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**Magnetic Force Microscopy study of layered
superconductors in vectorial magnetic fields**

Memoria presentada por

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Abstract

The thesis project is mainly focused on the study of the magnetic and structural properties in superconductors and thin flakes of layered materials. Particularly, a BCS prototype superconductor (β -Bi₂Pd), an iron based superconductor (Ca(Fe_{0.965}Co_{0.035})₂As₂), a cuprate superconductor (BiSr₂CaCu₂O₈ or Bi-2212) and graphene. To access the local magnetic properties of this materials we have used a low temperature magnetic force microscope (LT-MFM) capable to work between 2 and 300 K.

MFM is a widely used technique in the field of superconductivity, specially for vortex characterization and manipulation. It allows to map and manipulate not only the vortex lattice but also individual vortices, being an ideal tool to study their distribution and local pinning forces.

The vortex distribution in a superconductor at very low fields is still an open debate in the scientific community. For example, bitter decoration experiments performed in the single gap, low- κ superconductor, Nb, shows areas where flux expulsion coexists with regions showing a vortex lattice. Moreover, Scanning Hall Microscopy experiments have shown vortex chains and clusters in ZrB₁₂ ($0.8 > \kappa < 1.12$) at very low fields [1]. Both experiments were explained with the existence of an attractive term in the vortex-vortex interaction in superconductors with $\kappa < 1.5$. This regime is known as the *Intermediate Mixed State*. On the other hand, the existence of vortex free areas between cluster and stripes of vortices at very low fields was also reported in the multigap superconductor MgB₂ [2–4]. In this case, the authors propose that this behavior corresponds to a new state that they called *Type 1.5 superconductivity*, due to the existence of two different values of the Ginzburg-Landau parameter, κ , for the

two gaps of the compound. In addition, a recent theoretical work have also proposed that pinning may have an important role in the formation of the vortex patterns in MgB_2 [5]. Comparatively, $\beta\text{-Bi}_2\text{Pd}$ has a small, yet sizable, value of $\kappa \approx 6$. It has very weak pinning and is a single gap isotropic superconductor [6–8]. This allow us to characterize the vortex distribution at very low field in a material with only one gap and a moderate value of κ for first time. We have found vortex clusters and stripes as in the case of low- κ or multigap superconductors. But, in this case, we have found that they are associated with local changes in the value of the penetration depth of the superconductor.

In the fourth chapter of the thesis, we have focus in the local manipulation of superconducting vortices in the high-temperature cuprate superconductor Bi-2212. It has a two-dimensional layered (perovskite) structure and with superconductivity taking place in the copper oxide planes. When a magnetic field is applied tilted with respect to the c crystallographic axis, it decomposes in two perpendicular components, one parallel (Josephson vortices) and another perpendicular (Pancake vortices) to the CuO planes. In our work, we have achieved local manipulation of single PVs and we have experimentally determine the pinning force of isolated PVs and the attracting force between PVs and JVs.

$\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ is an iron based compound with extremely high sensitivity to pressure and strain. Due to the presence of Ca ions, small pressures result in dramatic changes in the ground state of the system. Moreover, the recent proposal of phase separation between superconducting and antiferromagnetic domains under the action of biaxial strain in this compound [9], opens a good opportunity to the local characterization of a system where magnetic and superconducting domains coexist, for first time. We have characterized the formation of alternating superconducting antiferromagnetic domains at low temperatures and related them with the separation of the material in two structural phases at higher temperatures.

Finally, in the last chapter of the thesis, we have focused in the exfoliation and deposition of layered superconductors and in the study of graphene/superconductor interphases. 2D superconductivity in thin films and crystals flakes has attracted the attention of many researchers in the last decade [10–18]. For example, superconduct-

ing crystals like BSCCO or TaS₂ have been successfully exfoliated down to a single layer and deposited in a substrate in the past [19–21]. In addition, a lot of work has been done trying to induce superconductivity in graphene in contact with a superconductor due to the proximity effect [10, 11, 22–26]. In our case, we have measured the magnetic profile of a Bi-2212 flake below the superconducting transition, developed an experimental procedure to localize graphene flakes deposited on top of a β -Bi₂Pd single crystal and demonstrated the possibility of deposit thin flakes of the β -Bi₂Pd superconductor on a substrate. We have also measured the vortex lattice at low temperatures of a β -Bi₂Pd single crystal with a graphene sheet deposited on top.

Resumen

Este proyecto de tesis está principalmente enfocado en el estudio de las propiedades magnéticas y estructurales en superconductores y capas delgadas de materiales laminares.

la microscopía de fuerzas magnéticas (MFM) es una técnica ampliamente utilizada en el campo de la superconductividad, especialmente para la caracterización y manipulación de vórtices. Esta técnica permite medir y manipular no solo la red de vórtices, si no, además, vórtices aislados. Ello la convierte en una herramienta ideal para el estudio de la distribución de los vortices y su fuerzas de pinning locales.

La distribución de los vórtices en un superconductor a campos muy bajos es aún un debate abierto en la comunidad científica. Por ejemplo, experimentos de decoración magnética llevados a cabo en el superconductor de un solo gap y pequeño κ , Nb, muestran áreas donde la expulsión de flujo coexiste con regiones donde está presente la red de vórtices. Además, experimentos de microscopía Hall, han mostrado cadenas y acumulaciones de vórtices en ZrB_{12} ($0.8 < \kappa < 1.12$) a campos muy bajos. Ambos experimentos fueron explicados mediante la existencia de un término atractivo en la interacción entre vórtices en superconductores con $\kappa < 1.5$. Este regimen es conocido como el *Estado Intemedio Mixto*. Por otro lado, la existencia de áreas libres de vórtices entre acumulaciones y cadenas de ellos a campos muy bajos ha sido tambien demostrada en el superconductor de varios gaps MgB_2 . En este caso, los autores propusieron que esto se debe a un nuevo estado conocido como *Superconductividad tipo 1.5*, debido a la existencia de dos valores diferentes del parámetro de Ginzburg-Landau, κ , para los dos gaps del compuesto. Además, un trabajo teórico reciente, ha propuesto que el pinning puede jugar un papel importante en la distribución de los

vórtices en MgB_2 . Comparativamente, el $\beta\text{-Bi}_2\text{Pd}$ tiene un gap pequeño con un valor de $\kappa \approx 6$. Tiene un pinning muy debil y es un superconductor con un solo gap isótropo. Esto nos ha permitido caracterizar la distribución de vórtices a campos muy bajos en un material con un solo gap y un valor moderado de κ por primera vez. Hemos encontrado acumulación y cadenas de vórtices como en el caso de los superconductores de bajo κ . Pero, en este caso, hemos encontrado que están asociados con cambios locales en el valor de la longitud de penetración en el superconductor.

En el cuarto capítulo de la tesis, nos hemos enfocado en la manipulación local de los vórtices superconductores en el superconductor de alta temperature Bi-2212 . Este material tiene una estructura consistente en láminas bidimensionales (tipo peroskita) con la superconductividad originándose en los planos de óxido de cobre. Cuando un campo magnético es aplicado inclinado con respecto al eje cristalográfico c , este se descompone en dos componentes, una paralela (vórtices de Josephson) y otra perpendicular (vortices pancake) a los planos de CuO . En nuestro trabajo, hemos conseguido manipular de manera local vortices pancake y hemos determinado de manera experimental la fuerza de pinning de vortices pancake aislados y la fuerza atractiva entre vórtices pancake y Josephson.

El $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ es un superconductor basado en hierro con una gran sensibilidad a la presión y la tensión. Debido a la presencia de los átomos de Ca , pequeñas presiones resultan en cambios dramáticos en el estado fundamental de este sistema. Además, con la reciente propuesta de una separación de fases sueprconductoras y antiferromagnética bajo el efecto de una tensión biaxial en este compuesto, ha surgido una gran oportunidad para la caracterización local de un sistema donde magnetismo y superconductiidad conviven por primera vez. Nosotros hemos caracterizado la formación de dominios alternativos superconductores y antiferromagnéticos a bajas temperaturas y los hemos relacionado con la separación del material en dos fases estructurales distintas a temperaturas mayores.

Finalmente, en el último capítulo de la tesis, nos hemos centrado en la exfoliación y transferencia de superconductores laminares y en el estudio de interfases grafeno/superconductor. La superconductividad 2D en láminas delgadas y en copos de critales ha atraído la atención de muchos investigadores en la última década. Por

ejemplo, cristales superconductores como el BSCCO o el TaS₂ han sido esfoliados con éxito hasta una monocapa y depositados en un sustrato en el pasado. Por otro lado, mucho trabajo se ha invertido tratando de inducir superconductividad en grafeno en contacto con un superconductor mediante el efecto proximidad. En nuestro caso, hemos medido el perfil magnético de un flake de Bi-2212 por debajo de la transición superconductora, desarrollado un procedimiento experimental para localizar flakes de grafeno depositados encima de β -Bi₂Pd y demostrado la posibilidad de depositar pequeños flakes del superconductor β -Bi₂Pd en un sustrato. Además, hemos medido la red de vórtices a bajas temperaturas de un monocristal de β -Bi₂Pd con una lámina de grafeno depositada encima.

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Introduction

1.1 Historical remarks

Superconductivity was first discovered by H. K. Onnes in 1911 [27] after he was able to liquefy He in his laboratory in 1908 opening a new branch in physics, the *low temperature physics*. Onnes expected a gradual approach of the resistivity to zero with decreasing the temperature, however he found that the resistance of Hg dropped to zero below 4.15 K. Onnes realized that he had found a new state of the matter characterized by zero resistivity, the *superconductivity*. One year later he also discovered that applying a strong magnetic field to superconducting Hg, the normal state was recovered. In the following years new superconducting materials were discovered: Pb, Nb etc. In 1933 Meissner and Ochsenfeld found that if a superconductor material is cooled down below its transition temperature, it expels any external magnetic field below a certain value called the *critical magnetic field*, H_C , [28]. This effect is called nowadays the *Meissner state*. Later, superconductors where the magnetic field can penetrate the

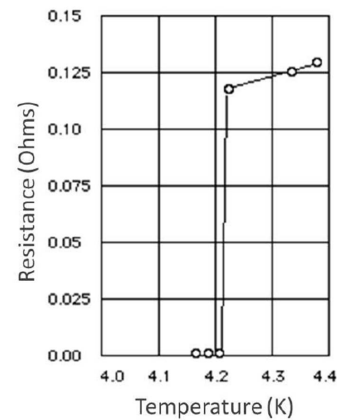


Figure 1.1: K. Onnes original measurement of the superconducting transition in Hg.

material without losing the zero resistivity were discovered and superconducting materials were split in two categories, type I and type II superconductors.

Type I superconductors present zero resistivity and perfect diamagnetism below T_C and H_C . Type II superconductors present zero resistivity below T_C and the the *upper magnetic critical field*, H_{C2} , but only perfect diamagnetism below the *lower magnetic critical field*, H_{C1} . Between H_{C1} and H_{C2} , the magnetic field penetrates the material in form of magnetic vortices that carry one single magnetic quantum flux, $\phi_0 = 2.067 \cdot 10^{-15}$ Wb. This regime is called the *mixed state*. For more details see references [29, 30].

1.2 Superconducting theories

These discoveries prompted the London brothers to propose the first phenomenological theory to explain the new phenomena in 1935 [31]. In 1950 a new superconducting theory was developed, the Ginzburg-Landau theory [32]. It describes the superconductivity in terms of an order parameter. Today the most advanced theory on superconductivity is the BCS theory [33], named after John Bardeen, Leon Cooper, and John Robert Schrieffer. The BCS theory is the first microscopic theory on superconductivity.

1.2.1 Ginzburg-Landau Theory

The thermodynamic superconducting properties can be described in terms of the Gibbs free energy G_S . Ginzburg and Landau assumed that close to the transition temperature the Gibbs free energy density can be expanded as function of a complex order parameter, $\psi = |\psi| e^{i\theta}$ [34] as:

$$G_S = G_N + a|\psi|^2 + \frac{b}{2}|\psi|^4 + \frac{1}{2m^*} \left| (i\hbar\nabla - e^*\vec{A})\psi \right|^2 \quad (1.1)$$

$m^* = 2m_e$ and $e^* = 2e$ are the superelectron mass and charge (m_e and e , are

the electron mass and charge), \vec{A} the vector potential and a and b parameters only dependent of the temperature with values $a \approx a_0[T/T_C - 1]$ and $b \approx b_0$ near T_C . The square of the order parameter is the super electron density, n_s . The order parameter ψ is zero above T_C and increases as the temperature decreases below T_C . Taking the derivative of equation 1.1 with respect to the order parameter they found what is now called the *first G-L equation*:

$$\frac{1}{2m^*}(i\hbar^2\nabla^2\psi - 2i\hbar e^*\vec{A}\cdot\nabla\psi - e^{*2}\vec{A}^2\psi) - a\psi - b|\psi|^2\psi = 0 \quad (1.2)$$

The free energy is also a minimum with respect to the vector potential \vec{A} . Taking the derivative of G_S with respect to \vec{A} , we obtain the *second G-L equation*:

$$\nabla \times (\nabla \times \vec{A}) + \frac{i\hbar e^*}{2m^*}(\psi^*\nabla\psi - \psi\nabla\psi^*) + \frac{e^{*2}}{m^*}\vec{A}|\psi|^2 = 0 \quad (1.3)$$

The G-L equations can be used to calculate the two principal length scales in a superconductor as we will introduce in the following.

1.2.1.1 Coherence length

Lets now study the following case: a semiinfinite superconductor from $x = 0$ to $x = \infty$ and a normal metal from $x = -\infty$ to $x=0$. Setting $\vec{A} = 0$ in the first G-L equation we obtain:

$$-\frac{\hbar^2}{2m^*}\nabla^2\psi + a\psi + b|\psi|^2\psi = 0 \quad (1.4)$$

Since the phase, θ of the order parameter is arbitrary, we can take ψ real ($\theta = 0$) and therefore, $\psi = \psi(x)$. Now, we can simplify the equation 1.4 to the one dimensional case:

$$-\frac{\hbar^2}{2m^*}\frac{d\psi^2}{dx^2} + a\psi + b|\psi|^2\psi = 0 \quad (1.5)$$

which has the solution:

$$\psi = \psi_{\infty} \tanh \frac{x}{\sqrt{2}\xi} \quad (1.6)$$

where ξ is a characteristic length of ψ . ξ is called the *coherence length* and is one of the two main parameters of the G-L theory. The order parameter ψ is zero inside the normal material and increases until ψ_{∞} over an intermediate width of ξ in the superconducting material.

1.2.1.2 Penetration depth

Now, we will consider the same semiinfinite geometry than in the previous section but with an homogeneous magnetic field in the Z direction, which has a potential vector $\vec{A} = A_y(x)$.

Substituting in the second G-L equation, we find:

$$\frac{d^2 A_y(x)}{dx^2} = \frac{\mu_0 e^{*2} |\psi|^2}{m^*} A_y(x) \quad (1.7)$$

and the solution for the vector potential inside the superconductor is:

$$A_y(x) = A_0 e^{(-x/\lambda)} \quad (1.8)$$

And therefore:

$$B_z(x) = B_0 e^{(-x/\lambda)} \quad (1.9)$$

where A_0 and B_0 are constants and λ is the *penetration depth*, the second characteristic length of the G-L theory. It represents the distance in which an external magnetic field decreases inside the superconductor a factor e^{-1} .

1.2.1.3 Type I and type II superconductors

Using the two characteristic lengths of the G-L theory, we can define the dimensionless quantity:

$$\kappa = \frac{\lambda}{\xi} \quad (1.10)$$

which is called the *G-L parameter* and is the most important parameter of the theory. It helps to separate the superconductor material in two classes, type I and type II superconductors. Values of $\kappa < 1/\sqrt{2}$ and $\kappa > 1/\sqrt{2}$, separates the G-L equations in two different branches of solutions. For $\kappa < 1/\sqrt{2}$ the energy difference between a normal and a superconducting domain is positive and for $\kappa > 1/\sqrt{2}$ is negative which means that for the superconductor becomes favorable the formation of many small superconducting and normal domains [29].

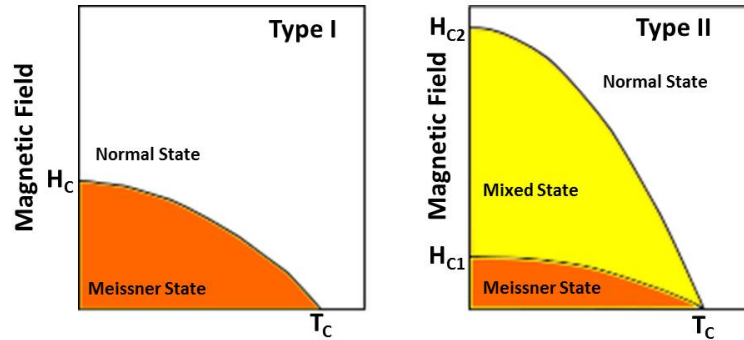


Figure 1.2: Phase diagram for type I (left) and type II (right) superconductors. In orange, the region presenting Meissner state. In white, the normal region. In yellow, the mixed state region.

Solving the G-L equations for $\kappa < 1/\sqrt{2}$ the B-T phase diagram for type I superconductors is found. In this phase diagram, there are only two regions, normal and Meissner state, separated by the critical field, with a dependence of the temperature following:

$$B_C(T) = B_C(0) \left[1 - \left(\frac{T}{T_C} \right)^2 \right] \quad (1.11)$$

For $\kappa > 1/\sqrt{2}$ a B-T phase diagram with three regions is found. The phase diagram is separated in normal state, Meissner state and mixed state. The two first regions are analogous to the regions in type I superconductors and the mixed state is a region where the magnetic flux is allowed to enter into the superconductor material in form of superconducting vortices that carry a flux ϕ_0 . Vortices represents singularities where the order parameter is suppressed and the material is in the normal state, allowing the formation of normal-superconducting interfaces. The three regions are separated by two critical fields, following the equations:

$$B_{C1}(T) = B_{C1}(0) \left[1 - \left(\frac{T}{T_C} \right)^2 \right] \quad (1.12)$$

$$B_{C2}(T) = B_{C2}(0) \left[1 - \left(\frac{T}{T_C} \right)^2 \right] \quad (1.13)$$

Where:

$$B_{C1}(0) = \frac{\phi_0}{4\pi\lambda^2} \ln(\kappa) \quad (1.14)$$

$$B_{C2}(0) = \frac{\phi_0}{2\pi\xi^2} \quad (1.15)$$

Both phase diagrams are schematized in the figure 1.2.

1.2.1.4 Vortex lattice

As it has been mentioned in the previous section, in type II superconductors, above a certain value, the magnetic field is not fully expelled from the superconducting material. It penetrates in form of magnetic vortices where the superconductivity is broken. As we have introduced using the G-L theory, in such an interface (normal-superconductor) the coherence length and the penetration length determines how the superconducting electron density of states and the magnetic field behaves.

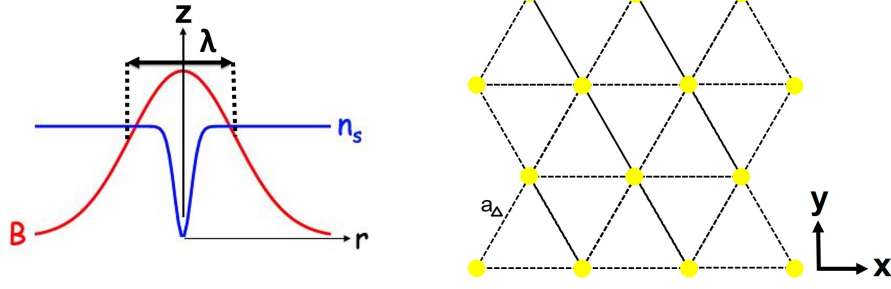


Figure 1.3: In the left panel, an scheme of the superconducting density of states (blue) and the magnetic field (red) inside a superconducting vortex. The center of the vortex is located at the center of coordinates in the scheme. In the right panel, an schematic representation of the Abrikosov vortex lattice of SC vortices with a lattice parameter a_{Δ} . SC vortices are represented as yellow circles. The lattice is schematized with dashed black lines.

Thus, we can separate vortices in two spatial scales, the first is the superconducting density of states scale and is determined by ξ . The vortex consist in a core region of 2ξ width where n_S is zero at the center and increases until it reaches a finite value outside this region. From the magnetic point of view, the highest field is in the core. The core is surrounded by a region of larger radius λ within which magnetic flux and screening currents flowing around the core are present together. The supercurrent density J_s of these shielding currents decays with distance from the core in an approximately exponential manner [30]. Both spatial dependence of the vortex structures are shown in figure 1.3.

Vortices have a repulsion interaction between them, for this reason they arrange in a hexagonal lattice (it is the configuration that minimizes such a system as it was proved by Kleiner, Roth y Autler [35]) called the Abrikosov lattice after Aleksei Abrikosov who first propose the existence of superconducting vortices in type II superconductors [36]. The parameter of the vortex lattice is:

$$a_{\Delta} = 1.075\sqrt{\phi_0/B} \quad (1.16)$$

which is only dependent in the value of the magnetic field. An schematic repre-

sensation of the vortex lattice is shown in figure 1.3.

1.2.2 BCS theory

In 1956, L.N Cooper demonstrated that the normal ground state of an electron gas, is unstable with respect the formation of bound electron pairs [37]. Cooper, developed his theory following an original idea of Frohlich [38]. Frohlich argument that an electron moving across a crystal lattice, due to its negative charge will attract the positive ions in the lattice. In the surroundings of the electron, there will be an accumulation of positive charge, changing locally the density of charge in the lattice and exciting a phonon. If a second electron is near this perturbation, it will be attracted by it absorbing a phonon (figure 1.4). Cooper considered a pair of electrons near the Fermi level whose attraction due to the phonon interaction was greater that the Coulomb repulsion, creating a bound state between both electrons. The attraction is maximum when the momentum of the electrons is equal and has opposite sign ($\vec{k}_1 = -\vec{k}_2$), the resulting cooper pair has momentum and spin equals to zero.

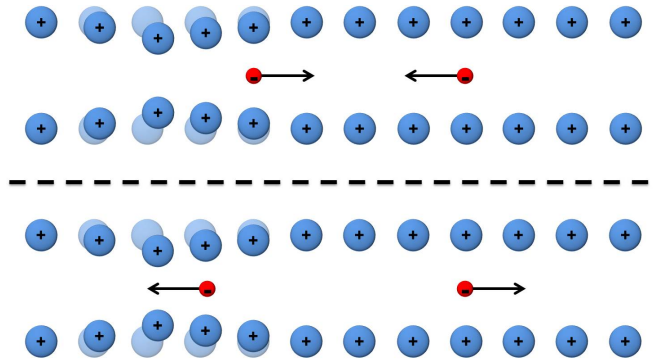


Figure 1.4: Scheme of the phonon mediated pairing of Cooper pairs. The atomic cores are represented with blue circles and the electrons with red circles. The direction of the movement of the electrons is schematized by a black arrow. The movement of the atomic lattice is represented by transparent circles. In the upper panel, an electron coming from the left, slightly distorts the atomic lattice. In the lower panel, another electron coming from the right is attracted by the accumulation of positive charge at the distortion.

One year later, J. Bardeen, L. N. Cooper y J. R. Schrieffer presented the basis of their new microscopic theory of superconductivity [33]. A theory that nowadays is

known as the BCS theory. They extrapolated Cooper's result to a system with many electrons. As Cooper pairs have zero spin, they behave as bosons and therefore they can condensate in a Bose-Einstein condensate. This state is described in the BCS theory with a macroscopic wave function that keeps the phase coherence a distance equal to the coherence length ξ .

1.2.2.1 Superconducting gap

Forming Cooper pairs, decreases the energy of the system a quantity equal to the energy of the bonding between electrons in the pair, 2Λ . In the ground state, copper pairs are condensed in a state with an energy Λ below the Fermi level and the first excited state has an energy Λ above the Fermi level. Λ is known as the superconducting gap.

1.3 Intermediate and Intermediate Mixed States

As it was presented before, below H_C (Type I SC) or H_{C1} (Type II SC) no magnetic field penetration is expected. Below this critical fields, both types of superconductors should behave as perfect diamagnets. But, some works have reported flux penetration below H_C in type I SCs [39, 40] and below H_{C1} in type II SCs [2–4, 40–43]. This unexpected behaviour can be explained as a *intermediate state* (IS) in type I SCs and a *intermediate mixed state* (IMS) in type II SCs.

1.3.1 Intermediate State

Lets consider the case of a type I superconducting sphere (demagnetization factor, $N=1/3$) in the presence of an external magnetic field in the Z direction. Below T_C , the magnetic field at the surface of the sphere is:

$$B_{surface} = \frac{3}{2}B_a \sin\Theta \quad (1.17)$$

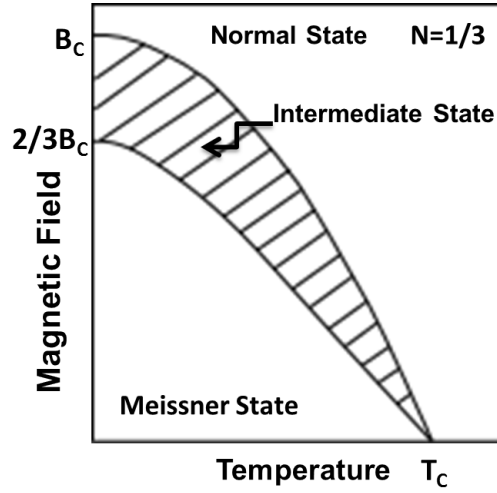


Figure 1.5: B-T phase diagram of a type I superconducting sphere. The curve $B=2/3B_C(T)$ separates the Meissner from the IS. The region where the IS takes places is dashed.

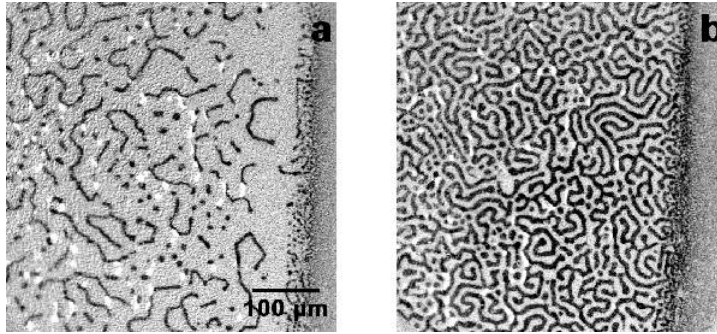


Figure 1.6: Typical IS patterns in an In sample with thickness $d=10\mu\text{ m}$ for increasing values of the applied magnetic field. Images a and b, correspond to $h=0.105$ and $h=0.345$, respectively ($h=H/H_C$) at $T=1.85\text{ K}$. SC domains are represented in black and have circular or lamellar shapes. The edge of the sample is along the right edge of the image. Adapted from [44]

where B_a is the external magnetic field and Θ the polar angle in spherical coordinates. If the external magnetic field is lower than $2/3B_C$, the surface field will be lower than B_C in all the surface, and the sphere will remain in the superconducting state. But, if the external magnetic field is greater than $2/3B_C$, from equation 1.17, there will be a range of angles where the surface field will exceed B_C and the sphere can not remain in the perfect superconducting state. In the range of external magnetic fields:

$$\frac{2}{3}B_C < B_a < B_C \quad (1.18)$$

The surface must decompose into superconducting and normal regions that keep the internal field below the critical value H_C . This state is known as the *intermediate state* (IS). The trigger of this state is the inhomogeneous distribution of the magnetic field on the surface due to the demagnetization factor of the samples. An scheme of the phase diagram for a superconducting sphere is shown in figure 1.5, where the dashed area represent the region where the IS takes place. The IS was observed in various type I superconductors in form of tongues or alternative domains of Meissner and normal states [39, 40, 44] (figure 1.6).

1.3.2 Intermediate Mixed State

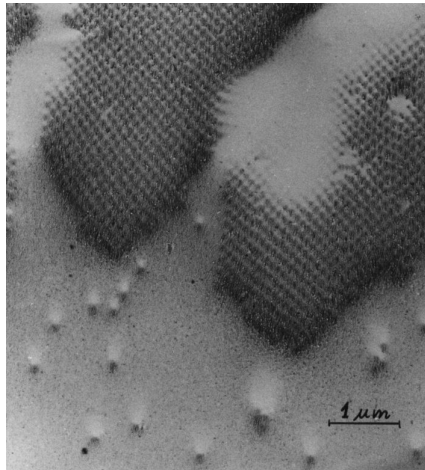


Figure 1.7: Magnetic decoration of a square disk $5 \times 5 \times 1 \text{ mm}^3$ of high purity polycrystalline Nb at 1.2 K and 1100 Oe, showing domains of Meissner and mixed states. Magnetic flux penetrates from the edges in form of fingers which are composed of vortex lattice. Adapted from [43].

Following the same arguments than for type I SC, if a magnetic field is applied to a type II superconductor, at certain fields below B_{C1} , the SC will decompose in domains in the Meissner state and domains in the mixed state, depending on its demagnetization factor [40]. This regime is called the *intermediate mixed state* (IMS). Experimentally it was found that the intervortex distance in the IMS domains

corresponds to the expected value corresponding to the inductance B_{C1} in equation 1.16. It was also found that the area occupied by the domains with zero induction decreases linearly with the magnetic field, to reach B_{C1} when entering the mixed phase [40]. An example of the IMS in a type II superconductor is presented in figure 1.7 where the magnetic flux penetrates into the Nb forming domains in the Meissner states and domains with a regular vortex lattice with $a_{\Delta} = 1.075\sqrt{\phi_0/B_{C1}}$, independent of the magnetic field.

1.4 Anisotropic Superconductors

In anisotropic superconductors, the electronic properties have a huge dependence on the direction of the space and new consideration have to be taken into account in order to understand their behaviour. For example, in cuprates, cooper pairs and vortices are confined into 2D copper oxide planes [45–51]. The penetration depth and the coherence length have to be separated in two components, one associated to supercurrents parallel (ξ_{\parallel} and λ_{\parallel}) and perpendicular (ξ_{\perp} and λ_{\perp}) to the superconducting planes [45, 46, 48]. Then, we can define the anisotropic factor, $\gamma = \xi_{\parallel}/\xi_{\perp} = \lambda_{\perp}/\lambda_{\parallel}$.

In highly anisotropic layered superconductors like BSCCO, when a magnetic field is applied perpendicular to the superconducting planes, it penetrates the material in form of stacks of 2D vortices in the CuO planes, called pancake vortices (PVs). If the magnetic field is applied parallel to the CuO planes, it penetrates the superconductor parallel to the CuO planes in form of Josephson vortices (JVs) [49][52].

1.4.1 Pancake vortices

In BSCCO and other highly anisotropic superconductors, the CuO planes are separated in the c-axis direction a distance $s > \xi_{\perp}$ and therefore they act as Josephson junctions [47–51]. A vortex perpendicular to these layers, which conventionally would be considered a uniform cylinder of confined flux, is interpreted in high anisotropic superconductors as a stacking of 2D pancake shaped vortices (PVs), one PV per layer with surrounding currents confined to the layer [45, 53–56]. PVs are so weakly cou-

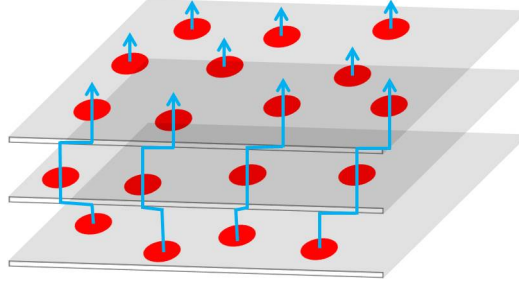


Figure 1.8: Stack of 2D pancake vortices in a layered superconductor. Red circles represents the 2D PVs while blue lines are a guide to the eye to connect the PVs at different layers (grey planes).

pled that thermal agitation can decouple the stack of PVs [57]. An scheme of PVs in different layers of a highly anisotropic superconductor is presented in figure 1.8.

1.4.2 Josephson vortices

In the case of an applied magnetic field parallel to the superconducting planes, the field penetrates in highly anisotropic superconductors in form of Josephson vortices (JVs) [51]. JVs do not have normal cores, but have rather wide non linear cores located between two superconducting layers [49, 51]. The structure of the core is similar to the structure of the phase drop across a flux line in two-dimensional Josephson junctions, where the phase changes 2π between the two layers over a distance of Λ_J [51]. For 3D superconductors, this length is given by $\Lambda_J = \gamma s$, and we can think of a central region of γs wide and s high as the core of the JV [51] (figure 1.9). Beyond this core, the screening of the z -axis currents is weaker than by in-plane currents, and the flux line is stretched into an ellipsoidal shape with a large width (λ_{\perp}) along the layers. An scheme of a JV is shown in figure 1.9.

Under an applied magnetic field parallel to the CuO planes, in the Y direction, they arrange in a strongly stretched triangular lattice along the direction of the layers with lattice parameters [51]:

$$a_z = \sqrt{2\phi_0 / \sqrt{3}\gamma B_x} \quad (1.19)$$

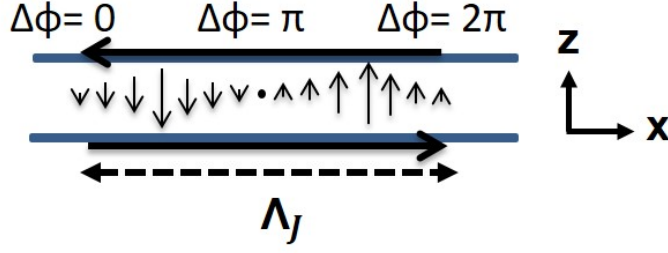


Figure 1.9: Scheme of a JV in a layered superconductor with the in plane magnetic field applied in the Y direction. Horizontal blue lines represents the SC planes and the black arrows the Josephson currents (vertical) and the supercurrents (horizontal) resulting from the JV. The phase difference between the CuO planes is summarized in the upper part of the image.

$$a_x = \sqrt{\sqrt{3}\gamma\phi_0/2B_x} \quad (1.20)$$

1.4.3 Crossing lattice

A huge variety of vortex configurations have been proposed when applying magnetic field tilted with respect the c axis in highly anisotropic superconductors [47, 49, 50, 52, 58]. We will focus in the crossing lattices of PVs and JVs. In this configuration, the JVs interact with the stacks of PVs splitting them in two branches giving a zig-zag like structure perpendicular to the CuO planes [47, 49, 50, 52].

A JV in the Y direction between two superconductor layers carries a current with opposite sign in the two layers ($\pm J$). The current, interacts with the stack of PVs in the Z direction with a Lorenz force $+F_y$ and $-F_y$ in the two different superconductor layers. As a result the PV stack is displaced a distance $+a$ and $-a$ in the two planes in the direction of the JV, causing a zig-zag like structure in the PV stack [47, 49, 50, 52], as is represented in figure 1.10 e and f. The amplitude of the distortion has been extensively studied by [49], finding that the maximum pancake displacement at the JV core position is:

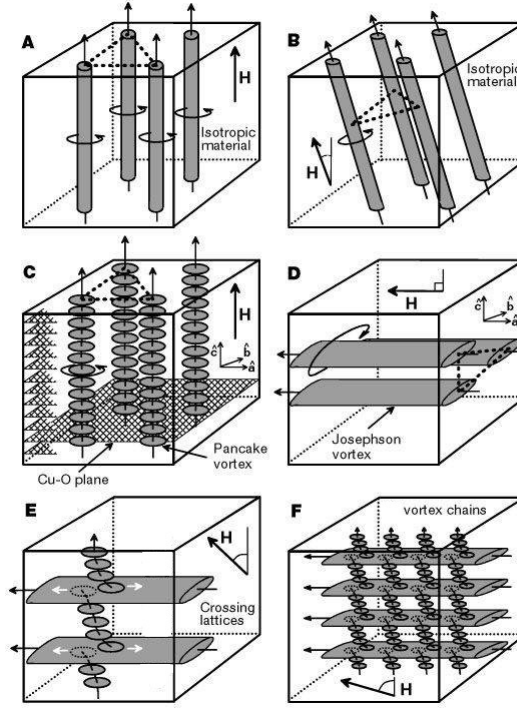


Figure 1.10: Vortices in isotropic and highly anisotropic superconductors. In a and b, the vortex lattice in an isotropic superconductor where the repulsion between vortices leads to the formation of hexagonal lattice. Curved arrows indicate circulating supercurrents around the vortex core. In c, hexagonal ordering of the vortex lattice in layered superconductors with the magnetic field applied along the c axis. In this case, vortices are formed of vertical stacks of 2D PVs situated in the CuO planes. In d, with the magnetic field parallel to the layers, crystalline anisotropy leads to the formation of elliptical JVs. In e, tilted vortices spontaneously decompose into coexisting orthogonal PVs and JVs. Where a PV stack intersects a JV stack, small PV displacements (indicated by white arrows) driven by the JV supercurrents lead to an attractive interaction. In f, the vortex chain state when all PVs stacks become trapped on vertical stacks of JVs. Adapted from [59].

$$a \approx \frac{2.2\lambda_{\parallel}}{\gamma \log(2\gamma s/\lambda_{\parallel})} \quad (1.21)$$

The distorted PV stack crossing a JV have less energy compared with other stacks, which makes favourable to add an extra stack on top of the JV and form PV rows along the JV [47] separated a distance[60]:

$$d \approx 2\lambda_{\parallel} \log \frac{B_{\parallel} \gamma^2 s^2}{\phi_0 \lambda_{\parallel}} \quad (1.22)$$

The existence of PVs rows decorating JVs have been confirmed in previous experimental works [61–68].

The crossing lattice of PVs and JVs causes a rearrangement of the phase distribution on the CuO planes and therefore in the JV structure. In a isolated JV in the Y direction, the phase difference, $\Delta\phi = \phi_1 - \phi_0$ (ϕ_1 and ϕ_0 are the phases at both CuO planes), between the top and bottom CuO planes changes by 2π over a distance Λ_J in the X direction. The phase difference is 0 and 2π at the edges and π at the centre of the JV (figure 1.11 b). Adding one PV in each layer, separated by a in-plane distance $2a$ in the Y direction, causes a change in the phase in each CuO plane. The phase changes by π between the extremes of the line that crosses a PV parallel to the JV. The phase changes by π in both layers at different positions, creating a narrow region, $2a$ width, where the phase difference between CuO layers is 2π instead of π in the centre of the JV [49] (figure 1.11 c). This phase distribution is very effective to pin the PV and JV lattice.

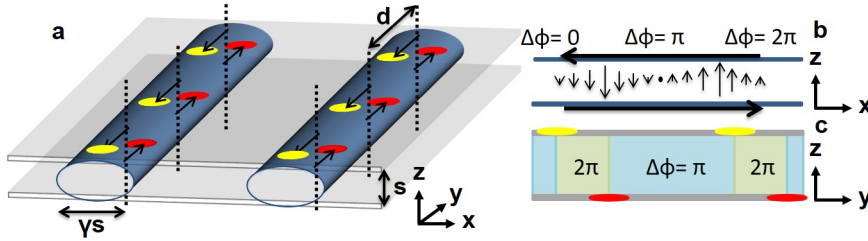


Figure 1.11: In a, we schematically describe PVs and JVs crossing lattice. The ellipse signals a Josephson vortex and the circles pancake vortices pinned to it. The relevant approximate length scales, such as JVs size and distance between pancake vortices is also shown. In b, we show a two dimensional scheme of the Josephson currents and phase difference in a JV. In c, blue areas represents areas where $\Delta\phi = \pi$ and green areas where $\Delta\phi = 2\pi$ as a consequence of the PVs displacement.

1.5 Iron Based Superconductors

Iron based superconductors (FeBSC) were first discovered by Kamihara et al. in 2006 [69]. They found that LaFePO transits to a superconducting state below 4 K. Two years later, they found superconductivity in LaFeAsO_{1-x}F_x with a T_C of 26 K [70]. Fe is a well known magnetic material and magnetism was thought to damage superconductivity. Actually, magnetism and superconductivity are considered as competing states. For this reasons, finding superconductor with Fe was a big surprise. Moreover, as in the case of cuprates superconductors, the BCS phonon-mediated coupling was not able to explain the formation of cooper pairs in this superconductors. Before 2008, the term *high-temperature superconductivity* (HTS) was reserved for the cuprates. Now the term HTS equally applies to both cuprates and FeBSC.

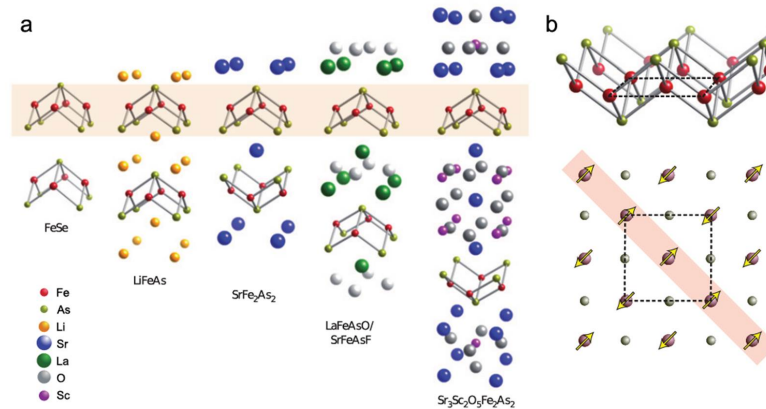


Figure 1.12: In a, crystal structure of different families of iron pnictides. Fe-As planes are highlighted as common features in all structures. In b, the FeAs plane from a frontal (top) and upper (bottom) point of view. Spins are aligned ferro and antiferromagnetic alternately in a structure called stripe like antiferromagnetic order. Adapted from [71].

Among the FeBSCs, the 122 family has focused a lot of attention in the last years. Specially the compounds derived from BaFe₂As₂ and CaFe₂As₂. Nematicity for instance, was first reported in a STM study of Co doped CaFe₂As₂ [72]. The electronic nature of this state was showed performing resistivity measurements on detwinned single crystals of Co doped BaFe₂As₂ by [73]. Recently, strain induced phase separation between superconducting tetragonal domains and non-superconducting orthogonal

domains was proposed by [9] in Co doped CaFe_2As_2 .

FeBSC are also promising compounds to the study of superconductivity in the 2D limit. In FeBSC, superconductivity has its origin in the 2D Fe-As layers, similar to the CuO planes in the cuprates. In fact, a recent work has obtained signs of 2D superconductivity in the novel FeBSC $\text{Ca}_{1-x}\text{La}_x\text{Fe}_{1-y}\text{Ni}_y\text{As}_2$ [74]. This compound presents an extra As-As chain spacer-layer that increases the FeAs layer interdistance up to $s = 10.34 \text{ \AA}$, and it is expected to be strongly anisotropic. The authors in [74], argue that they have obtained crystals with $\gamma \approx 30 \xi_c(0)$ smaller than the FeAs layer interdistance, s , leading to a 2D superconducting behaviour at accessible reduced temperatures.

1.5.1 Phase diagram

FeBSC have 2D lattices of 3d transition metal ions as the building block, sitting in a quasi-ionic framework composed of rare earth, oxygen, alkali or alkaline earth blocking layers. They present similar phase diagrams, with a magnetic ordered phase in the parent compound and a superconducting dome developing with doping. They also present orthorhombic transition at small doping.

Some compounds, for instance, LaFeAsO , shows first order transition between magnetic and superconducting phases and in other compounds like the 122 family, both states coexist for certain doping levels. FeBSC magnetic phases are metallic with linear dependence of the resistivity with the temperature. They also shown an structural phase transition which is often coupled with the magnetic transition. Above them, a nematic transition has been reported in some compounds. Superconductivity emerges as a dome at finite doping levels with the optimal doping level located where the magnetic transition extrapolates to zero temperature. For some materials there is a region where magnetism and superconductivity coexist. An schematic representation of the generic phase diagram of FeBSC is presented in figure 1.13.

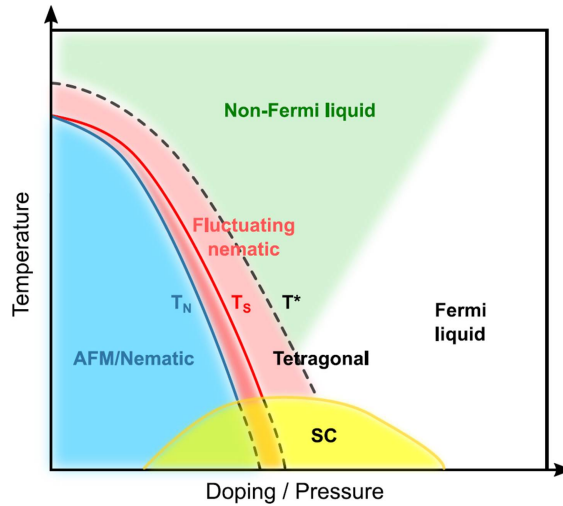


Figure 1.13: Generic temperature versus doping/pressure phase diagram for the FeBSC. The parent compound usually presents a structural/magnetic transition that reduces its temperature with increasing doping/pressure. The structural and magnetic transitions are coupled or separated depending on the compound. Above the structural transition and usually coupled to it and to the magnetic one there is an electronic nematic phase. Superconductivity emerges in a dome-shape with finite doping/pressure with the optimal doping usually coinciding with the extrapolation of the magnetic phase to zero temperature. Adapted from [75].

1.5.1.1 Electronic structure

The Fermi Surface (FS) of FeBSC is dominated by the d_{xy} , d_{yz} and d_{xz} orbitals of Fe and the out of plane orbital of the As, with which Fe is in tetrahedral coordination in a 2D layer (figure 1.12).

The electronic band structure has been calculated using the local density approximation [78], showing that the electronic properties are dominated by five Fe d states at the Fermi energy, with a FS consisting of at least four quasi-2D electron and hole cylinders. These consist of two hole pockets centred at the Brillouin zone (BZ) centre and two electron pockets centred at $(0, \pm\pi)$ and $(\pm\pi, 0)$ in the tetragonal unit cell (figure 1.14 c). Two non-equivalent As positions result in the folding of the BZ to include two Fe atoms per unit cell and to put the electron pockets at $(\pm\pi, \pm\pi)$ as shown in figure 1.14 d. A fifth hole band is also proposed to sit at $(0, \pm\pi)$ in the folded BZ, and its presence may be very sensitive to structural details [79].

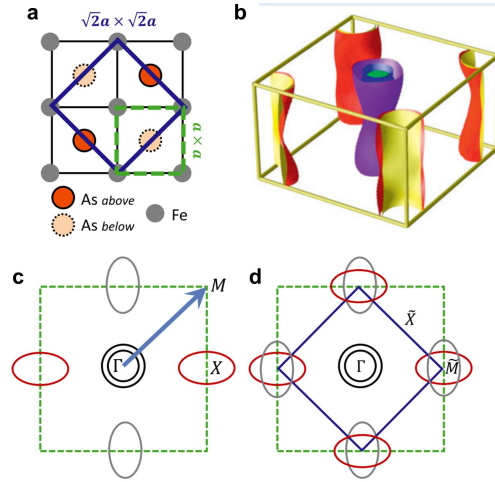


Figure 1.14: In a, FeAs lattice indicating As above and below the Fe plane. Dashed green and solid blue squares indicate 1- and 2-Fe unit cells, respectively. In b, FSs of BaFe_2As_2 with 10% substitution of Co, calculated using DFT using experimental atomic positions and drawn using the folded BZ representation with two Fe per unit cell. In c, schematic 2D Fermi surface in the 1-Fe BZ whose boundaries are indicated by a green dashed square. The arrow indicates folding wave vector QF . In d, Fermi sheets in the folded BZ whose boundaries are now shown by a solid blue square. Adapted from [76] and [77].

1.5.1.2 Magnetism

The nature of magnetism in the FeBC parent compounds is a hotly debated topic, the electronic structure suggests that the same magnetic interactions that drive the antiferromagnetic (AFM) ordering also produce the pairing interaction for superconductivity [76]. As predicted before experiments [80], AFM order in all FeAs-based superconducting systems is found to have a wave vector directed along (π, π) in the tetragonal unit cell with a real-space spin arrangement consisting of AFM stripes along one direction of the Fe sublattice and ferromagnetic stripes along the other (figure 1.12).

It was predicted by DFT calculations [81] and confirmed by experiments [82] that the magnetic ground state of FeTe has a double-stripe-type antiferromagnetic order in which the magnetic moments are aligned ferromagnetically along a diagonal direction and antiferromagnetically along the other diagonal direction of the Fe square lattice,

as shown schematically in figure 1.15 a. Meanwhile, DFT calculations predict that the ground state of FeSe has the single-stripe-type antiferromagnetic order, similar to those in LaFeAsO and BaFe₂As₂, as shown in figure 1.15 b.

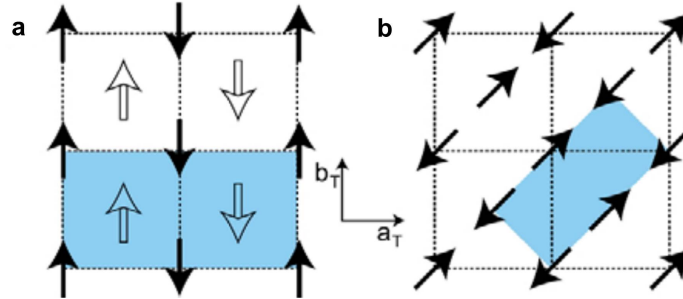


Figure 1.15: In a, double-stripe-type antiferromagnetic order in FeTe. The solid and hollow arrows represent two sublattices of spins. In b, single-stripe-type antiferromagnetic order in BaFe₂As₂. The shaded area indicates the magnetic unit cell. Adapted from [82]

The energetic stability of $(\pi, 0)$ antiferromagnetic ordering over (π, π) ordering in FeTe has been studied by [81]. They found that it can be described by the nearest, second nearest, and third nearest neighbor exchange parameters, J_1 , J_2 , and J_3 , respectively, with the condition $J_3 > J_2/2$. Authors in [83] found that Te height from the Fe plane is a key factor that determines antiferromagnetic ordering patterns in FeTe, so that the magnetic ordering changes from the $(\pi, 0)$ with the optimized Te height to the (π, π) patterns when Te height is lowered.

1.5.1.3 Superconducting gap

The symmetry of the superconducting gap function $\Lambda(\mathbf{k})$ has turned out to be a subject of debate in FeBSC. Figure 1.16 schematically presents various possible scenarios. The conventional s-wave state (a) has a gap with the same sign everywhere on the FS. The simplest scenario for FeBSC is the s₊- state (b) in which the gaps on hole and electron FSs are treated as constants and only differ in sign.

Theorists realized early on, however, that because of the multiorbital nature of FeBSC, an s₊- gap function on each pocket necessarily has an angular variation that may be substantial. Due to this angular variation, it is possible that four nodes

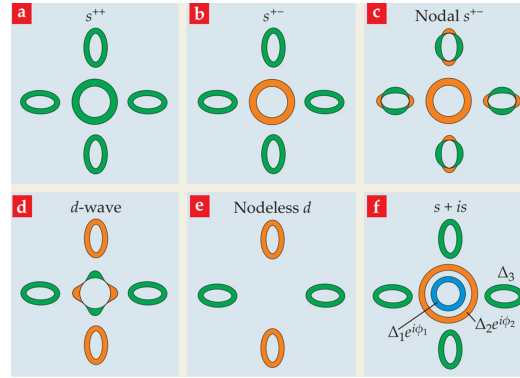


Figure 1.16: Schematic representation of the different scenarios proposed for pairing symmetries in FeBCS, colors represent the phase of the order parameter at each pocket. Adapted from [79].

develop on each FS (c). Such nodes have been called accidental, since their position is not set by symmetry. In contrast, a d-wave gap (d), by symmetry, must have its nodes along certain directions in reciprocal space. But if there is no central hole pocket, a d-wave state need not have nodes (e). The presence or absence of the nodes is highly relevant, as it completely changes the low-temperature behavior of a system compared with a conventional s-wave superconductor.

An even more subtle issue is the actual structure of the gap function phase in a generalized s+- state. We considered the case when the phase changes by π between hole and electron pockets, but in multiband systems other cases are possible, for example, a sign change, as in s+-, but now between different hole pockets, or phase differences which are not integer multiples of π (f). In the second case, superconducting order breaks time-reversal symmetry and is therefore dubbed s + is.

1.6 Motivation

The vortex distribution has been studied in a huge amount of superconducting system like type I, BCS prototype type II or High T_C superconductors. The knowledge of their interaction and distribution had a remarkably advance in the last decades. But, there are still open questions on this matter. For example, the study of the vortex distribution is usually made on the mixed state of type II SCs at magnetic fields well

above H_{C1} . Some works have advanced in the understanding of the IMS in type II SCs, but the mechanism of formation of vortex patterns below H_{C1} is still an open debate [2–4]. In particular, the possible mechanisms of the vortex distributions at very low fields and the current controversies on this matter are discussed extensively in [43]. Moreover, the majority of the previous works have been focused in the “passive” characterization of the vortex lattice but not in its local manipulation with scanning techniques. Some recent works have successfully manipulated Abrikosov vortices in 3D superconductors [84–87] but the manipulation of 2D pancake vortices and Josephson vortices in highly anisotropic systems have not been achieved yet. In particular, the force exerted on a PV by a JV has not been measured yet. On the other hand, the coexistence between superconductivity and magnetism have attracted a lot of attention in the last decade. Several theoretical and experimental attempts to understand the interplay between both states have been done in the last years. But, the play of the magnetism in the superconducting state and the pairing mechanism of the copper pairs in this systems remains unclear [71, 76, 80, 88–90]. In addition, the local characterization of a system where magnetic and superconducting domains coexist has not been achieved yet. The recent proposal of phase separation between superconducting and antiferromagnetic domains under the action of biaxial strain in CaFe_2As_2 Co doped, opens a good opportunity to the local characterization of this coexistence [9].

From an experimental point of view, answer those question needs a scanning probe technique capable to measure the topographic and magnetic profiles in areas of several tens of microns at low temperatures in a short period of time and under tilted magnetic fields. In order to manipulate the superconducting vortices in a controlled way, the scanning technique also has to be able to interact with them when it is necessary and do not perturb them when it is not desired. For these reasons, during the thesis, a set-up with a magnetic force microscope of low temperatures working in combination with a homemade three axis superconducting magnetic coil was employed as the main technique. The magnetic force microscopy is the only technique that allow to measure simultaneously the topography and the local magnetic profile of samples. This property is of vital importance to determine the possible effect of pinning at topographic defects on the vortex distributions, or to correlate topography

and magnetic profiles in samples showing magnetostructural transitions for example. In addition MFM has proved to be an effective tool to local manipulation of magnetic structures.

Our set-up have allowed us to characterize areas up to $20 \times 20 \mu m^2$ at low temperatures in a few minutes with tilted applied magnetic fields and interact with magnetic structures on the samples in a controlled way. Three system where selected in the thesis due to their specific properties to try bring some light in the topics we have presented in the previous paragraphs, β -Bi₂Pd, Bi₂Sr₂CaCu₂O₈ and Ca(Fe_{0.965}Co_{0.35})₂As₂.

Experimental methods

In this thesis, I have used magnetic force microscopy (MFM) at low temperatures to investigate the local properties of several superconductors. MFM allow to measure the magnetic field distribution at low temperatures in big areas ($20 \times 20 \mu\text{m}^2$ at 2 K) in rough or nanostructured samples where the differences in high are too big to another techniques like scanning tunnelling microscopy (STM). MFM also allow to single vortex manipulation [91]. For this reasons, MFM has become one of the most interesting techniques to study the local magnetism in different systems.

I have separated this chapter in three sections. In the first one, I will discuss our experimental set-up. Then, in the second section I will present a detailed characterization of the MFM probes at low temperatures. Finally in the third section, I will introduce the *solution growth method* that we have used to grow several crystals during the thesis.

2.1 Set-up

A Low Temperature Atomic Force Microscope (LT-AFM) from Nanomagnetics Instruments was employed during the thesis. It was used to characterize the superconducting vortex lattice and magnetic domains of several samples using the MFM mode. The microscope was used in combination with a commercial cryostat, a homedesigned

variable temperature insert (VTI), a vibration isolation stage and a homemade three axis magnetic vector magnet.

2.1.1 Cryostat, VTI and vibration isolation stage

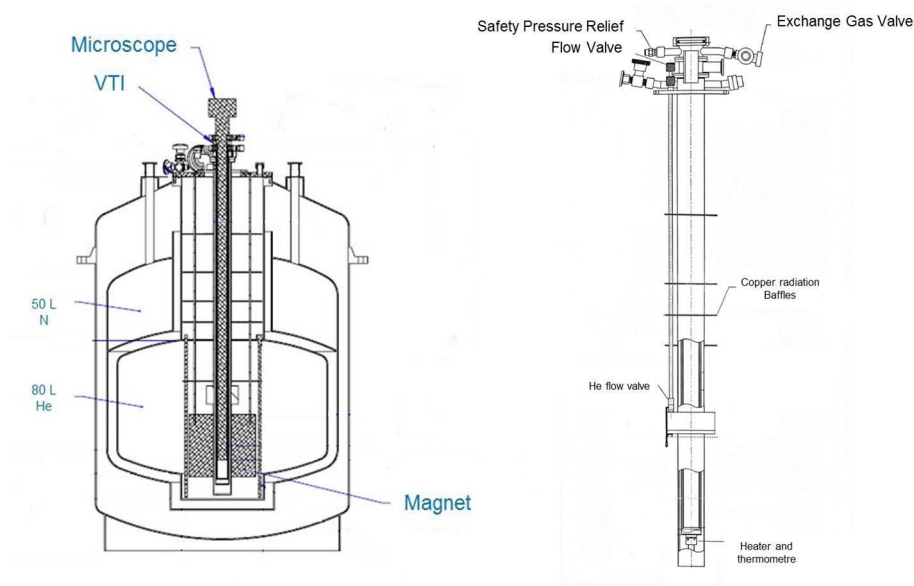


Figure 2.1: Schemes of the homedesigned cryostat and the VTI. Different chambers and walls are represented as black lines. In the left panel, the 80 L liquid He chamber is observed at the bottom, covered by the 50 L liquid Nitrogen chamber on top of it. Inside the cryostat, the position of the superconducting magnet, the microscope and the VTI are also shown. In the right panel, an scheme of the VTI shows the different valves to control the gas flow, the heater and thermometer to control its temperature and the copper radiation baffles in between.

Our cryostat consists in a 80 L liquid He main chamber where the VTI and the superconducting coil are placed. To isolate the liquid He bath from room temperature, the chamber is covered on top by a 50 L liquid nitrogen bath and both chambers are surrounded by a high vacuum compartment separated by stainless steel walls. The isolation of the liquid He bath allow us to perform experiments down to 2 K for a couple of days without refilling the cryostat. The Cryostat is specially designed to fit a commercial VTI and a magnet inside. An scheme of the cryostat is shown in figure 2.1.

The VTI (scheme in figure 2.1) is actually a double-layered vacuum can with two

spaces in between. It is designed to fit inside the magnetic coil in the He chamber. The inner space of the VTI is designed to fit our LT-AFM inside. To perform the measurements, the inner space is pumped to high vacuum and then filled with helium gas to a desired pressure (typically 0.5 atmospheres) to control the thermal contact between the liquid He bath and the microscope. The operating principle can be briefly described as follows. Through a narrow capillary, the helium liquid from the bath is siphoned into the outer space of the VTI, carefully controlled by a needle valve. Meanwhile, the gaseous helium is pumped out through a mechanical pump. Thus, the cooling power is generated by the evaporation process of liquid helium and cold gas flowing through the outer space. It was designed by Dr. Norbert Marcel Nemes from the *Universidad Complutense de Madrid* (UCM).

There are two working modes for VTI, that is, one-shot mode and continuous-flow mode. In one-shot mode, the needle valve is fully opened for a while, and a large amount of liquid helium is transferred into the outer space. Then, the needle valve is totally closed and no liquid comes in. Through sustained pumping, the base temperature could be achieved with a typical value of 1.3 K, which depends on the heat load and pumping speed. In continuous-flow mode, the needle valve is kept open at a delicate position and the liquid helium flows into VTI continuously. As the gaseous helium is pumped out, a wide range of temperature can be stabilized by controlling the temperature of the He gas with a 50 Ω heater on the bottom of VTI. The heater response is fixed by a commercial Cryocon Temperature controller. The VTI provides excellent thermal response with greater sample thermal stability allowing to a perfect control of the temperature at the microscope during the experiments with oscillations below 0.01 K.

The He evaporated and pumped out from the cryostat is heated and directed to a recovery line to liquefy it again at the *Servicios de Apoyo a la Investigacion Experimental* (SEGAINVEX) facilities.

The cryostat is placed on an isolation stage that makes negligible the vibrational noise during the measurements.

In figure 2.2, a picture of the cryostat, the isolation stage, the mechanical pump,



Figure 2.2: Picture of our set-up at the lab. In the image are shown the electronics to control the superconducting magnet (a), the temperature controller (b), the mechanical pump used to control the gas flow in the VTI (c), the isolation stage (d), the cryostat (e), the heaters to warm the cold He pumped from the VTI (f), the He recovery line (g) and the electronics of the LT-AFM (h).

the heaters, the recovery line and the control electronics is presented.

2.1.2 Three axis magnetic vector magnet

A three axis homemade magnetic vector magnet is placed inside the cryostat, in the liquid He bath. The magnet design is presented in reference [92] and consist in five superconducting coils made of NbTi wire, one coil for z axis field and two sets of split coils for the xy-plane field. The five coils are mounted in an Al cage. In figure 2.3 a and b, an scheme and a real picture of the coil are presented.

The magnet allow us to generate a magnetic field in any direction of the space up to fields of 5 T in the Z direction and 1.2 T in the X and Y direction, using a current of about 100 A. We have measured the magnetic field as a function of the distance and find a homogeneous field within a sphere around the center of the coil system of 0.2% for the magnetic field along the z axis, and of 1% for the magnetic field in the plane (Fig2.3 c). The three coil systems are equipped each with a persistent mode switch which allows maintaining the magnetic field constant when the liquid

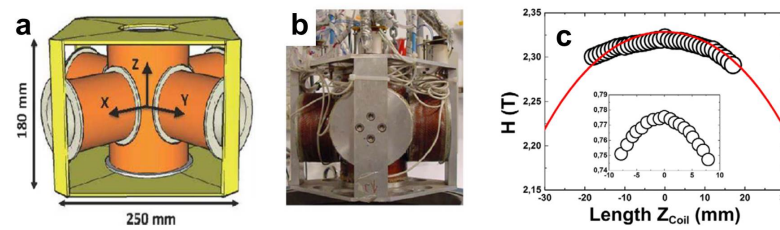


Figure 2.3: In a, and scheme of our homemade three axis vector magnet. The superconducting coils are represented in orange and the Al cage in yellow. One long coil is used to generate the z-axis magnetic field. For the in-plane field, we use two crossed split coil systems centred on the z-axis coil. The three directions of the space, X, Y and Z are marked with black arrows on the scheme of the coils, together with the real dimensions. In b, a real picture of the vector magnet. In c, we show the magnetic field vs z-axis position, with respect to the centre of the magnet when the z-coil is energized (50 A) (main panel) and when the x or y coils are energized (75 A, inset). Red line is a guide to the eye.

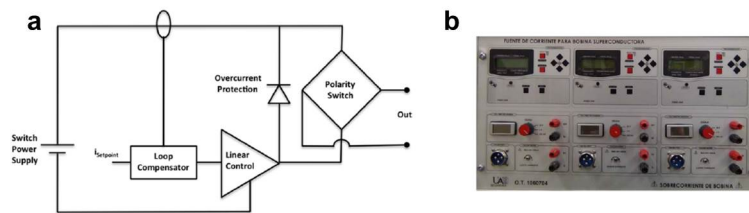


Figure 2.4: In a, we show a scheme of the current power supply for the magnet. In b, we show a photograph of the power supply. It is rather compact, 50 cm high and 80 cm long.

helium is above the magnet. The magnet is energized using a power supply with three independent current sources, each one has a commuted internal commercial stage of 5 V 100 A, followed by a voltage to current converter consisting of a stage providing linear regulation which uses MOSFET power transistors. Figure 2.4 shows an scheme of the circuits and a photograph of the power supply. The power supply was designed and made at SEGAINVEX mostly by M. Cuenca.

2.1.3 Low Temperature Microscope

The LT-AFM can be divided in two main parts, the insert and the head. The insert can be attached to the microscope head using low temperature connectors, what allow to connect it to multiple microscope heads with different equipments (AFM, SHPM, STM, etc). Radiation buffers are placed along a stainless steel tube that gives mechanical shielding and guide to all the necessary wires. It has a KF 40/50 connector on the top that fits in the variable temperature insert (VTI) space of the cryostat. An schematic representation of the microscope is presented in figure 2.5.

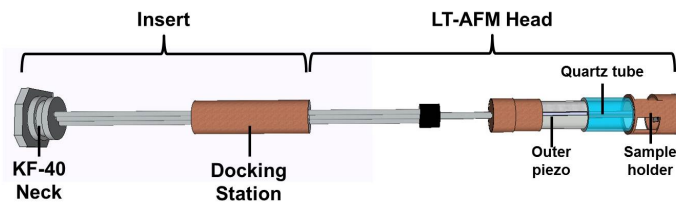


Figure 2.5: Scheme of the LT-AFM microscope. In the picture the hole microscope, insert and head, is shown. On the top of the microscope, the KF-40 neck that fits on the VTI. In the middle, the docking station to attach the insert to different microscope heads. In the head, the outer piezo, the quartz tube and the sample holder are shown.

The microscope head is formed by the AFM holder, two concentric lead zirconate piezotubes, a quartz tube, and the sample holder. A real picture of the LT-AFM head is shown in figure 2.6).

2.1.3.1 AFM holder

The AFM holder is attached to the inner piezotube by two screws. It has an commercial AFM alignment holder from NanoSensors, glued on top of a small piezo stack element, which is sandwiched between two alumina plates. The AFM probe is fixed on the AFM holder using a spring connected to the body of the holder. The AFM holder also has a Zirconium ferrule tube used to align the end of an optical fibre with respect the AFM probe. The optical fibre is used to control the cantilever displacement with the so-called *optical laser interferometer method*. The piezo bel-

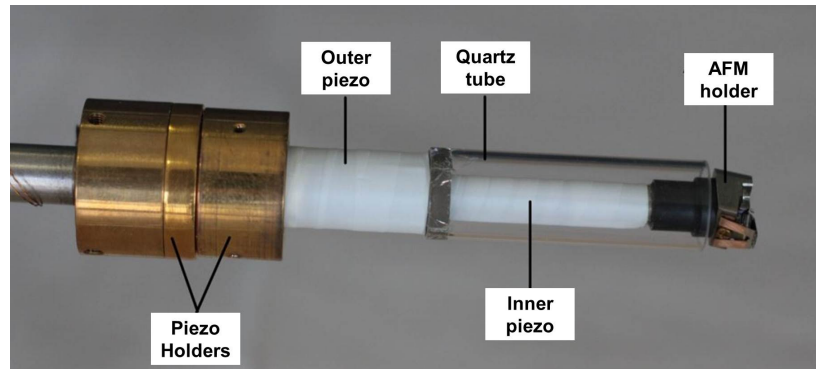


Figure 2.6: Real picture of the head of the LT-AFM. It shows the piezo holders, the two piezo tubes (wrapped in Teflon in the picture), the quartz tube and the AFM holder.

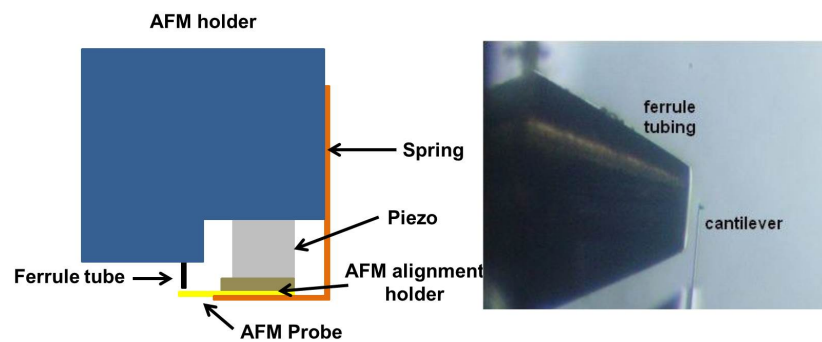


Figure 2.7: In the left panel, an scheme of the AFM holder. The body of the holder is represented in blue. The ferrule tube is shown in black, the AFM probe in yellow, the AFM alignment holder in brown, the piezo in grey and the spring in orange. In the right panel, a picture zoomed in the ferrule tube and the probe.

low the alignment holder is used to control the fibre-probe distance. An schematic representation of the AFM holder is presented in figure 2.7.

2.1.3.2 Sample holder

The sample holder is a hollow cylinder made of Phosphor bronze with a hole at the top that fits in the quartz tube. At the bottom, it has a plate where the sample is glued and a connector to bias the sample. At the side, it has a leaf spring used to attach it to the quartz tube. A picture of the sample holder is presented in figure 2.8.

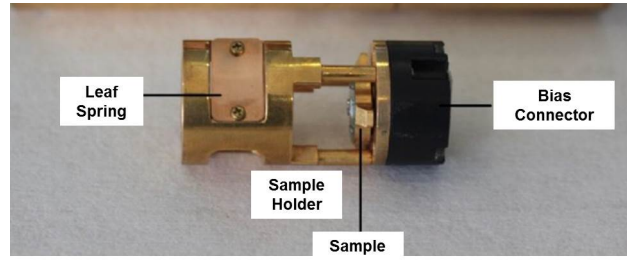


Figure 2.8: Real picture of the sample holder. In the image are visible, the leaf spring used to attach the sample slider to the quartz tube, the plate where is sample is glued and the bias connector.

2.1.3.3 Scanning and tip oscillation system

The inner piezotube is used to oscillate and scan the AFM probe over the samples. It has quadrant electrodes and a circular electrode at its apex as is schematized in figure 2.9. If and opposite voltage is applied to reciprocal electrodes, the tube will bend as is show in figure 2.9 a. On the other hand if the same voltage is applied to all quadrature electrodes with respect to the inner electrode, the tube will extent or contract in the Z direction. It has a $\approx 20\mu m$ scan range in the XY plane and a $\approx 1.5\mu m$ retract range in the Z direction at 2 K applying a voltage difference of 200 V between electrodes. The single electrode at its apex is used to oscillate the AFM probe by applying and oscillating difference of potential to the electrode as is schematized in 2.9 c.

2.1.3.4 Approaching-retracting mechanism

The outer piezotube and the quartz tube are used to perform the approaching an retracting movement of the sample with respect to the AFM probe using the so-called *stick-slip method* [93]. The piezotube has quadrant electrodes and the quartz tube is glued to its end.

The principle of the *stick-slip method* is schematized in figure 2.10. First, the sample holder is attached to the quartz tube with the leaf spring and is approached to a safe distance of the AFM probe by hand (A). Then, an exponential voltage is applied to the outer piezotube in about 3 ms contracting (retracting) it (B). During

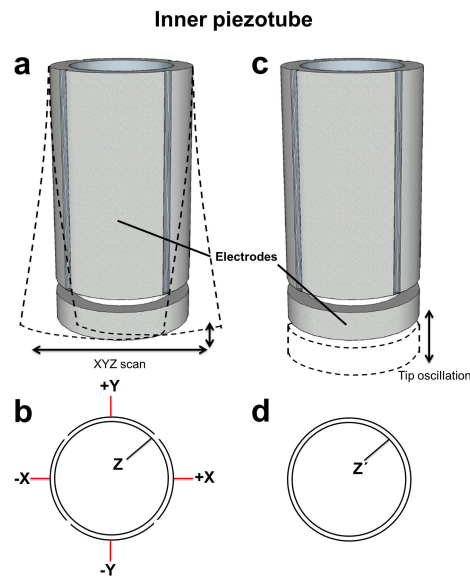


Figure 2.9: Scheme of the inner piezotube and its electrical contacts. On top, the motion of the piezotubes is schematically represented by dashed lines. On the bottom, the electrical contacts on the piezotube are shown as curved black lines. In a, an scheme of the scan movement of the inner piezotube is shown. In b, an scheme showing the five contacts to perform the scan movement, denoted by X, Y and Z. In c, an scheme of the oscillatory movement of the single electrode at the apex. In d, an schematic view of the electrical contacts of the single electrode to perform the oscillatory movement, denoted by Z' .

the exponential pulse, the sample holder moves together with the quartz tube due to the friction between them. Finally, the voltage is turned to zero in less than $1\mu\text{s}$ and the outer piezo is retracted (contracted). As a consequence, the sample holder slides on the quartz tube due to its inertia, approaching (retracting) the sample to the AFM holder (C). This slider mechanism can move few hundred grams at 4 K, successfully. Note that the success of the method depends on the equilibrium between the inertia and the friction force of the sample holder, which is controlled by the pressure of the leaf spring against the quartz tube. For this reason the quartz tube has to be carefully cleaned and the leaf spring tested at room temperature before all the measurements.

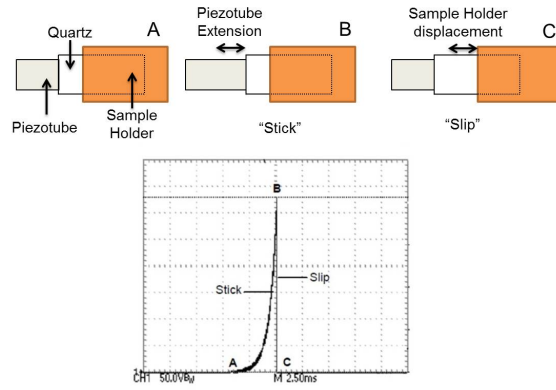


Figure 2.10: Scheme of the stick-slip method to move the sample holder. On the upper panel, an scheme of the LT-AFM head, showing the outer piezotube, the quartz tube and the sample holder. Dashed lines are used to represent the portion of the quartz tube inside the sample holder. A, B and C, represent the three steps during the sample holder displacement described in the text. In A, the sample holder is attached to the quartz tube. Then, between A and B an exponential voltage pulse is applied to the piezotube to extend or contract it. Finally between B and C the voltage is turned down to zero in less than $1 \mu\text{s}$. As a result the sample holder is displaced a given distance. In the lower panel, a real image of the pulse on an oscilloscope, with the three steps presented on the top panel marked with A, B and C letters.

2.1.3.5 Optical laser interferometer method

As it was introduced before, the optical laser interferometer method [94] [95] [96] was used in our LT-AFM to detect the displacement of the AFM probe. In this method, a laser (I) is pointed to the cantilever through an optical fibre. At the end of the fibre, some of the light is reflected by the surface (I_{rs}) and some escape the fibre and goes to the cantilever that acts as a mirror, then the laser is reflected (I_{rc}) through the fibre, back to the source where they interfere. The interferometer pattern is a function of the optical path of each beam and therefore of the fibre cantilever separation, which allows to monitor the bending of the cantilever. It is schematically represented in figure 2.11. The photocurrent at the interferometer can be described as follows [97]:

$$I_{inter} = I_0 \left[1 - V \cos\left(\frac{4\pi d_{f-c}}{\lambda}\right) \right] \quad (2.1)$$

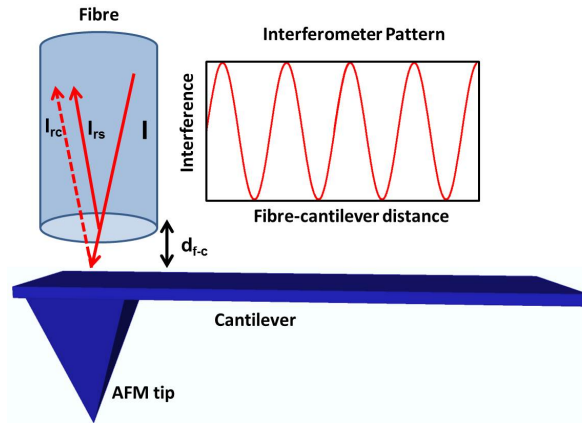


Figure 2.11: Scheme of the interferometer sensor method. The laser beam, I , travels through the fibre. At the end of the fibre, some of the light is reflected by the surface (I_{rs}) and some escapes the fibre and goes to the cantilever that acts as a mirror, then the laser is reflected (I_{rc}) through the fibre, back to the source where I_{rs} and I_{rc} form an interferometer pattern (red line in the plot) as a function of the fibre-cantilever distance, d_{f-c} .

$$I_0 = \frac{I_{max} + I_{min}}{2} \quad (2.2)$$

$$V = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} \quad (2.3)$$

where I_0 is the midpoint current, V the visibility, d_{f-c} the fibre-cantilever separation, R the reflectivity and λ the laser wavelength. The slope of the interference is:

$$m = 4\pi I_0 \frac{V}{\lambda} \quad (2.4)$$

As all the magnitudes except d_{f-c} are constant, the photocurrent can be used to measure the fibre cantilever distance and therefore, the oscillation of the cantilever. To maximize the accuracy of the measurements, the equilibrium distance between the fibre and the cantilever is chosen to make the slope of the interferometer pattern maximum. This point is determined by measuring the interference pattern when changing the cantilever-fibre distance with the piezo beneath the cantilever. The

piezo is driven between 0 – 125 V forward and backward, with respect to the fibre. An example of the interferometer patten recorded at room temperature in our microscope is presented in figure 2.12.

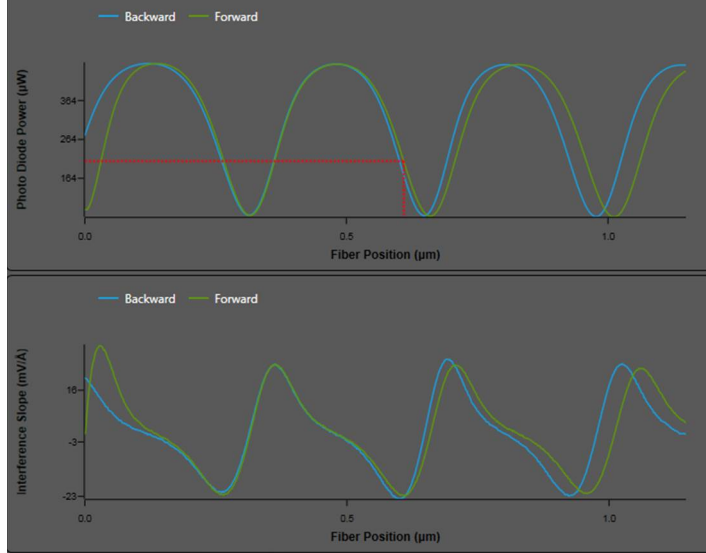


Figure 2.12: In the upper panel, the interferometer pattern obtained by our LT-AFM at room temperature as a function of the fibre position with respect to the resting cantilever. In the lower panel, the slope of the interferometer patten. The green and blue lines represents the interferometer patten obtained when approaching and retracting the AFM probe to the cantilever. The pattern is used to lock the fibre-tip distance at the maximum slope, marked by red dashed lines in the image.

2.1.3.6 LT-AFM controller

The bending of the cantilever is measured by the interferometer and received by a digital Phase Lock Loop (PLL) card, which excites the cantilever at the desired frequency and measures the frequency shift, the phase and the Amplitude of the output signal from the cantilever.

The LT-AFM controller has a very low noise power supply unit. It has four channels of low noise high voltage amplifiers to drive scan piezo. A Digital PID loop is operated at 250 kHz for the feedback, which gives analog 24 Bits signals to drive the Z position and 32 Bits digital output for the software. A sample slider card produces exponential pulses up to 400 V for the stick-slip mechanism. A diagram of the control

mechanism of our LT-AFM microscope is presented in figure 2.13. Both, electronics and software were developed by Nanomagnetism Instruments LTD.

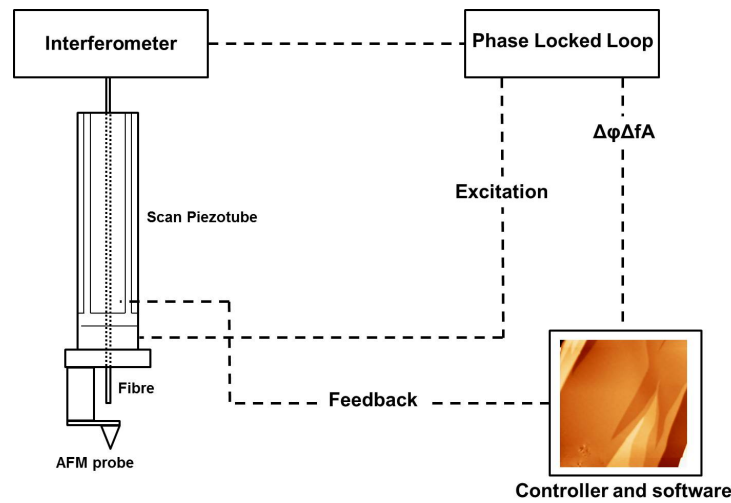


Figure 2.13: LT-AFM control scheme. In the image, different elements of the LT-AFM head are schematized using black lines. Dotted lines are used to represent the fibre position inside the piezotube. Dashed lines are used to represent the connection between the microscope and the different element in the control electronics. The information of the AFM probe displacement is recorded in the interferometer and transmitted to the Phase Locked Loop (PLL) card which excites the cantilever at the resonance frequency and measure the frequency shift, phase and amplitude changes. Either frequency shift or amplitude change is used for a feedback which is operated by the controller. The scan and coarse approach mechanism is also managed by the controller.

2.1.3.7 Operational modes

Several measurement modes have been introduced in AFM to be able to measure variety of forces in different conditions [98]. In this section we will introduce the two main modes used during the thesis in our LT-AFM, the *dynamic mode* and the *MFM mode*:

2.1.3.7.1 Dynamic mode In the dynamic mode the AFM cantilever is oscillated at a given amplitude at its resonance frequency and placed near to the sample (5-15nm). In such scenario, the tip is near enough to the surface to interact via short

range Van der Waals (VdW) forces with the surface [98, 99]. The oscillation amplitude should be large enough to ensure that the restoring force at the lower turning point is larger than the attractive force between tip and sample. This will avoid an instability, which would stop or at least seriously distort the oscillation. The tip-sample interaction causes a shift in the resonance frequency which is used to measure the force acting on the tip [98, 99].

If we approximate the cantilever and the tip as point-mass spring (figure 2.14), we can consider the AFM probe as a damped oscillator due to friction forces, with some driving force, and into a force field created by the tip sample interaction, then its movement can be described by a linear, second-order differential equation [98]:

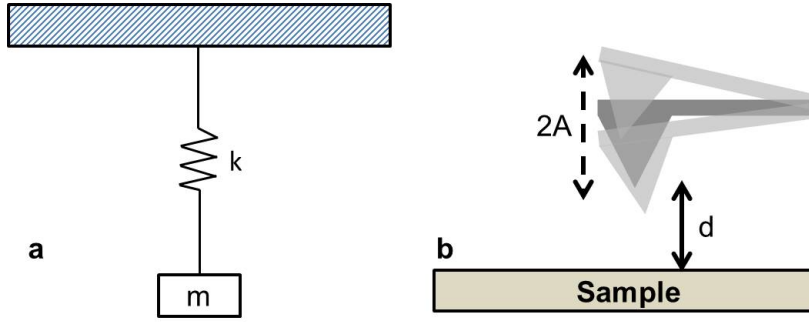


Figure 2.14: Point-like mass spring as an approximation of the AFM cantilever. In a, a simple point like mass spring as an approximation of the cantilever movement. In b, an scheme of the oscillating tip is showed.

$$m\ddot{z} + \delta\dot{z} + k(z - z_0) = F_d + F_{t-s} \quad (2.5)$$

where $F_d = F_0 \cos(\omega t)$ is the driving force provided by the piezotube to oscillate the cantilever at an angular frequency ω , F_{t-s} is the force due to the tip sample interaction and δ is the damping factor which can be calculated as [98]:

$$\delta = \frac{k}{f_0 Q} \quad (2.6)$$

where Q is the quality factor of the oscillator and $f_0 = \sqrt{\frac{k}{m}}$ the resonance frequency of the free oscillator. To solve equation 2.5, F_{t-s} is expanded into a Taylor series:

$$F_{t-s}(z) = F_{t-s}(z_0) + \frac{\delta F_{t-s}(z_0)}{\delta z}(z - z_0) \quad (2.7)$$

and equation 2.5 can be rewritten as:

$$m\ddot{z} + \delta\dot{z} + k(z - z_0) = F_d + F_{t-s}(z_0) + \frac{\delta F_{t-s}(z_0)}{\delta z}(z - z_0) \quad (2.8)$$

$$m\ddot{z} + \delta\dot{z} + \left[k - \frac{\delta F_{t-s}(z_0)}{\delta z}\right](z - z_0) = F_d + F_{t-s}(z_0) \quad (2.9)$$

The term $k - \frac{\delta F_{t-s}(z_0)}{\delta z}$ in equation 2.9 is called the effective spring constant, k_e . Solving 2.9, we will find:

$$z(t) = z_0 + A\cos(2\pi f_e t - \phi) \quad (2.10)$$

Equation 2.10 represents an harmonic oscillator with angular frequency ω_e different that the angular frequency of the free oscillator and with a phase shift of ϕ . The frequency of the oscillator is:

$$f_e = \sqrt{\frac{k}{m} - \frac{1}{m} \frac{\delta F_{t-s}}{\delta z}} = \sqrt{f_0^2 - \frac{1}{m} \frac{\delta F_{t-s}}{\delta z}} \quad (2.11)$$

Taking into account that $k \gg \frac{\delta F_{t-s}}{\delta z}$, equation 2.11 can be reduced to:

$$\Delta f \approx \frac{f_0}{2k} \frac{\delta F_{t-s}}{\delta z} \quad (2.12)$$

Therefore, when the cantilever is brought into a force field, the resonance frequency will be shifted and the force gradient can be measured by measuring the shift in the resonance frequency or if the excitation frequency is kept constant, by measuring the change in amplitude (ΔA) or the change in phase ($\Delta\phi$) of the oscillation as is seen in Figure 2.15.

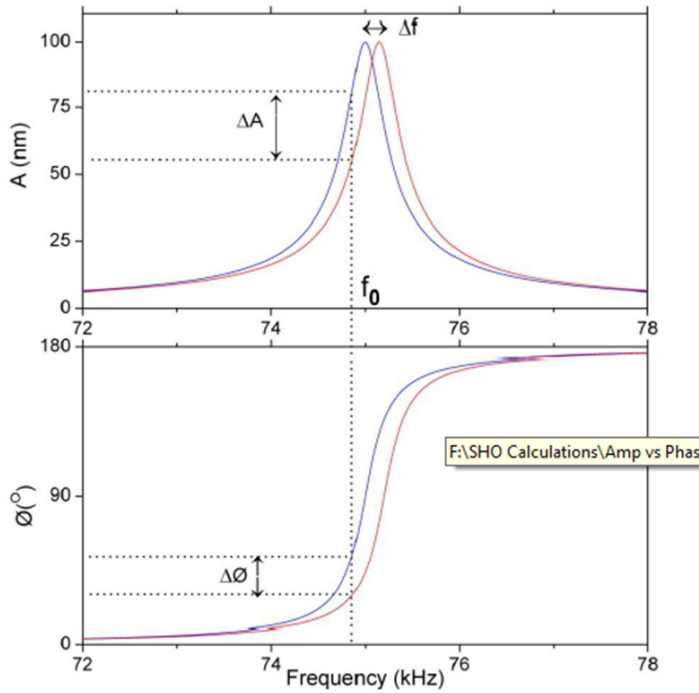


Figure 2.15: Oscillation amplitude (A) and phase (ϕ) versus frequency. In the plots, the blue line represents the amplitude and phase of the cantilever far from the surface and the red line their shift due to the interaction with the sample. In the upper panel, the oscillation amplitude decreases by ΔA when the tip is oscillated at a frequency f_0 due to the shift in the oscillation frequency Δf . In the lower panel, the corresponding change in the oscillation phase $\Delta\phi$ is shown.

In our system, in the dynamic mode, topographic images are measured by keeping constant the excitation frequency and the amplitude of the oscillation. A feedback is used to keep the amplitude constant by changing the length of the scan piezotube. The topography is measured using the change in the length of the piezotube.

2.1.3.7.2 MFM mode In MFM, the interaction between a magnetic probe and a magnetic sample is measured.

The topography and the magnetism of a magnetic sample can be measured independently using the a modification of the dynamic mode, called *two pass mode*. In this mode, the operational parameters are chosen such that either, the non-magnetic or the magnetic interaction becomes dominant. This is achieved due to the small contribution of the magnetic forces at small distances ($< 5\text{nm}$) and the VdW forces

at big distances ($> 50nm$) [98].

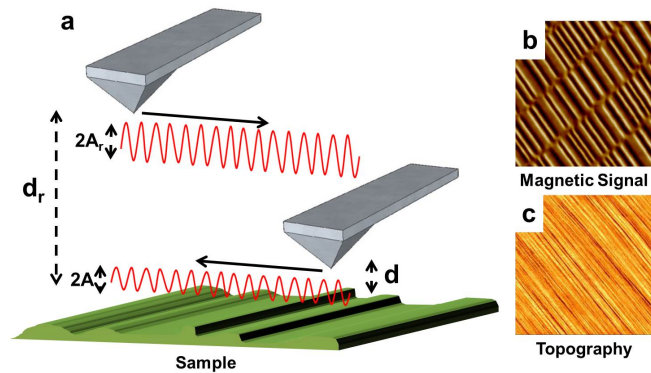


Figure 2.16: In a, an schematic representation of the MFM two pass mode. A first scan near the surface at a distance, d , and amplitude, A , is made to record the topographic profile. Then, the tip is retraced a distance d_r and scans the same profile at a constant height, d_r , and amplitude A_r to measure the long range magnetic interaction. The trajectory of the tip during the scans is represented with a red line. In b and c two examples of the magnetic and topographic images obtained in a Hard Disk Drive (HDD).

In the MFM two pass mode, the topography is measured in a first scan (forward) in the same way that in the dynamic mode. Then, the probe is retraced a large distance and a second scan is performed (backward). In the second scan, the tip is oscillated at the same frequency that the first scan but with the amplitude feedback opened. In this case, the length of the piezotube is used to keep the tip-sample separation constant, using the information of the topographic scan. Then, the tip-sample magnetic interaction is recorded by measuring either ΔA or $\Delta\phi$. An scheme of the two pass mode is presented in figure 2.16.

The MFM contrast is associated to the magnetic domains in the sample. Assuming that the magnetization of the tip is in the axial direction, when the stray field from the sample is parallel to the tip magnetization there is an repulsive force which is typically represented in MFM images as dark contrast and if the stray field is antiparallel the contrast will be light.

2.2 Characterization of MFM probes for low temperature experiments

During the thesis mainly two AFM probes were used: *Nanosensors Point Probe Plus Force Modulation Mode - Reflex Coating* (PPP-FMR) and *Nanosensors Point Probe Plus Magnetic Force Microscopy - Reflex Coating* (PPP-MFMR) probes. Both probes are made of Silicon with a cantilever length of $225 \mu\text{m}$, width of $30 \mu\text{m}$, thickness of $3 \mu\text{m}$ and spring constant of 3 nN/nm . The difference between them is that PPP-MFMR probes have a CrCo alloy layer of $\approx 20\text{nm}$ deposited on the tip and the cantilever, allowing to magnetic characterization of the samples. PPP-FMR probes were used to preliminary topographic characterization and PPP-MFMR probes to magnetic characterization. Figure 2.17 shows the AFM probes geometry with a rectangular cantilever and a sharp tip at the end.

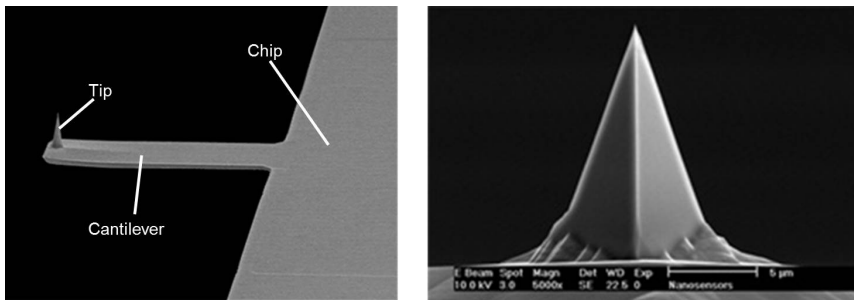


Figure 2.17: SEM images of an AFM probe. In the left panel, the silicon chip where the probe is lithographed, the rectangular cantilever and the pyramidal tip. In the right panel, a zoom on the tip showing its pyramidal shape.

The CrCo alloy that cover the PPP-MFMR probes is a ferromagnetic material, and therefore it presents an hysteresis cycle in its magnetization when changing the magnetic field. At room temperature the coercive field ($\approx 300\text{Oe}$) and magnetic moment ($10\text{-}13 \text{ emu}$) are provided by the manufactured, but for lower temperatures a magnetic characterization of the probe must be done in order to obtain its coercive tip to a proper tip magnetization. For this purpose we have measured the ferromagnetic domains of a Hard Disk Drive (HDD) as a function of the temperature from 300 K to 2 K .

Ferromagnetic domains of HDD are known to have coercive fields much greater

(2000-5000 Oe) [100] than typical MFM probes. This makes them the perfect candidates to characterize the hysteresis cycle of MFM probes as their magnetic state will not change by the small magnetic fields needed to change the state of the tip.

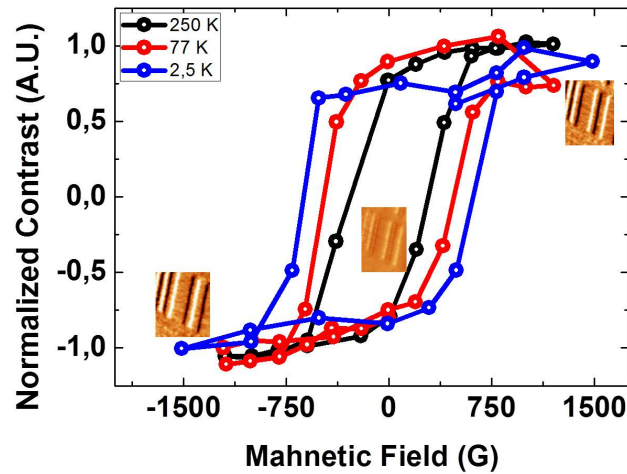


Figure 2.18: Ferromagnetic cycles of a PPP-MFMR tip measured at 205 K, 77 K and 2.5 K, using a HDD as a sample. Three MFM images of the magnetic domains at 1500, 0 and -1500 Oe at 2.5 K of the HDD are also shown. In the images, the switching of the tip magnetization is recorded as a change in the contrast from dark to light and vice versa in the magnetic domains of the HDD.

The hysteresis cycle of a typical PPP-MFMR probe was recovered by measuring the surface of a HDD at different fields and constant temperature for several temperatures. We have calculated the magnetic moment of the tip (in arbitrary units) as the difference in contrast between domains walls of the HDD in the MFM image [101]. The hysteresis cycles for 250 K, 77 K and 2.5 K are represented in figure 2.18. The coercive field of the tip changes from ≈ 300 Oe at RT to ≈ 1000 Oe at 2.5 K.

This characterization is of huge importance to a proper interpretation of the MFM images, as they are the result of the interaction of the tip magnetization and the stray field of the sample [98].

2.2.0.1 MFM features

To illustrate the importance of the characterization of the MFM tips at low temperatures, we will present the particular cases of MFM images of superconducting vortices when a PPP-MFMR tip is magnetized below and above its coercive field.

If the tip is not magnetized up to its coercive field, different domains with different orientation will appear at the tip and could cause artefacts in the magnetic image [102–104]. A good example are the series of images that are presented in figure 2.19. In the images, superconducting vortices were measured in $\beta - Bi_2Pd$ at 2 K with a tip magnetized up to 500 Oe at 5 K, which is well below the coercive field. As it was presented in the introduction of the thesis, superconducting vortices are known to have circular shapes with radius determined by λ . In our experiment, the superconducting vortices appear as star-like features instead of as circles, pointing out the existence of a complicated distribution of the magnetic domains of the tip. A simple explanation of the origin of the star-like features will be discussed in the following:

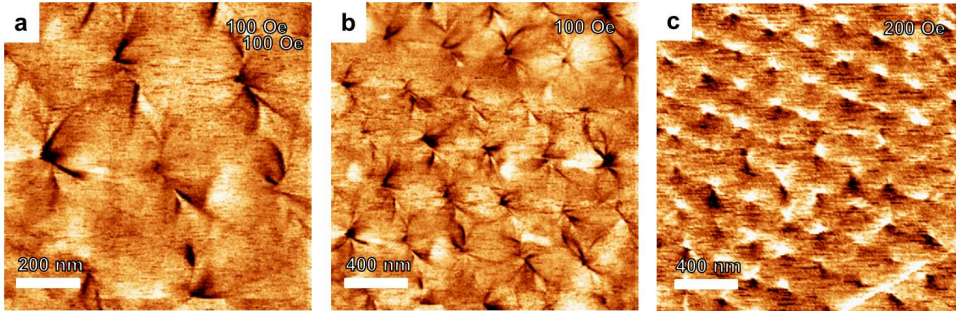


Figure 2.19: Examples of star-like features at superconducting vortex positions when a MFM tip is not magnetized up to its coercive field. The three MFM images were taken at 2 K. Images a and b were taken at 100 Oe while c was taken at 200 Oe.

The tip-vortex interaction can be approximately written as [105]:

$$\frac{\delta F}{\delta z} = m_x \frac{\delta^2 H_x}{\delta x^2} + m_y \frac{\delta^2 H_y}{\delta y^2} + m_z \frac{\delta^2 H_z}{\delta z^2}. \quad (2.13)$$

Where $m_i; i = x, y, z$ are the components of the magnetic dipolar moment of the tip and $H_i; i = x, y, z$ are the components of the field created by the vortex. The field created by a vortex can be described as [29]:

$$B = \frac{\phi_0}{2\pi\lambda^2} K_0((r/\lambda)e^{-\frac{2\pi z}{d}}) \quad (2.14)$$

where K_0 is the modified Bessel functions of second order and r and z the radial and vertical distances from the vortex core.

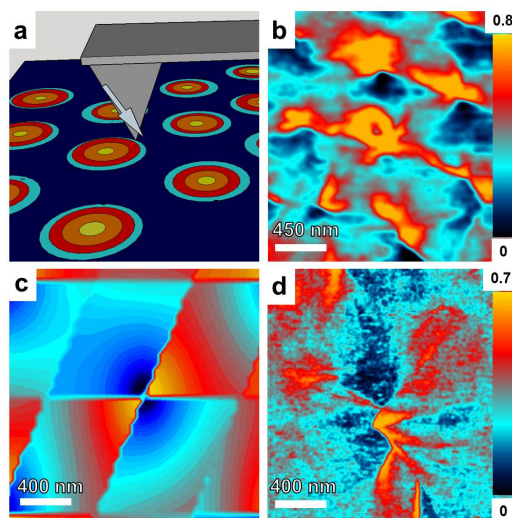


Figure 2.20: In a, we schematically shown a MFM tip with with a non-zero in plane component of the dipolar moment (grey arrow on the tip) and the vortex lattice (coloured circles bellow). In b, we show star like vortices obtained at 2 K and 100 Oe with a tip magnetized with 300 Oe at 5 K. In c and d we compare a simulation and an MFM image of a single vortex measured with a tip with an in-plane magnetization component. The same colour scale is used at the scheme, the simulation and the MFM images.

Typically the components X and Y of the dipolar moment of the tip are neglected due to the predominant Z magnetization of the tip [104]. To explain the star shaped vortices, we assume that this is no longer the case, as schematically show in Fig.2.20 a. We consider a tip with non zero X an Y components. By calculating spatial maps of the force gradient sensed by the cantilever using equation 2.13, we find that indeed finite x and y components of the magnetization provide star shaped vortices as is shown in Fig.2.20 c. The starlike features obtained by our simple model, are

very similar to the vortices shapes obtained in the experiments. Real MFM images of starlike vortices are shown in figures 2.20 b and d together with the result of the simulations to highlight this fact.

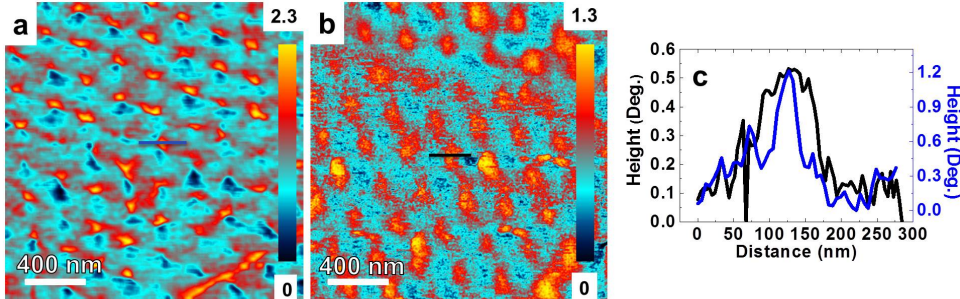


Figure 2.21: Here we show images taken at the same temperature (2 K) and magnetic fields (300 Oe) with a tip having in-plane components of the magnetization (a) and only z-component (b). The blue and black profiles shown in c were taken at the lines with the same colour in a and b. The profiles provides a measure of the spatial resolution, which is improved when the tip has an in-plane anisotropy.

In order to compare the images obtained by a MFM tip magnetized below and above its coercive field, we have measured the vortex lattice at the same area and the same magnetic applied field with different tip magnetization conditions. In figure 2.21 two cases are shown, one with the tip magnetized at 500 Oe (below its coercive field) and another with the tip magnetized at 1500 Oe (above its coercive field). Interestingly, when the tip is not magnetized above its coercive field, the lateral resolution is improved as is shown in figure 2.21 c. The vortex profile in figure 2.21 a is thinner than in figure 2.21 b. We suggest that the MFM lateral resolution can be improved by tuning the magnetization of the tip or what is the same, the amount of magnetic domains in the Z, X and Y direction.

2.3 Crystal growth

Binary phase diagrams contain the information of all known crystallographic structures for a pair of elements as a function of the composition and the temperature. An example of a simple binary phase diagram for arbitrary elements, A and B, is presented in figure 2.22.

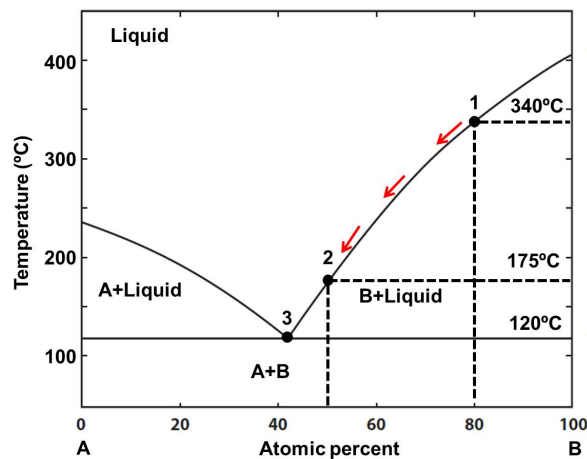


Figure 2.22: Example of a binary phase diagram. Black curved line represent the solid-liquid equilibrium line. Point 1 and 2 are the initial and final points in the growing process described in the text and 3 is the eutectic point. Dashed lines are used to point the exact temperature and composition of points 1 and 2. The red arrows represents the evolution of the composition of the liquid phase as explained in the text.

The phase diagram in figure 2.22, consists in a series of vertical, horizontal and curved lines. The vertical lines, represent different known stable compositions of A-B crystals. Curved lines are the solid-liquid equilibrium lines of the compounds, meaning that at a given composition, above this line, the mixture will be in the liquid phase and below this line, a determined solid phase or phases will grow. The horizontal lines represents the temperatures where a different solid will grow

The *solution growth method* is a powerful technique to grown single crystals using the information provided by binary phase diagrams [106]. In order to grow single crystal of a determined composition, the desired amount of A and B is placed inside two alumina crucibles with glass wool in between and sealed in a quartz ampoule in a He atmosphere using a blowtorch. The mixture is heated up to melt it, and then cooled down to the desired temperature where the crystals will start to grown, for example, for a composition of 80 percent of B in figure 2.22, 340°C (1). Then, the ampoule is slowly cooled down to a temperature above the eutectic point, as the temperature decreases, the crystal becomes bigger and the liquid mixture composition varies following the liquid line (2). If the temperature is decreased below the eutectic

point (3), the remnant liquid will solidify enclosing the B crystals, making especially hard to separate them. For this reason, the alumina crucibles has the glass wool in between. Taking quickly the ampoule from the furnace and spinning it, will separate the liquid and the solid phases at both sides of the glass wool that acts as a filter. Then, clean and intact B crystals can be obtained for their research. An schematic representation of the growing process is presented in figure 2.23

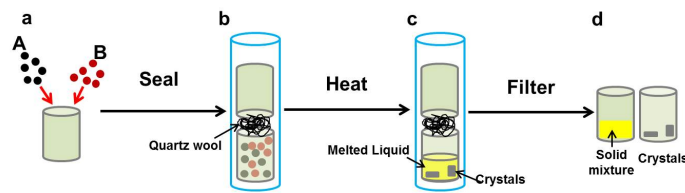


Figure 2.23: Scheme of the solution growth method. Alumina crucibles are represented as beige cylinders and the quartz tubes with blue lines. In a, the desired elements A and B (black and red circles) are placed inside the alumina crucibles. Then in B, they are encapsulated between two alumina crucibles with quartz wool in between (black line) and sealed in a quartz tube in vacuum. In c, the quartz ampoule is heated in a furnace to melt the elements inside and grow the crystals. Finally, in d, the crystals are separated from the flux excess by spinning the quartz ampoule.

Sometimes, the interesting phase of a given crystal is not stable at ambient conditions. This phase could be stable for example above 500° and transits to another (uninteresting) phase below this temperature. In this case, it is still possible to obtain the desired crystals by forcing them to cool down very quickly from a temperature where they are stable to the room temperature by immersing them in water or liquid nitrogen for example. Doing so, the crystal will not transit to the low temperature phase and will remain in the desired phase [106]. With this process, the remnant liquid is not filtered and the crystals will be immersed in an amorphous solid with the composition of the remnant liquid.

In conclusion, knowing the binary phase diagram of two elements allow to grow binary crystals using the *solution growth method* in a simple way.

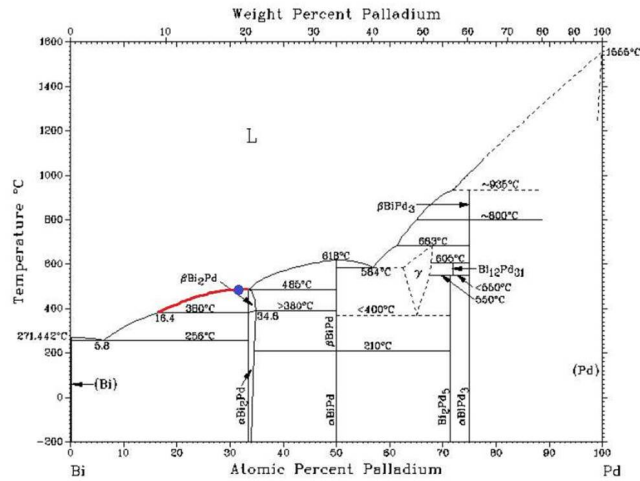


Figure 2.24: Phase diagram of Bismuth and Palladium system. The red line represents the liquid-solid equilibrium line for $\beta - Bi_2Pd$ and the purple circle the initial liquid concentration during the growth.

2.3.1 $\beta - Bi_2Pd$ single crystals growth

During the thesis, single crystals of $\beta - Bi_2Pd$ were grown using slight excess of Bi [107], [108]. We grew our samples from high purity Bi (Alfa Aesar 99.99%) and Pd (Alfa Aesar 99.95%). Bi and Pd were introduced in alumina crucibles and sealed in quartz ampoules at 140 mbar of He gas using a blowtorch. Then, the ampoules were heated from room temperature to $900^\circ C$ in 3 h, maintained 24 h at this temperature, slowly cooled down to $490^\circ C$ in 96 h and finally cooled down to $395^\circ C$ in 200 h in a furnace. This temperature is about $15^\circ C$ above the temperature for the formation of the $\alpha - Bi_2Pd$ phase [109] (figure 2.24). To avoid formation of the α phase, we quenched the crystals down to ambient temperature by immersion in cold water. We obtained large crystals of $5mm \times 5mm \times 3mm$ (inset in figure 2.25).

To characterize them, we made x-ray diffraction on crystals milled down to powder (Fig.2.25, using x rays with wavelength 1.54 \AA). We find $\beta - Bi_2Pd$ (I4/mmm, see Ref.[111]) with refined lattice parameters $a = b = 3.36(8) \text{ \AA}$ and $c = 12.97(2) \text{ \AA}$ and no trace of $\alpha - Bi_2Pd$. We made in total twelve growths, varying slightly the conditions for the quench, growth temperature and initial composition, and obtained always crystals with a resistivity vs temperature very similar to the one shown in Fig 2.25.

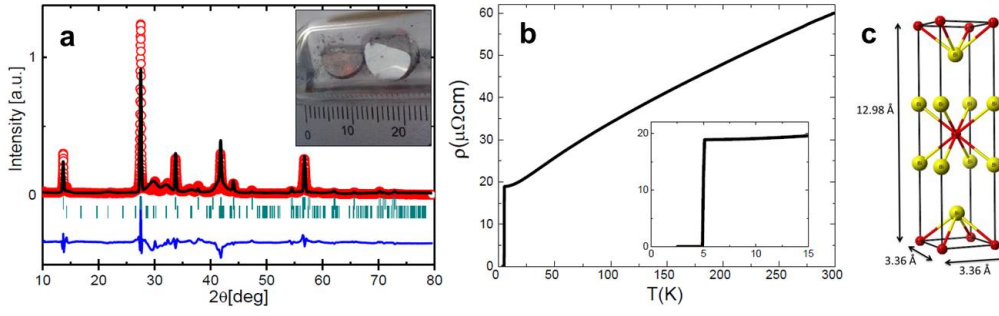


Figure 2.25: Powder diffraction pattern of β - Bi_2Pd . Red symbols are the experimental points. The black line is the best fit to β - Bi_2Pd diffraction pattern [110]. Residuals are given by the blue line. The two series (upper and lower) of vertical green strikes represent, respectively, the position in 2θ scale of the reflections from the β - Bi_2Pd ($I4/mmm$) and α - Bi_2Pd ($C12 = m1$) phases. The inset show a photograph of one β - Bi_2Pd crystal. In b, the temperature dependence of the resistivity. In c, the unit cell with the lattice parameters of β - Bi_2Pd

To ensure that the crystals composition is homogeneous on the hole crystal and discard the presence of Bismuth or Pd clusters in it, we have performed Energy-dispersive X-ray spectroscopy (EDX) measurements in a scanning electron microscope (SEM) at the *Servicio Interdepartamental de Investigacion* (SIdI) of the Universidad Autonoma de Madrid. We found that the 2:1 stoichiometry is constant in the hole crystal and that there is not any presence of Bi or Pd precipitates on it. We have measure the very same sample before and after exfoliate it using the Scotch tape method, finding the same results in the outer and inner layers of the β - Bi_2Pd crystals. A SEM image of the crystal with the three different areas where EDX experiments were performed is presented in figure 2.26.

EDX experiments reveals and almost constant composition of $\approx 65at.\%$ of Bi and $\approx 35at.\%$ of Pd on the hole sample, pointing out the very good quality of our crystals. An example of the EDX spectra on the crystals is also presented in figure 2.26 b.

The superconducting vortex lattice in β - Bi_2Pd at very low fields was measured during the thesis and our result will be presented in chapter 3.

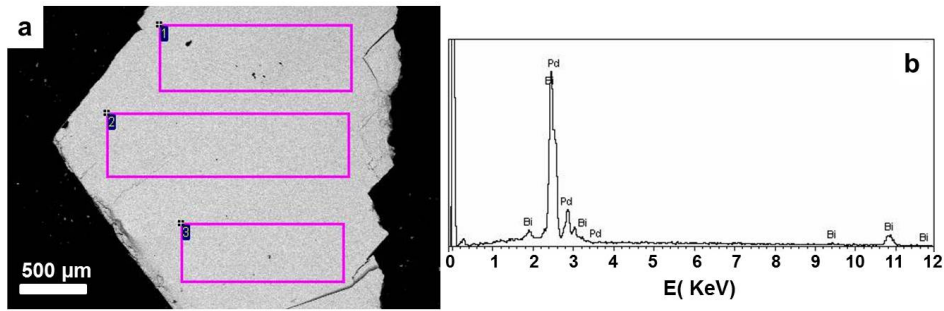


Figure 2.26: In a, a SEM image of an $\beta - Bi_2Pd$ crystal. The pink rectangles points the areas where different EDX spectres were measured. In b, the EDX spectra measured in the area marked with a 2 in the SEM image. Each peak of the EDX spectra is marked with the symbol of the corresponding element. The spectres at different areas of the crystal shown a perfect match with the Bi_2Pd composition.

2.4 Summary and conclusions

In conclusion, we have successfully implemented an experimental set up that allows us to perform AFM-MFM measurements between 1.8 K and 300 K, applying magnetic fields in any direction of the space up to 5 T in the Z direction and 1.2 T in the X and Y directions. With this set up, we have characterized for the first time the hysteresis cycle of MFM commercial probes as a function of the temperature from 1.8 to 300 K. We have also found that the MFM images of superconducting vortices show star-like features at vortex positions when the MFM tip is magnetized bellow its coercive field. Interestingly, at some magnetization fields, bellow the coercive field of the tips, the lateral resolution is improved with respect to tips magnetized above their coercive field. In addition, during the thesis, single crystals of $\beta - Bi_2Pd$ and Mn_5Si_3 were successfully grown via the solution growth method and characterized in our lab, presenting different properties as superconductivity or anomalous hall effect.

Vortex lattice at very low fields in the low κ superconductor $\beta - Bi_2Pd$

3.1 Introduction

Historically, magnetic microscopy techniques have been of huge importance in the study of superconducting materials. The first visualization of the vortex lattice was done by Essman and Trauble [112] in Pb by Bitter magnetic decoration in 1967 (figure 3.1 a). The Bitter decoration technique consists in depositing small magnetic particles on the surface of the superconductor. Under a magnetic field, the magnetic particles will be aligned with the magnetic flux lines, pointing the position of the individual vortices. Bitter decoration was also the first technique used to visualize the vortex lattice in high T_C superconductors in 1987 by Gammel et al. [113] in YBCO (figure 3.1 b). The MFM was used for first time to measure the vortex lattice in a superconductor in YBCO by Moser et al. [114] in 1995 in MgB₂ (figure 3.1 c).

A few pioneers works of magnetic bitter decoration have studied the *intermediate state* in Type I and the *intermediate mixed state* (IMD) in Type II superconductors, proving the coexistence between domains of normal and Meissner state in type I and between Meissner and mixed state in type II superconductors [40, 42, 43, 115, 116].

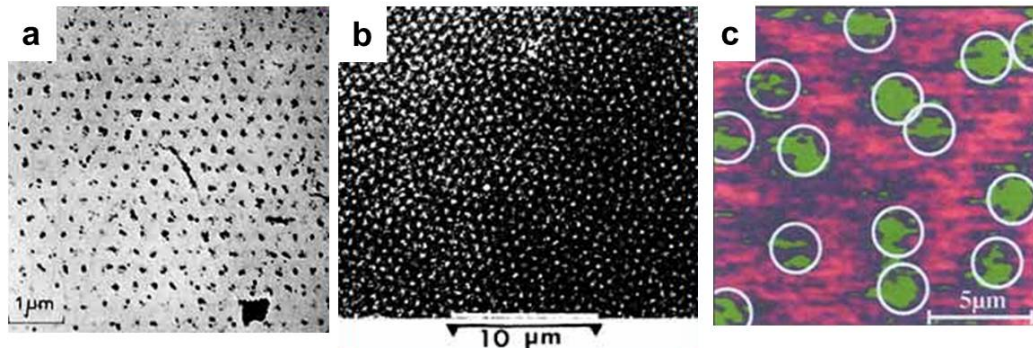


Figure 3.1: a shows the first magnetic image of superconducting vortices in 1967, it was measured by bitter decoration technique in Nb. Black dots points the position of the vortices, revealing an hexagonal lattice. b shows the first visualization of the vortex lattice in a High Tc superconductor (YBCO) in 1987, also by bitter decoration technique. White dots points the position of the vortices. c shows the first image of superconducting vortices measured by MFM in MgB₂ in 1995. Vortices are shown as green spots, in this case showing disordered arrangement. Images from references [112] [113] and [114]

3.1.1 Single and multi band superconductors in the IMS

At fields below H_{c1} , in type II superconductors, the internal magnetic field can be strongly inhomogeneous. Because of the full flux expulsion of the Meissner state, demagnetizing effects can dominate the magnetic field behaviour. The sample separates into domains with zero induction $B = 0$ and an induction close to $\mu_0 H_{c1}$ between $(1 - N)\mu_0 H_0 < \mu_0 H < \mu_0 H_0$ [40].

Thin films of Nb, TaN, PbIn and other materials show a IMS [40–43]. In high quality single crystal of Nb with $\kappa = 1.1\sqrt{\frac{1}{2}}$, flux expulsion coexists with regions showing a vortex lattice. Small angle neutron scattering finds exactly the intervortex distance expected at H_{c1} in the vortex domains [42]. The area occupied by the domains with zero induction decreases linearly with the magnetic field, so that the magnetic induction reaches the value corresponding to the mixed phase [42, 43]. The morphology of the IMS domains is mainly governed by geometric barriers preventing domain nucleation [116], surface barriers which hamper the entrance of flux lines into the sample [117, 118], VL anisotropies [42] as well as pinning forces [119]. A few selected images of different domains geometries in the intermediate mixed state in

Nb are presented in figure 3.2. Nb has only a single superconducting gap, but a highly complex Fermi surface with three bands crossing the Fermi level [42].

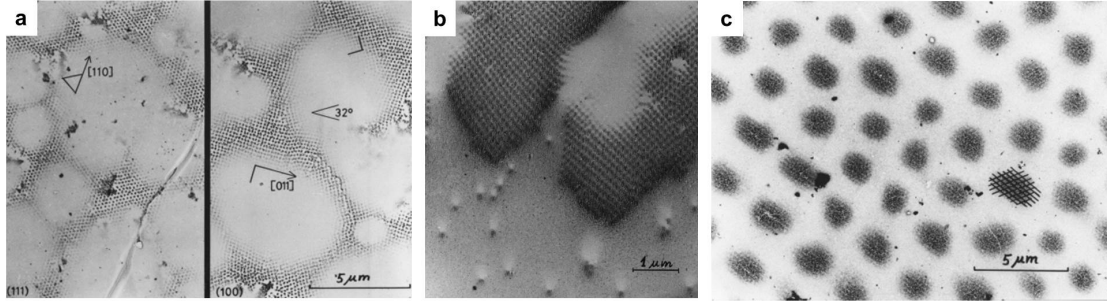


Figure 3.2: Different domains morphologies in the IMS observed in Nb by bitter decoration technique. In a, a high purity Nb disk 1 mm thick, 4 mm diameter, at 1.2 K and 600 Oe ($H_{c1} = 1400$ Oe). Round islands of Meissner phase are surrounded by a regular vortex lattice. In b, a square disk $5 \times 5 \times 1 \text{ mm}^3$ of high purity polycrystalline Nb at 1.2 K and 1100 Oe. Magnetic flux penetrates from the edges in form of fingers which are composed of vortex lattice. In c, high-purity Nb foil 0.16 mm thick at 1.2 K and 173 Oe. It shows round islands of vortex lattice embedded in a Meissner phase. Images adapted from [43, 120].

Recently, the interest in the IMS in low- κ superconductors has been renewed thanks to the advances in the studies of new materials and visualization techniques. SHM experiments have shown vortex chains and clusters in ZrB_{12} ($0.8 > \kappa < 1.12$) at very low fields [1]. SHM measurements showed two different behaviours in ZrB_{12} , one at temperatures close to T_C , characterized by an Abrikosov lattice with a first neighbour distance, $d = (0.75)^{1/4} \left(\frac{\Phi_0}{B}\right)^{1/2}$ and another at lower temperatures characterized by the formation of vortex clusters and stripes with first neighbour distances almost independent of the magnetic field.

Authors in [1] claim that the formation of vortex chains and clusters arises from the combined effect of quenched disorder and the attractive vortex-vortex interaction in the type-II/1 phase at lower temperatures. They observed that at the clusters, non-pinned vortices tend to form the triangular arrangement with pinned vortices at the centre (figure 3.3). The averaged first-neighbour distance inside the cluster exhibits a very weak dependence on the external field (figure 3.3). They associated the regular Abrikosov lattice observed at higher temperatures with the type-II/2 phase dominant at these temperatures.

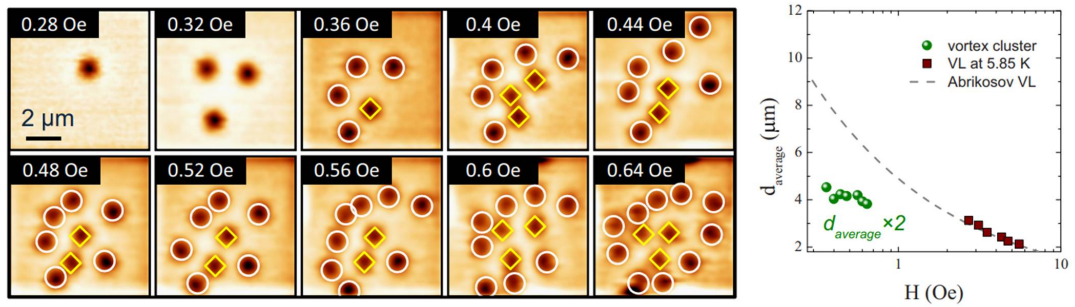


Figure 3.3: Vortex cluster formation in ZrB₁₂. In the left panel, SHM images observed at 4.2 K after FC with progressively increasing magnetic fields, showing the formation of a vortex cluster. The symbols indicate the location of the vortices. Squares points the position of vortices pinned on defects and circles of vortices not pinned on defects. In the right panel, averaged nearest-neighbour distance as a function of the applied magnetic field for the vortex cluster (green circles). The nearest-neighbour distance for the VL at 5.85 K is shown by the squares, which follows the triangular arrangement of the Abrikosov VL (dashed line). Adapted from [1]

On the other hand, 2H-NbSe₂ and MgB₂ show two superconducting gaps with strong interband interactions[121–123] and both are extreme type II superconductors, with $\kappa \gg \frac{1}{\sqrt{2}}$ [124]. Several experiments to characterize the vortex lattice bellow H_{C1} have been done in this two materials using different magnetic microscopic techniques [2–4].

Bitter decoration studies at very low fields, showed a remarkable different behaviour between 2H-NbSe₂ and MgB₂ [2]. Decoration measurements in 2H-NbSe₂ showed a distorted hexagonal lattice (figure 3.4 a) while in MgB₂, they showed vortex accumulation in clusters at $H=1$ Oe and in stripes at $H=5$ Oe (figure 3.4 b and c). Clusters and stripes are separated by vortex free areas, whose size is of a few intervortex distances. Further Scanning squid measurements in MgB₂ showed accumulation of vortices in clusters with an intervortex distance almost independent of the magnetic field [4]. Moreover, SHM measurements at very low fields showed an hexagonal lattice for NbSe₂ with the intervortex distance expected for the applied magnetic field (figure 3.5 a and b) and vortex accumulation in clusters and stripes in MgB₂ [3] (figure 3.5 c and d).

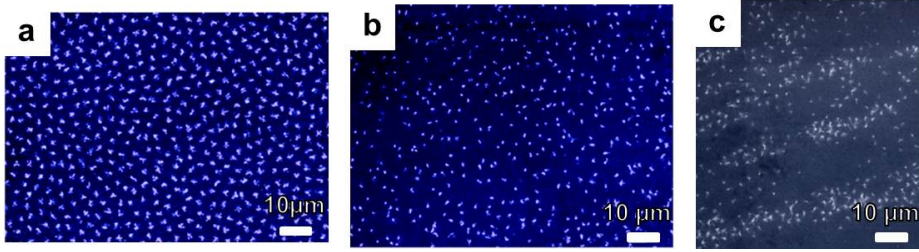


Figure 3.4: Bitter decoration images of the vortex structure at very low fields in 2H-NbSe₂ and MgB₂. Vortex positions are shown as white dots on the blue background. In a, vortices in NbSe₂ in a distorted triangular lattice at T = 4.2 K and H = 1 Oe. In b, vortices in MgB₂ are accumulated in clusters at T = 4.2 K and H = 1 Oe. In c, vortices in MgB₂ are accumulated in stripes at T = 4.2 K and H = 5 Oe. Images adapted form [2].

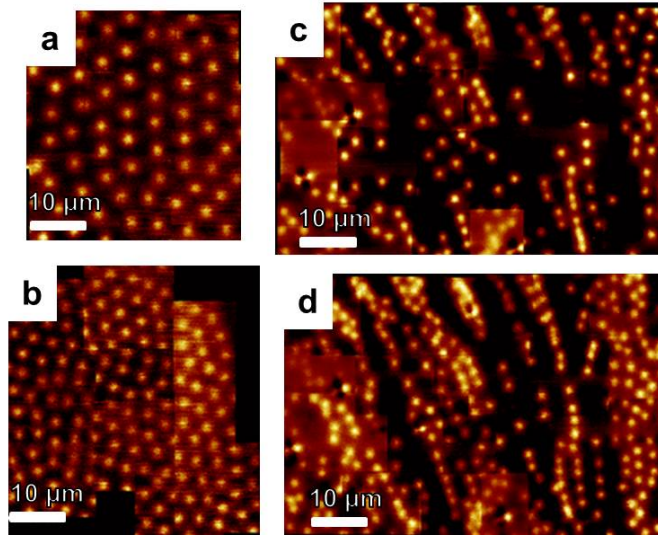


Figure 3.5: Scanning Hall Microscopy images of the vortex structure at very low fields in 2H-NbSe₂ and MgB₂. In a and b, SHM images of a distorted triangular vortex lattice in NbSe₂ at 4.2 K and 2 Oe. In c and d, SHM images of stripes and clusters of vortices in MgB₂ at 4.2K and 2 Oe.

The hexagonal lattice founded in 2H-NbSe₂ was ascribed to vortices nucleated in the mixed state at temperatures where $H_0 = 0$ [3]. Vortices are formed at high temperatures, when $H_{c1}(T)$ is negligible and remain trapped when cooling. Vortices then, form hexagonal lattice with a first neighbour distance, $d = (0.75)^{1/4} \left(\frac{\Phi_0}{B} \right)^{1/2}$ and are retained at low temperatures by surface barriers [125]. No vortex free areas have been reported in 2H-NbSe₂.

Authors in [2–4] propose that the existence of vortex free areas between cluster and stripes in MgB_2 can be explained in term of a new state that they called Type 1.5 superconductivity, based on the semi-Meissner state predicted by [126]. They argue that the two gaps of MgB_2 have different λ and ξ and subsequently different κ , one below $\sqrt{\frac{1}{2}}$ and another above $\sqrt{\frac{1}{2}}$. As a consequence, the vortex-vortex interaction is the result of the competition between short-range repulsion and long-range attraction. This, leads to the appearance of vortex clusters and stripes. They also argue that the vortex stripes are independent of the crystal lattice and therefore they can not be related to pinning due to topographic features.

A recent theoretical work have proposed that the vortex patterns in MgB_2 can be also explained as a result of the interplay between repulsive-attractive vortex-vortex interaction, due to vortex-core deformations and pinning [5].

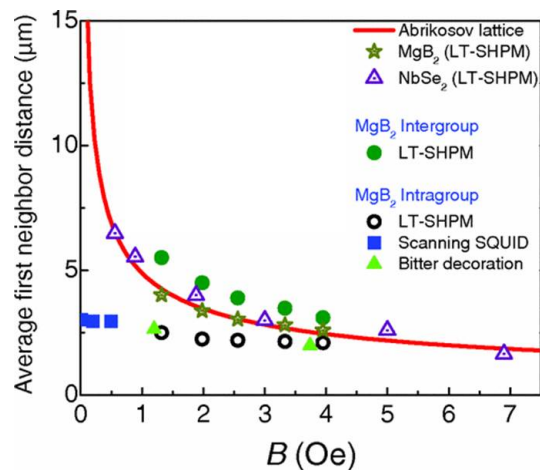


Figure 3.6: Plot of the theoretical and experimental values of the intervortex distances in $NbSe_2$ and MgB_2 obtained by different techniques at very low fields. Intervortex distances in $NbSe_2$ show good agreement with the expected Abrikosov lattice (red line). Vortices in MgB_2 show two different intervortex distances, one intergroup distance which agrees with the expected evolution of the Abrikosov lattice and another intragroup distance which remains almost independent of the magnetic field. Adapted from reference [3].

Figure 3.6 summarizes intervortex distances obtained from the experiments mentioned above in $NbSe_2$ and MgB_2 . In $NbSe_2$, the intervortex distance fits the expected evolution with the applied magnetic field for an Abrikosov lattice. The intervortex distance in MgB_2 is separated in two groups, the intragroup and the intergroup dis-

tances. The intragroup distance remains almost constant when changing the magnetic field while the intergroup distance follows the expected behavior of the Abrikosov lattice [3].

Comparatively, $\beta - Bi_2Pd$ has a small, yet sizable, value of $\kappa \approx 6$. It has very weak pinning and is a single gap isotropic superconductor [6–8].

3.1.2 Previous works on $\beta - Bi_2Pd$ crystals

In 2012 Imai et al. [127] grew $\beta - Bi_2Pd$ single crystals and suggested the possibility of a multigap behaviour. Their macroscopic measurements appeared to be consistent with multigap superconductivity. Their specific heat measurements showed a peculiar behaviour below T_C when changing the temperature, similar to the observed in the two gap superconductors MgB_2 and $Lu_2Fe_3Si_5$ (the jump magnitude at T_C , a fast increase at low-temperature and a small shoulder in between) [128–130] (figure 3.7 b). Moreover, their H_{C2} measurements as a function of temperature were also consistent with the two gap scenario as they showed positive curvature near the critical temperature similar to other multi-gap superconductors [131–134] (figure 3.7 c).

Several works have been done since in order to understand the electronic structure and the gap behaviour of $\beta - Bi_2Pd$. Both, vortex lattice and superconducting gap behaviour studied by STM are actually close to prototypical single gap s-wave behaviour [6]. Single gap was also confirmed by ac calorimetric measurements by [8].

3.1.2.1 STM and specific heat measurements

Recent STM and specific heat measurements performed in $\beta - Bi_2Pd$ single crystals grown in the LBTUAM have probed the single gap behaviour of $\beta - Bi_2Pd$ [6, 8]. STM measurements were done by Dr. Edwind Herrera at the LBTUAM and specific heat measurements were done by the group of Prof. Peter Samuely at the *Centre of Low Temperature Studies* in Slovakia. Both measurements were done in $\beta - Bi_2Pd$ samples of the same series that we have grown and measured in this work (see section

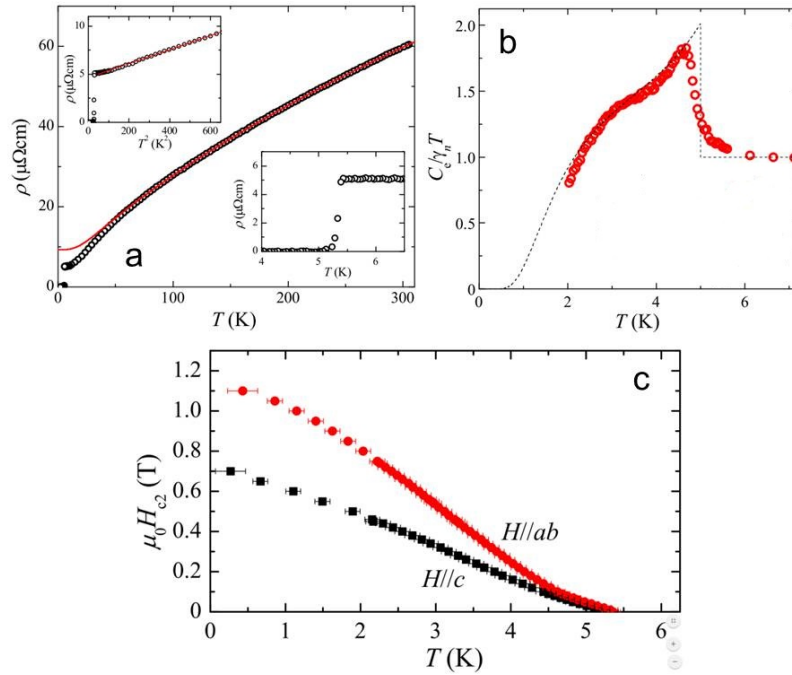


Figure 3.7: In a, the evolution of the resistivity with the temperature for $\beta - Bi_2Pd$ obtained by [127]. Insets show ρ near T_C and ρ at temperatures less than 25 K plotted as a function of T^2 . Black circles represents the experimental data and red line a fit proportional to T^2 . b shows the behaviour at low temperatures of the normalized electronic specific heat in zero field. The red circles are the experimental data and the dashed black curve was calculated using the two-band model [127]. In c, the evolution of the upper critical field with the temperature, also measured by [127]

2.3). In figure 3.8 a the evolution with the temperature of the normalized experimental tunnelling conductance in $\beta - Bi_2Pd$ is presented, together with the evolution of the superconducting GAP with the temperature extracted from it (Figure 3.8 b). Both, conductance measurements and GAP evolution are consistent with a single GAP BCS superconductor with $\Delta = 0.76$ meV [6]. The specific heat represented in figure 3.8 c was obtained using an ac technique [135, 136]. The electronic contribution of the specific heat perfectly fits the BCS single GAP theory [8]. The fit reproduces very well the jump at the anomaly and the shape of the experimental curve. In the data there is not any signature of an additional second GAP.

In a recent STM experiment in epitaxially grown thin films, authors find superconducting properties that are very different from the bulk behaviour [137]. The critical

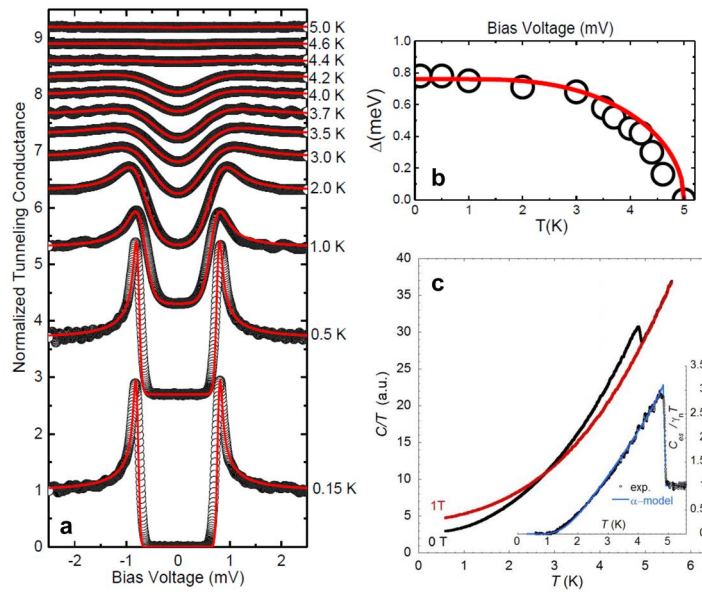


Figure 3.8: In a, the normalized tunnelling conductance curves for $\beta - Bi_2Pd$ at different temperatures are presented. Experimental data (black circles) matches the BCS single GAP fit (red line). The evolution of the superconducting GAP obtained from the curves in a is shown in b (black circles), matching the prototypical BCS single GAP behaviour (red line). In c the evolution of the specific heat with the temperature measured by [8] at 0 and 1 T. The inset shows the normalized electronic specific heat showing the sharp jump in the superconducting transition. The continuous blue line is the theoretical curve based on the BCS theory. Adapted from references [6, 8].

temperature is somewhat larger and two gaps appear in the tunnelling conductance. Furthermore, a zero bias peak appears in the centre of the vortex cores, indicating the formation of vortex bound states [121, 138, 139]. Authors argue that these states could be topologically non-trivial.

3.1.2.2 Fermi Surface

The first calculation of the electronic band structure and Fermi surface (FS) in $\beta - Bi_2Pd$ were made by Shein and Ivanovskii [140] (figure 3.9 a), finding that the Pd 4d and Bi 6p states are responsible from the metallic character of the material. They studied the system with and without spin orbit coupling (SOC) determining that the effect of the SOC is of minor importance. The FS can be divided in four

main structures: a 2D hole-like deformed cylinder parallel to the K_z direction (green colour in 3.9 a), a hole-like pocket centred in the Γ -point, electron-like 3D pockets overlapping the 2D hole like deformed cylinder and one pocket inside the 3D electron like pockets (yellow in 3.9 a) [140]. FS calculations have probed that there are anisotropies of chemical bonding which causes that Bi/Bi layers are less coupled than Bi/Pd layers. This result is also consistent with STM topographic measurements of [6] and with our own result as it will be discussed.

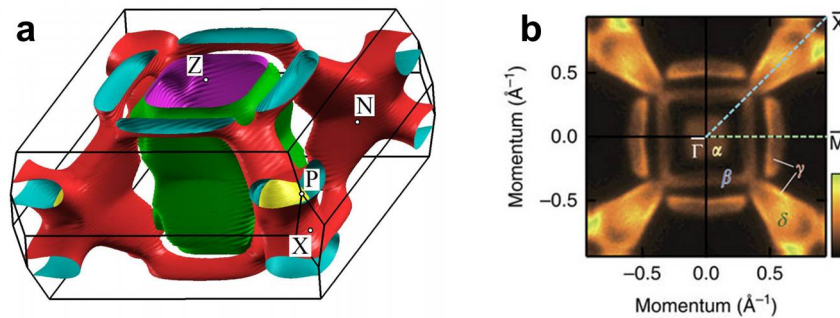


Figure 3.9: In a, the calculated Fermi surface of $\beta - Bi_2Pd$ is shown with the first Brillouin zone [140] and in b, the Fermi surface of $\beta - Bi_2Pd$ recorded by angle-resolved photoemission spectroscopy (ARPES). Two electron-like and two hole-like Fermi surfaces are denoted by α , β and γ , δ , respectively [141].

A later work [141] also have found topological protected states in $\beta - Bi_2Pd$ using angle-resolved photoemission spectroscopy (ARPES). The FS obtained by ARPES mostly agrees with the calculations of [140]. The resulting FS obtained in [141] is presented in figure 3.9 b. It represents the projection in the K_x - K_y plane of the 3D FS. Photoemission reveals a Dirac cone well below the Fermi level[141]. Spin resolved measurements provide polarized bands close to the Dirac cone. The same authors suggest that topologically non-trivial spin polarized bands crossing the Fermi level might rise up to the surface.

3.2 MFM and SOT characterization

Previous works have studied the bulk properties and the vortex lattice at high fields with STM [6, 7]. However, prior to our work, the study of the vortex lattice below and near H_{C1} has not been reported yet. This was our main motivation and the

starting point of our study in this compound.

3.2.1 Topographic characterization

For topographic characterization of the sample, AFM measurements were made in a disk like sample approximately 1 mm thick and 1 cm radius, glued with low temperature silver epoxy to our LT-AFM sample holder and exfoliated at room temperature using scotch tape. Topographic measurements were taken using the dynamic mode described in section 2.1.3.7.1 with typical sample tip separation around 10 nm.

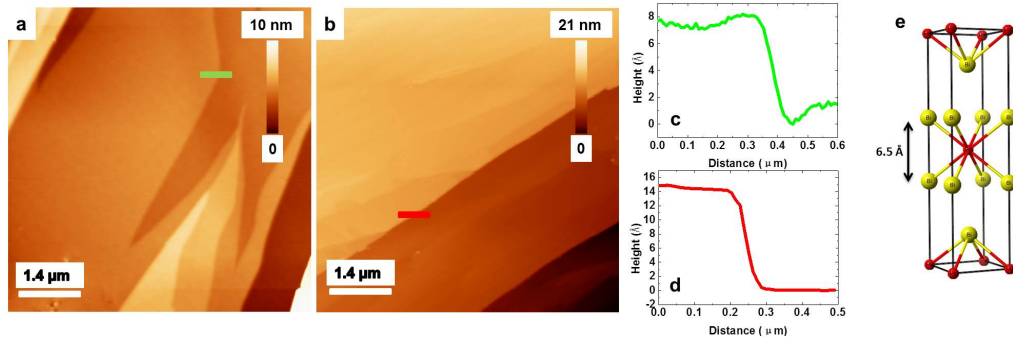


Figure 3.10: Different areas of the $\beta - Bi_2Pd$ crystals after exfoliation with scotch tape at RT. Both images were measured at 2K. In c and d, the topographic profiles corresponding to the green and red lines in the a and b images are shown. Image a has atomic flat areas separated by steps of few Angstroms while b has steps up to tens of nanometres. In e, the unit cell of $\beta - Bi_2Pd$ with the distance between Bi layers highlighted.

After exfoliate it, the sample presents a combination of very clean areas with flat terraces and atomic steps and areas with steps up to some tens of nanometres. The atomic flatness of the surface makes it a very good candidate to SPM measurements including MFM. Figure 3.10 a and b show two $7 \times 7 \mu m^2$ topographic images took at 2 K. In c, the profiles of the green and red lines on the images are shown. The height of the step marked with the green line corresponds with the distance between Bi atoms in the unit cell as it is shown in figure 3.10 e, pointing out that the surface is terminated by Bi as it was observed in STM measurements by [6]. The flatness of the sample allows measuring, at 2K, areas up to $10 \times 10 \mu m^2$ large.

3.2.2 Magnetic characterization

The magnetic profile of the sample was mapped together with its topography using the MFM mode described in section 2.1.3.7.2. For magnetic imaging, the tip-sample separation was kept constant at 120 nm during the scan and the MFM probe magnetized up to 1500 Oe at 10 K.

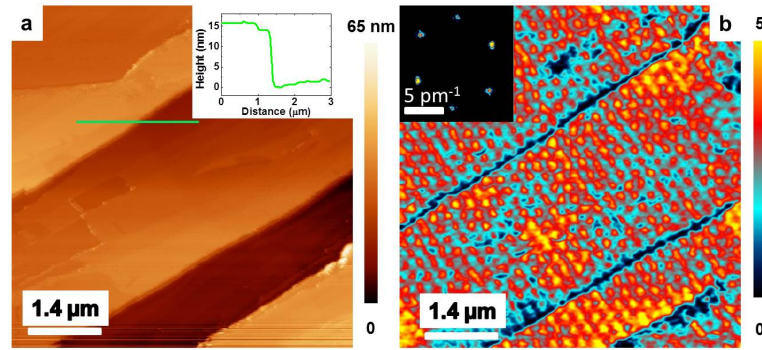


Figure 3.11: In a, we show a topographic image, with a line cut in the inset. Note that the height of the observed steps is of about 10 nm. In b, we show a vortex lattice image taken with MFM at 2K and 300 Oe in the same area, together with its Fourier transform (inset). The lattice is hexagonal over the whole area. The diagonal blue lines in the magnetic image are features due to the long range electrostatic interaction with the steps of the topography.

Fig.3.11 shows simultaneous topographic and magnetic images acquired at 2 K and 300 Oe after Field Cool (FC). The topography shows terraces separated by steps of $\approx 10nm$, produced during the cleaving of the sample. In the simultaneous magnetic signal a vortex lattice is observed over the complete scanned area. We observe a hexagonal vortex lattice over the whole image. Dark blue contrast is also seen in the magnetic images at the position of the topographic steps due to the long range electric interaction between the steps and the MFM tip [101, 142].

3.2.2.1 Evolution of the vortex lattice with the applied magnetic field

We have observed vortices at extremely small applied magnetic fields, using a combination of Squid on the Tip microscopy (SOT) with a set up described in [143] and MFM measurements. SOT measurements were done always in FC conditions by

Dr. Yonathan Anahory and Dr Lior Embon in the group of Prof. Eli Zeldov at the *Weizmann Institute* in Israel with a tip-sample separation of several microns.

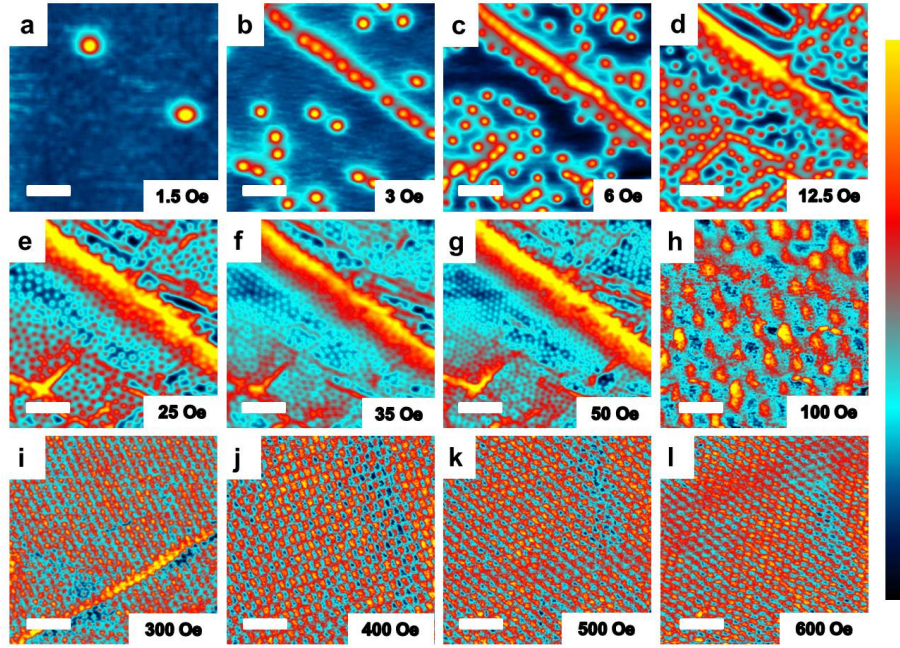


Figure 3.12: Evolution of the vortex distribution in $\beta - Bi_2Pd$ with the applied magnetic field. Images a-g were taken with SOT and present a disordered distribution at the lowest field that become more orderer at higher fields, vortex accumulation in clusters and linear features is also shown. Images from h-l were taken with MFM, a regular Abrikosov lattice is clearly shown in all images. The lattice becomes denser as the magnetic field increases, as expected. The color scale represents the out-of-plane field, with span of 2 in a, 3.5 in b-c, 8.4 in d, 7.2 in e and 7.0 in f-g Oe in the SOT images and of 2 Hz in the MFM images, h-l. The scale bar in white bar is $4\mu m$ in a-g, 560 nm in h and $1\mu m$ in i-l.

The measurements at very low magnetic fields using SOT and at higher fields with MFM provide a radically different behaviour. The vortex lattice is disordered at the lowest fields and becomes gradually more ordered, reaching the hexagonal arrangement for fields close to 100 Oe. Above this field, vortices always arrange in a hexagonal lattice. Selected SOT and MFM images are shown in Figure 3.12. From visual inspection it is clear that, at the lower fields, the vortices are randomly arranged. Upon increasing the field, the flux line lattice becomes gradually more ordered in a hexagonal arrangement, expected for Abrikosov flux line lattice as is clearly seen in the images above 100 Oe.

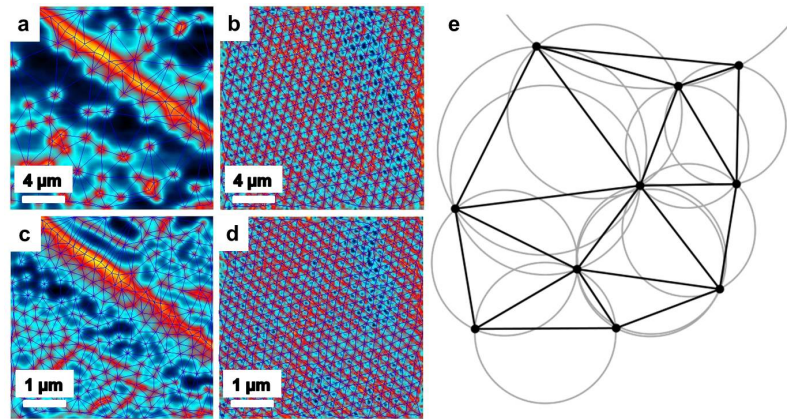


Figure 3.13: Examples of Delaunay triangulated images. Images at the left column were taken with scanning SQUID at low fields (5 Oe and 12.5 Oe) and images in the right column were taken with MFM (300 and 400) Oe. In e, an example of the Delaunay triangulation method. Black dots represents the vortex positions and black straight lines the inter-vortex distances, black circles are the circumcircle corresponding to each triangle formed by three vortices.

To quantitatively describe the vortex distribution, Delaunay triangulation was performed for all SOT and MFM images. In the SOT images, along the defects, above $H \approx 10$ Oe, we do not fully resolve isolated vortices. We have used the small peaks observed in the local magnetic field profile to identify vortex positions. To independently verify that the count is right, we have integrated the magnetic field in the SOT images along the defects and verified that the resulting flux coincides with expected value from the number of vortices we use in the triangulation. A few images are shown in Fig. 3.13 as an example, together with a Delaunay triangulation scheme. A Delaunay triangulation for a set of vortices in a plane is a triangulation such that no vortex in the set is inside the circumcircle of any triangle in the triangulation [144].

The results of the triangulation are presented in figure 3.14. The colour map correspond to the probability of the inter-vortex distances extracted from the Delaunay Analysis. For fields below ~ 100 Oe, the histogram broadens and the distances between vortices become wide-spread. At very low magnetic fields we can observe inter-vortex distances ranging between half and twice the expected inter-vortex distance for a hexagonal lattice ($d = (0.75)^{1/4} \left(\frac{\Phi_0}{B}\right)^{1/2}$). Image inspection shows the strong spread in distances is due to location of vortices at lines, with the formation of vortex chains. This accumulation leaves vortex-free areas in between. Interestingly,

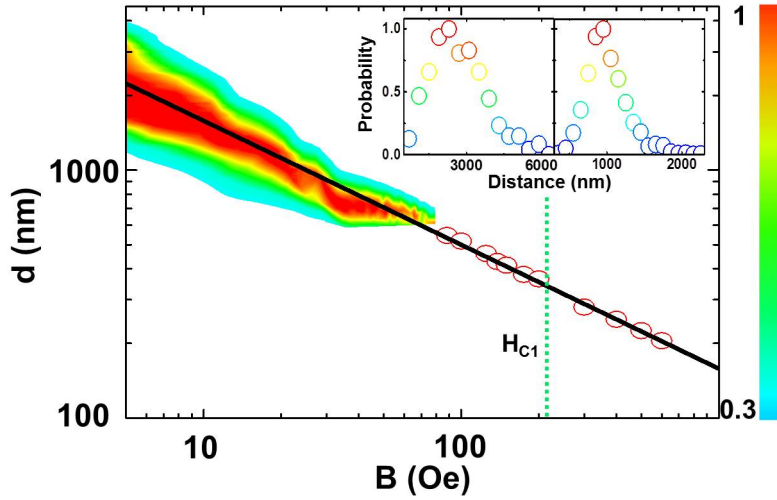


Figure 3.14: Intervortex distances vs the applied magnetic field. The black line represents the expected intervortex distance ($d = 1.075\sqrt{\frac{\phi_0}{B}}$), the red circles the measured intervortex distances with the MFM and the colour map the distribution of probabilities of intervortex distances obtained with the triangulation of the SOT images. Two different regimes are found, one at fields bellow $H_{C1} \approx 100$ Oe, where the vortex distribution is wide spread and other above $H_{C1} \approx 100$ Oe, where the vortices are ordered forming the Abrikosov lattice. The lower critical field measured by [8] is represented by the green dotted line. Colour scale is as represented by the bar at the right. In the insets, the histograms obtained for 3 and 25 Oe.

the histogram is peaked at the expected intervortex distance d , although it is skewed at large distances, reflecting that pinning is limited by intervortex repulsion. It is important to note that the vortices arrange in an hexagonal lattice well bellow the H_{C1} of the sample, 225 Oe [8].

3.2.2.2 Penetration depth at the defects

Simultaneous topographic images were not recorded in SOT measurements and we can not unambiguously determine that the magnetic features are related with topographic steps, despite is the most probable scenario. Vortices located at defects give weaker spots in the SOT images. The value of the magnetic field at the vortex centre is smaller than the value we find for vortices located far from the defect. This is nicely

visible at lower fields when vortices are well separated and do not overlap, (see figure 3.15 b), where the vortices arranged in a chain like structure, present weaker spots that the ones far from the chain. There are a few vortices in the SOT images that are not arranged along the main line and also show weaker spots, for example the one marked by an yellow arrow in figure 3.15 c. At higher magnetic fields, vortices cluster along lines close to these positions are found (figure 3.15 d). These vortices are thus also located close to a defect.

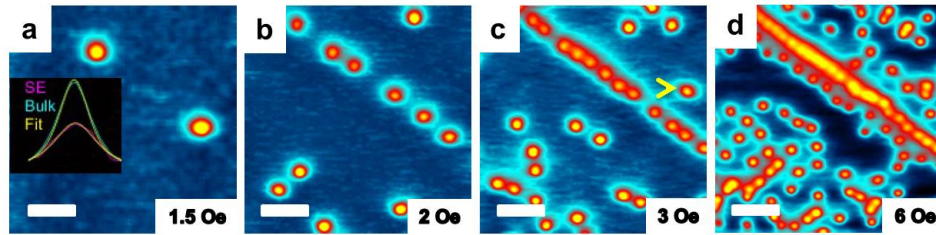


Figure 3.15: Selected images of SOT in $\beta - Bi_2Pd$. In a, two vortices on a defect. In b, c and d the evolution of the same area when increasing magnetic field. Vortices on the defects have weaker spots. In the inset the profiles of two vortices, one on the defect and another far from it together with the fitting described in the text. In c, the yellow arrow points a vortex far from the stripe which also shows a weaker spot. Visual inspection of d, shows that there is a vortex cluster at this position at higher fields, pointing out that there is also a defect at this position. 2 (a), 3.5 (b-d). The scale bar is white is $4\mu m$

In the inset of Fig. 3.15 a we show magnetic field profiles along two vortices showing weak and bright spots respectively. We can fit the profile of the isolated vortex to a monopole located at a distance $\lambda + d_{SOT}$ from the SOT where λ is the penetration depth and d_{SOT} is the distance from the tip to the sample surface. We estimate the value of d_{SOT} by taking $\lambda = 132 \pm 20$ nm [8] for the bright spots. We then leave the penetration depth as a free parameter to fit the profile of the weak spots and find $\lambda_D = 270 \pm 40$ nm which is about two times the value found elsewhere.

3.2.3 Origin of the variation in λ

The dependence of λ and ξ with the strain produces an effective interaction between the crystal and the vortex lattices [145]. Also, the stress produced by flux pinning

has been proposed as a source of magnetostriction effects in superconductors [146]. A recent theoretical work has demonstrated that strain can induce a square vortex lattice in the tetragonal superconductors [147]. The coupling between crystalline elasticity and the vortex lattice can be treated using the dependants of the critical temperature with the pressure dT_c/dP [145, 148–150]. Generally, vortices are repelled from places where the internal strain is bigger if $dT_c/dP > 0$ and are attracted to those places if $dT_c/dP < 0$. The value of dT_c/dP in $\beta - \text{Bi}_2\text{Pd}$ is unknown and therefore we can not unambiguously prove that the vortex accumulation at the defects in our crystals are due to strain effects, but it is known that the non-centrosymmetric α - Bi_2Pd crystallizes at 3.8 K, 1.2K below the β - Bi_2Pd . At low magnetic fields, we find that vortices are accumulated along defects, which is compatible with $dT_c/dP < 0$.

Let us remark that the pressure dependence of T_c and of H_{c2} in MgB_2 is sizeable, so that one can also in principle expect some coupling between strain and the vortex lattice[151].

3.2.4 Origin of the flux landscape

All our experiments are in field cooled conditions, so we quench, during cooling, vortices at locations where the free energy landscape is more favourable [116–118, 125]. For the lowest magnetic fields, we find strong gradients in the vortex distribution. To analyse this further, we calculate the elastic energy associated to pairs of vortices, F , at different locations in our images. We compare the result for vortices located at a defect and giving weak spots in SOT with the elastic energy for pairs of vortices far from the defects. To this end, we use $F = \frac{\phi_0^2}{4\pi\mu_0\lambda^2} \log(\kappa) + \frac{\phi_0^2}{4\pi\mu_0\lambda^2} K_0(d/\lambda)$ for the free energy per unit length of two vortices interacting with each other at a distance d [29]. The first term comes from the energy of superfluid currents, giving the line tension of the vortex, and the second term the interaction energy between vortices. K_0 is the modified Bessel function of the second kind. We then calculate F for vortices far from defects using the bulk λ and for vortices at the defects we use the measured λ_D . Below $\approx 50Oe$ the intervortex distances vary from 0.5 to 4 μm and the second term of the interaction energy remains negligible with respect to the first term, giving a difference in free energy between both situations of $\Delta F \approx 2 \times 10^{-11}$ independent of

the intervortex distance.

We can now compare ΔF with the pinning energy of a vortex pinned at a normal inclusion in β - Bi_2Pd using $U_{core} = \frac{B_c^2 \pi \xi^2}{\mu_0}$ [152], with B_c the thermodynamic critical field and ξ the superconducting coherence length ($\xi \approx 25$ nm, [6, 153]). We find a value which is smaller than ΔF , $1 \times 10^{-11} Jm^{-1}$. Thus, single vortex pinning at defects play an unimportant role in the vortex distribution on $\beta - Bi_2Pd$. Moreover, it is difficult to think of normal inclusions as big as $\xi \approx 25$ nm to pin isolated vortices. Thus, pinning seems to play a minor role in β - Bi_2Pd at low fields. The accumulation of vortices at the defects can be explained with the lower free energy caused by the experimentally determined changes in λ .

It is quite remarkable that simple estimation provide such clear results, and are probably particularly valid when vortices are very far apart at the lowest magnetic fields we have studied. For higher magnetic fields, the vortex lattice density increases and the previous two-vortex interaction approximation is no longer valid. For fields above ≈ 50 Oe we have consider the free energy of a vortex, interacting with his first six neighbours arranged in a hexagonal lattice using: $F = n \frac{\phi_0^2}{4\pi\mu_0\lambda^2} \log(\kappa) + 3n \frac{\phi_0^2}{4\pi\mu_0\lambda^2} K_0(a/\lambda)$ [29], where a is the lattice parameter of the Abrikosov lattice determined by $a = (0.75)^{1/4} \left(\frac{\Phi_0}{B}\right)^{1/2}$ and n the number of vortices per unit area. The difference of energy between six vortices close to a defect with λ_D and six vortices far from the defect with λ changes with the intervortex distance. We find that when vortices at the defect are closer than about 400 nm, it is no longer energetically favourable to add new vortices to the defect. This can explain the vortex distribution at low fields shown in Fig. 3.14. We find that the cross-over field is of ≈ 200 Oe. In the experiment, we find that already at $\approx 100Oe$ the distance histogram closes and the vortex lattice becomes hexagonal. We believe that, given the crude approximations made, the agreement is remarkable and provides a simple but successfully explanation to the vortex landscape at low fields.

It is also noteworthy that the average value of the intervortex distances follows d for all magnetic fields. This finding has not been previously reported, to our knowledge, at low magnetic fields and in presence of strongly inhomogeneous vortex distributions. Instead the usual pictures consist of clusters of vortices with widely differing

intervortex distances that are often smaller than d .

At the beginning of this chapter, we have introduced previous measurements in MgB_2 where vortex clusters, stripes and vortex free areas in between were found. Authors of [2–4] point out that the attractive-repulsive vortex-vortex interaction in the Type 1.5 superconducting state due to the existence of multiple superconducting gaps can explain this vortex distribution. Results in $NbSe_2$ are by contrast quite different. A distorted hexagonal lattice was found at very low fields with first neighbour distance following $d = (0.75)^{1/4} \left(\frac{\Phi_0}{B}\right)^{1/2}$. This was explained as trapped vortices that remains at the sample after FC bellow H_{C1} due to surface barriers [2, 3].

The vortex patterns that we have measured in β - Bi_2Pd showed both behaviours. At very low fields, the patterns are similar to the ones described for MgB_2 with vortex stripes, clusters and vortex free regions. At higher magnetic fields, still below H_{c1} , we observe by contrast a hexagonal, defect free, vortex lattice.

β - Bi_2Pd is clearly a single gap superconductor, so vortex clustering cannot be associated to multiple gap opening here [6, 8]. Thus, the vortex patterns at very low fields in $\beta - Bi_2Pd$ are due to the distortion of the hexagonal lattice formed during the FC process by pinning at crystalline defects due to differences in the values of the penetration depth. At very low fields, the vortex-vortex repulsion potential is small, and pinning dominates, leading to the observed vortex clustering. At higher fields, the vortex-vortex repulsion potential impedes pinning of isolated vortices and the vortex lattice arranges as a whole in a regular hexagonal lattice.

3.2.4.1 Evolution of the vortex lattice with the temperature

The penetration depth is expected to increase as the temperature approaches T_C , therefore, the superconducting vortex profile will be broader as the temperature increases. Eventually, at T_c , the magnetic profile will become homogeneous over the hole surface of the superconductor [29, 154].

We have measured the magnetic profile of the same area at different temperatures and constant magnetic field. After cool down the sample to 2.75 K in a perpendicular

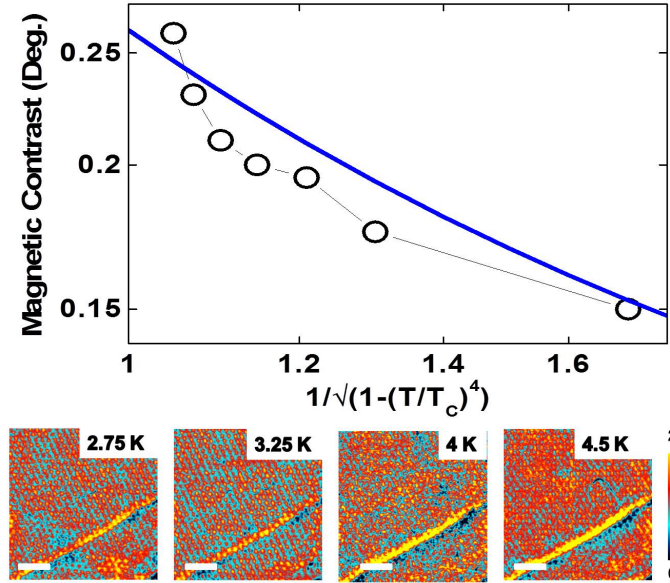


Figure 3.16: Evolution of the vortex lattice with the temperature. In the upper panel, a plot of the evolution of the magnetic contrast with the temperature is shown. The experimental data (black circles) was obtained from MFM images measured under a perpendicular magnetic field of 300 Oe at the same area. The blue line represents the expected magnetic contrast due to the theoretical evolution of λ as explained in the text. Both curves have the same behaviour, proving that the evolution of the vortex width is dominated by the evolution of λ . In the lower panel, four of the MFM images used to obtain the plot. Scale bar in white is $1\mu\text{m}$.

magnetic field of 300 Oe, the temperature was increased to 3, 3.25, 3.5, 4, 4.5 and 5 K recording the magnetic profile at each temperature. The evolution of the vortex profile width with the temperature is presented in figure 3.16. Experimental data was obtained as the difference between the MFM signal at the centre of the vortices and the background between vortices. Vortices magnetic profile become broader as the temperature increases. This can be related to the natural increase in the penetration depth, $\lambda(T) = \frac{\lambda(T=0)}{1-(T/T_C)^4}$, that characterizes the decay of the magnetic field outside of the vortex core. In the image, the relation between the experimental data and the theoretical evolution of the penetration depth is clear. We have calculated the theoretical value of the contrast as the difference of the magnetic field at the centre of a vortex and in between its neighbours in an hexagonal lattice using equation [?]. At 5 K, no vortex lattice was found in the sample, in agreement with the resistive

T_C [6].

In our measurements we have not founded any evidence of vortex clustering or lattice distortion at the lower temperatures associated to the attractive interaction between vortices in the type II/1 regime reported in ZrB_{12} [1].

3.2.4.2 Orientation of the vortex lattice

Previous works in the same $\beta - Bi_2Pd$ crystals have found that at high fields, the hexagonal vortex lattice is oriented with one of its axis along a crystallographic direction [6]. AFM has no atomic resolution and therefore, it can not determine the directions of the crystals lattice, but they can be inferred from the direction of the atomic steps that are easily measured with an AFM.

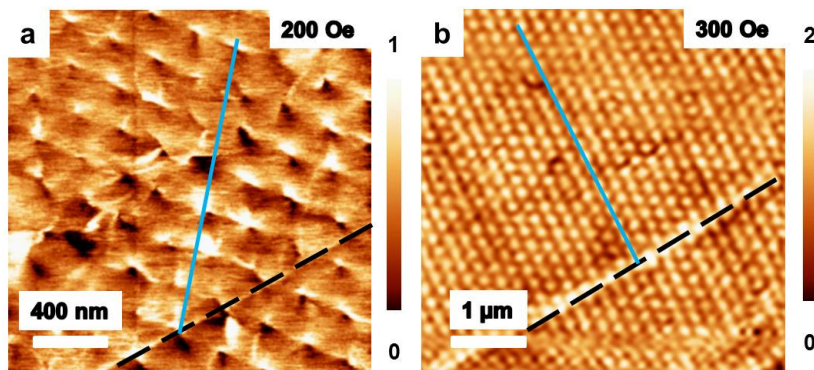


Figure 3.17: Example of the two vortex lattice orientations found. In a, the vortex lattice at 200 Oe form $\approx 30^\circ$ with respect to the topographic step (dashed black line in the image). In b, at 300 Oe, one of the main directions of the vortex lattice is normal to the topographic step. In both images the direction of the vortex lattice os highlighted with a light blue line.

Here, we have found that at low magnetic fields, bellow $\approx 200Oe$, the vortex lattice is preferentially oriented with one axis following the steps. It can be related to the accumulation of vortices at the defects due to the differences in penetration depth. At higher magnetic fields, when the distances are practically homogeneous over the whole field of view, the vortex lattice is oriented at 90° to the defects (figure 3.17). This hints that another mechanism, possibly the interaction between the hexagonal vortex lattice and the tetragonal crystalline symmetry observed at higher magnetic

fields [6], is fixing the orientation of the vortex lattice.

3.3 Summary and conclusions

In conclusion, we have observed two different regimes in the patterns of pinned superconducting vortices at low magnetic fields in the single gap superconductor β -Bi₂Pd. We have shown that lines of vortices form at defects due to pinning at very low magnetic fields, while at higher fields the vortex lattice acts as a whole, leaving a regular hexagonal lattice even below H_{c1} . Crystalline strain close to defects determines the vortex arrangements at low fields and leads to sizeable modifications of the local superconducting screening properties, as shown by the measured increase in the penetration depth λ close to defects. The mutual influence of crystalline strain and the vortex lattice has been extensively studied at high magnetic fields. Here, we show that this mutual influence also modifies vortex arrangements at very low magnetic fields. At fields slightly above H_{C1} , where vortices are arranged in a regular Abrikosov lattice, we have found that the magnetic profile of the vortices follow the expected behaviour when changing the temperature. We have not found any evidence of vortex clustering when decreasing the temperature associated to Type II/1 superconductivity in ZrB₁₂ [1].

Manipulation of the crossing lattice in *Bi₂Sr₂CaCu₂O₈*

4.1 Introduction

Bismuth strontium calcium copper oxide, or BSCCO, is a family of high-temperature superconductors having the generalized chemical formula $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$, with $n = 2$ being the most commonly studied compound, also called Bi-2212. Discovered in 1988 [155], BSCCO was the first high-temperature superconductor which did not contain a rare earth element. It is a cuprate superconductor, an important category of high-temperature superconductors sharing a two-dimensional layered (perovskite) structure with superconductivity taking place in the copper oxide planes.

The crossing lattice of Josephson vortices (JVs) and pancake vortices (PVs) in Bi-2212 have attracted a lot of attention in the scientific community in the last decades. Theoretical works have described the interaction between PVs and JVs at different regimes [47, 49, 50, 52, 58] and experimentalist have imaged the crossing lattice by several techniques like magneto optical (MO) imaging [63–65] (figure 4.1 a), bitter decoration [61, 62] (figure 4.1 b), and Hall microscopy [66–68] (figure 4.1 c).

Those works have characterized the crossing lattice at different polar angles and strengths of the applied magnetic field. They have found good agreement between

theory and the experimental values of the lattice parameters of the JV lattice and the distribution of PVs on top of JVs [66, 67]. They have also achieved PVs manipulation in some extent [68]. But, the local manipulation of single PVs and the experimental measurement of the force between PVs and JVs has not been achieved yet. On the other hand, local manipulation of single vortices was achieved in YBCO, another anisotropic high- T_C superconductor. Authors in [84, 91] have demonstrated that MFM allow to interact with the vortex lattice, making possible to manipulate individual or groups of vortices to study their dynamics and interactions.

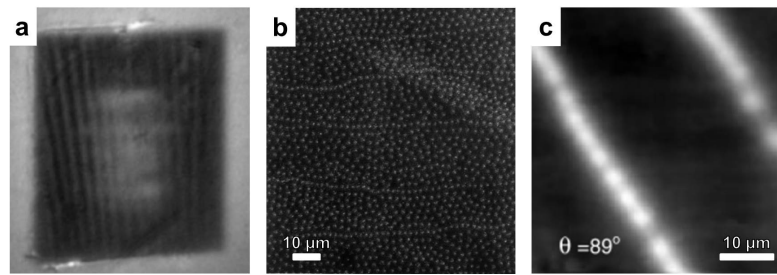


Figure 4.1: In a, a MO image measured in a Bi-2212 single crystal showing vortex chains. Obtained at $T = 72$ K, $B_{\perp} = 13.8$ Oe and $B_{\parallel} = 60$ Oe. In b, bitter decoration image in a Bi-2212 single crystal showing vortex chains with vortices in between. Obtained at $T = 72$ K, $B_{\perp} = 12$ Oe and $B_{\parallel} = 32$ Oe. In c, a SHM image of PV chains in a Bi-2212 single crystal at $T = 81$ K, $B_{\perp} = 0.8$ Oe and $B_{\parallel} = 35$ Oe. In the three images, vortices appear as white dots on the black background. Adapted from [61, 65] and [59].

4.1.1 Interaction between JVs and PVs

At small fields and high anisotropy factor, γ , PVs do not influence much the structure of JVs. However, there is a finite interaction energy between PV stacks and JVs (crossing energy) due to the PVs displacements under the action of the JVs in-plane currents [49]. This interaction causes an effective attractive force between PVs and JVs. The force per unit length along the c -axis, between a PV stack and a JV stack was calculated by [49] as:

$$f_x = \frac{1.4\phi_0^2}{4\pi^2 a_z \gamma^3 s^2 \log(\lambda_{ab}/s)} \quad (4.1)$$

Where ϕ_0 is the quantum of flux, a_z the lattice parameter of the JV lattice in the c-axis direction, s the distance between superconducting layers and λ_{ab} the penetration depth for superconducting currents in the a-b plane.

4.1.2 Manipulation of the crossing lattice in Bi-2212

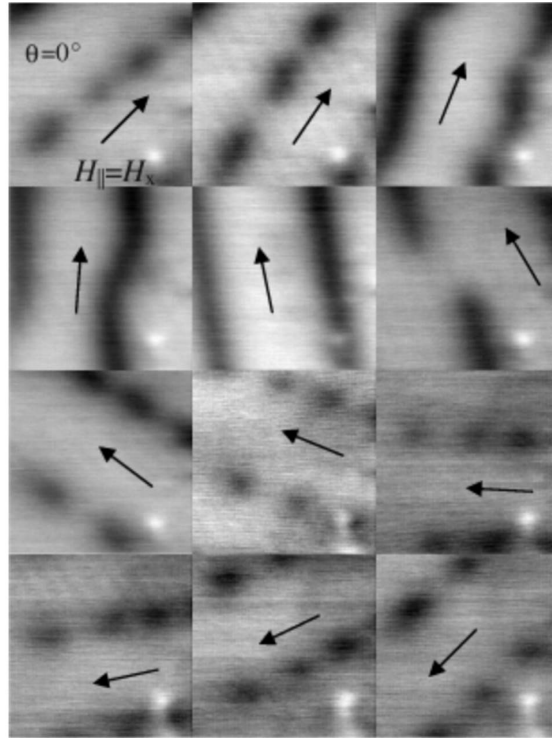


Figure 4.2: SHM images at $T = 80$ K and $H_{\parallel} = 27.5$ Oe, with the JV lattice rotated by (left to right, top to bottom, anticlockwise rotation): 0° , 15° , 30° , 45° , 60° , 75° , 105° , 120° , 135° , 150° , 165° , and 180° . PVs appear as black dots in the grey background. The black arrow indicates the direction of the magnetic field Scan size $28 \mu\text{m} \times 28 \mu\text{m}$. Adapted from [68].

Previous works have been able to manipulate the crossing lattice in Bi-2212,. They were able to drag PVs with the JVs by changing the direction of the in-plane magnetic field at temperatures close to T_C [59, 68]. An example is presented in figure 4.2, where the crossing lattice was successfully rotated in Bi-2212 at 80 K.

Authors in [68] argue that the attraction force between JVs and PVs at 80 K in their crystals is three times larger than the pinning force of the PVs. Therefore changing the direction of the JVs by rotating the in-plane the magnetic field, drags

the PVs with them.

4.1.3 Manipulations of superconducting vortices with MFM

4.1.3.1 Force of a MFM tip on a vortex

MFM tips exerts a given force on magnetic samples, this fact is often a disadvantage, as the tip-sample interaction could change the magnetic state of the sample and somehow introduce artefacts in the measurement [102–104, 156]. In the current chapter, we deliberately have used this force to manipulate superconducting vortices. We have magnetized the tip parallel to the vortices to give attraction force between them. Such a force, will decrease as the tip-sample separation increases.

To obtain an insight of the force acting on the vortices, we have followed the calculation of the tip-vortex interaction made by [157]. This model, treats both, tip and vortex, as monopoles. The model assumes that the tip is and infinitely long and narrow cylinder with its mains axis and magnetization parallel to the Z axis and the vortex as a monopole residing at a distance λ bellow the surface of the superconductor, which fills the half space $z < 0$ with a magnetic field [158, 159]:

$$\vec{B}(\vec{r}, z) \approx \frac{\phi_0(\vec{r} + (z + \lambda)\hat{z})}{2\pi(R^2 + (z + \lambda)^2)^{3/2}} \quad (4.2)$$

Where r is the radial distance from the tip, z the vertical distance and R the radius of the tip. Thus, the force acting on the tip due to the interaction with the superconducting vortex is:

$$\vec{F}(\vec{r}, z) \approx m\vec{B}(\vec{r}, z) \quad (4.3)$$

where m is the dipolar moment per unit length of the tip. Maximizing the force of equation 4.3 in z , we obtain:

$$F_z^{max} = \frac{m\phi_0}{2\pi(z+h_0)^2} \quad (4.4)$$

where h_0 is the offset in the tip-sample separation due to the approximation of the monopole model. The maximum lateral force is approximated as:

$$F_{lat}^{max} = \alpha F_z^{max} \quad (4.5)$$

Here, α is a constant of proportionality with a value between 0.3 and 0.4 [158, 160].

4.1.3.2 Vortex manipulation in YBCO

Previous measurements have probed the possibility of drag vortices in the High- T_C superconductors, YBCO [84, 91]. In particular, the authors in [84] have measured the interaction of a moving vortex with the local disorder potential. They found an unexpected and marked enhancement of the response of a vortex to pulling when they wiggled it transversely. They probed that wiggling the vortex along the fast scan of the MFM image, allows to move the vortex along the slow axis of the image when the magnetic tip is near enough to the sample and therefore the magnetic force between tip and sample increases. A schematic representation of the process is presented in figure 4.3 together with real MFM images of stationary and dragged vortices.

4.2 AFM/MFM studies

We have presented how previous works have manipulated individual vortices in YBCO and groups of PVs trapped on JVs in Bi-2212 at temperatures close to T_C . But, prior to our work, individual manipulation of PVs on JV, i.e. manipulation of the superconducting phase on a JV, has not been studied yet. Moreover, the force to move a PV out a JV remains unknown. This was our main motivation and the starting point of our study in this compound.

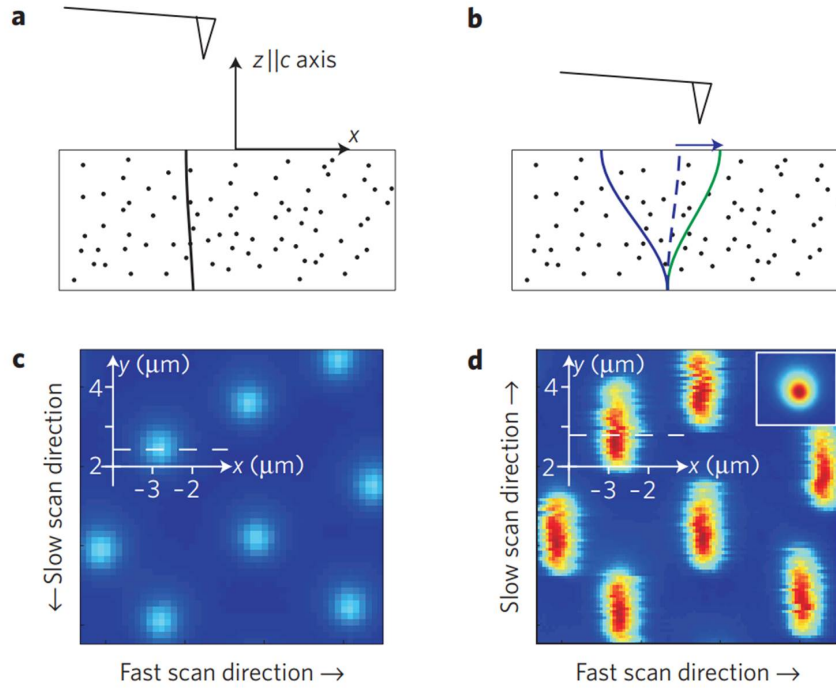


Figure 4.3: Scheme of vortex movement by MFM. The MFM tip (triangles) attracts a vortex (thick lines) in a sample with random pinning sites (dots). In a, the applied force F_{lat} is too weak to move the vortex due to the big tip-sample distance. In b, the vortex moves right, as the tip rasters over it in the direction indicated by the arrow. The blue line illustrates the initial vortex position, the dashed blue line shows an intermediate position and the green line shows the final configuration. In c and d, MFM scans for two different scan heights, $z = 420$ nm ($F_{lat}^{max} \approx 6$ pN), not enough to perturb vortices (c) and $z = 170$ nm ($F_{lat}^{max} \approx 12$ pN), enough to drag the vortices (d). Inset: Scan at 5.2 K, showing a stationary vortex. Adapted from [84].

We make several manipulation experiments. First, we show that PVs can be displaced by exciting them with the tip motion and turning the magnetic field. Then, we show how the tip motion can move PVs from one JV to another. We have also crossed JVs after induce a JV lattice at an angle with respect to a strongly pinned JV. Finally, we have studied the PVs entry in the sample at low temperatures, determining that it is governed by pinning. Our experiments show that phase patterns in superconductors, even when these are strongly pinned, can be controlled by the action of small forces and the direction of the magnetic field.

AFM/MFM measurements were performed in the set up of our lab described in

section 2.1 in a Bi-2212 single crystal. The crystal was grown by the group of Prof. Kadowaki at the *University of Tsukuba* in Japan, following a procedure similar to the one described in [161]. It has a superconducting critical temperature of ≈ 88 K.

4.2.1 Topographic characterization

For topographic characterization of the sample, AFM measurements were made in a Bi-2212 single crystal glued with low temperature silver epoxy to our LT-AFM sample holder and exfoliated at room temperature using scotch tape. The cleaving of the surface occur in the c-axis [162, 163]. We have aligned the crystal with the main axis of our coil system, to apply the Z component of the magnetic field along the c axis and B_x and B_y along the in-plane crystalline axis. Topographic measurements were taken using the dynamic mode described in section 2.1.3.7.1 with typical sample tip separation around 10 nm.

After exfoliate it, the sample presents very clean areas with flat terraces and atomic steps. The atomic flatness of the surface have allowed us to measure areas of $\approx 10 \times 10 \mu\text{m}^2$ at 5 K. An example of the crystal surface is presented in figure 4.4 together with the unit cell and a plot of evolution of the magnetization with the temperature near the superconducting transition.

4.2.2 Obtaining the Crossing Lattice

We have imaged the crossing lattice in our Bi-2212 single crystal using the MFM mode described in 2.1.3.7.2. First, we have cooled the sample under an applied magnetic field of 30 Oe in the Z direction down to 5.3 K and measured the resulting PV distribution. We obtained the regular Abrikosov lattice with the intervortex distance expected for the applied magnetic field (figure 4.5).

After measure the Abrikosov lattice, we ramped the field in Z down to zero and applied a magnetic field of 200 Oe in the Y direction. We obtained images as shown in Fig.4.6 a. The Abrikosov lattice is interspersed with lines of PVs pinned on JVs.

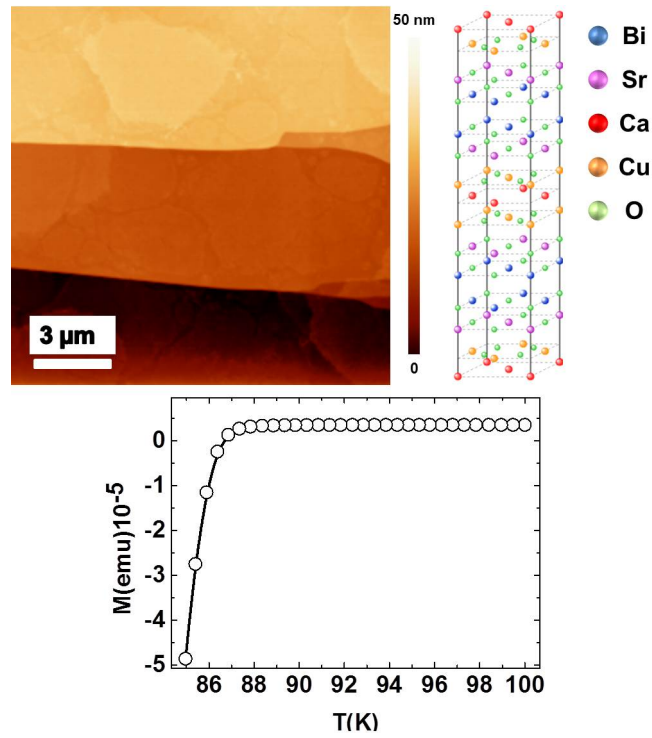


Figure 4.4: In the upper panel, in the left, an AFM topographic image of the cleaved Bi-2212 single crystal and its unit cell (right). The topography shows atomically flat terraces with steps ≈ 15 nm high. In the lower panel, the evolution of the magnetization of the same Bi-2212 crystal with the temperature near $T_C \approx 88$ K. The line is a guide to the eye.

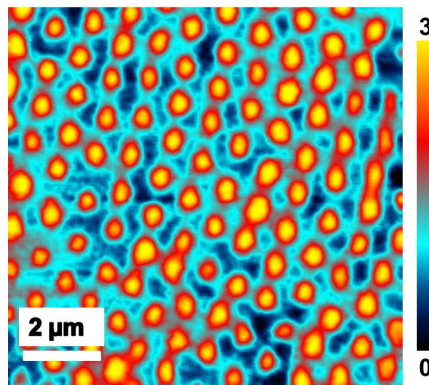


Figure 4.5: MFM image of the regular Abrikosov lattice. Measured in the Bi-2212 single crystal after FC at 5.3 K with a perpendicular magnetic field of 30 Oe.

To eliminate as far as possible PVs, we heated the sample quickly above 70 K and cooled it again to 2 K. This freed the PVs from their pinned positions and more JVs decorated with PVs are visible (figure 4.6 b). After repeating this process several

times we have obtained areas with almost every PV pinned on top of a JV (figure 4.6 c). The same process was always used to obtain the decorated JVs in the following sections.

Images in figure 4.6 were used to calculate the anisotropic factor of our Bi-2212 crystal (for the details of the calculation see chapter 1). Using the distance between JVs, we have calculated $\gamma = 250$ and $a_z = 15.4$ nm. The size of the JVs was also calculated using the relation $a_{JV} = \gamma s$ and $b_{JV} = s$ [51], finding $a_{JV} = 375\text{nm}$ and $b_{JV} = 1.5\text{nm}$.

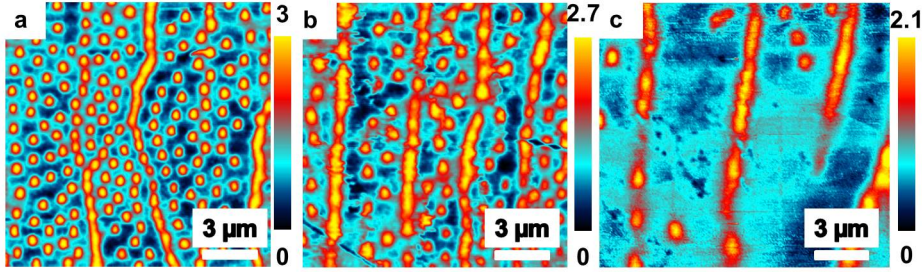


Figure 4.6: Decorated JVs with PVs. Images obtained with $B_{\parallel} = 200$ Oe along the Y direction, at 5.3 K. In a, the vortex arrangement after FC with $B_{\perp} = 30$ Oe and turn it to zero and B_{\parallel} to 200 Oe at 5.3 K. Some decorated JVs are visible with a significant number of trapped PVs in between. In b, a different configuration after heat the sample up to 70 K and cool it down again to 5.3 K. More decorated JVs are visible and the number of PVs in between have decreased. After repeat the same process several times, almost all the PVs are decorating JVs (c). The field of view has moved during heating, so that the images are not taken at exactly the same position.

The penetration depth, λ_{ab} , of Bi-2212 single crystals was experimentally determined by previous work using different methods [164–167]. This works have reported values between 180-270 nm for λ_{ab} . Thus, the lateral size of the PVs and the JVs is comparable. Therefore, only one PV row fits inside a JV.

4.2.3 Evolution of the crossing lattice with the temperature

We have measured the evolution of the crossing lattice at the same area for different temperatures. Images in figure 4.7 were measured at 5.5 K, 12 K and 15.5 K.

They show that the vortices width increases with the temperature. At 20 K the flux distribution becomes homogeneous over the surface and no magnetic contrast was obtained. This, suggest that PVs are able to move far enough from their equilibrium position to overlap between them. Figure 4.7 also represents the evolution of the magnetic profile of the same vortex at different temperatures, it shows that the magnetic contrast decreases as the temperatures increases. The potential well of the vortices was extracted from the evolution of this magnetic profile. It represents the thermal energy associated to each temperature of the experiment versus the vortex profile width.

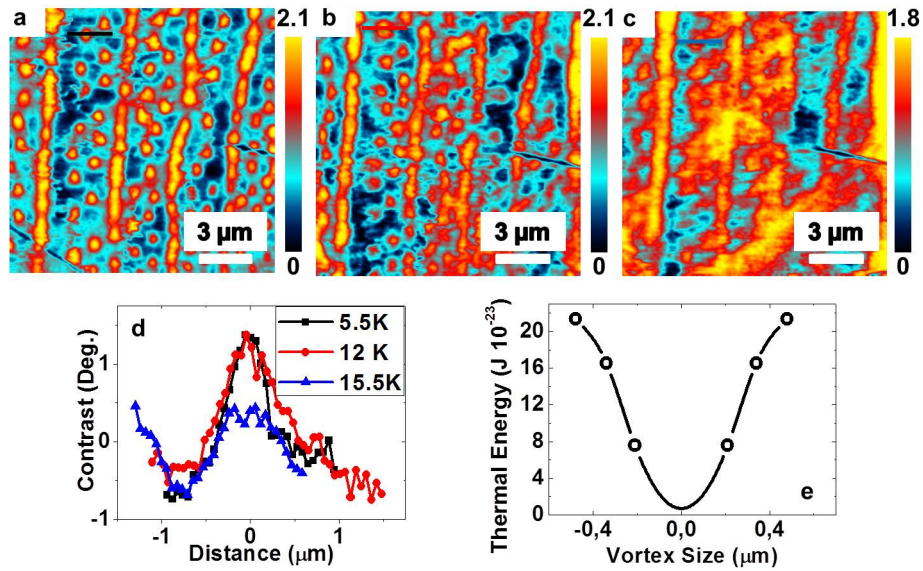


Figure 4.7: Thermal motion of PVs. In a, b and c, MFM images measured at 5.5 K, 12 K and 15.5 K respectively at the same area. The magnetic field is $B_y = 200$ Oe in the three images. The size of the PVs increases with temperature due to thermal motion. At 20 K the magnetic distribution becomes homogeneous in the whole field of view. In d, the magnetic profiles, measured at the same PV at the three temperatures. The PV is marked by black red and blue lines in the images. In e, the potential well of the PVs extracted from the data in d, the line is a guide to the eye.

The melting temperature, $T_m^* \approx 20$ K, obtained from figure 4.7 is far below the melting temperature reported by previous works, $T_m \approx 80$ K, for the same material at low fields [154, 168–170]. Moreover, according to [47], the dependence of the melting temperature with the tilted angle of the magnetic field is not enough itself to explain

such a low melting temperature. More likely, the melting of PVs is not exclusively a consequence of large thermal fluctuations. It has been pointed out by [84, 91, 160] that lateral magnetostatic forces during MFM imaging could also lead to a depinning of vortices. In our experiment, dragging due to the magnetostatic attraction between PVs and tip could also play a significant role, which suggest that the vortex-probe interaction is big enough to force PVs to move outside their equilibrium position at temperatures above 20K. This result gives a powerful tool to vortex manipulation in this system, as it shows that PVs can be manipulated by the MFM probe at reasonable low temperatures.

4.2.4 Manipulation of the crossing lattice

We have successfully manipulated the crossing lattice in our crystal. As a first step, we have manipulated a disordered arrangement of PVs. Then, we have successfully manipulated PV rows pinned on JVs.

4.2.4.1 Manipulation of PVs

We have successfully manipulated PVs combining the action of the in-plane magnetic field and the force of the MFM tip on the PVs. Figure 4.8 shows the evolution of an arbitrary arrangement of PVs when changing the angle of the in-plane magnetic field. The fast scan axis of the MFM is parallel to the X direction in all images. Figure 4.8 a, shows the original configuration of PVs, with round shapes and well localized positions at 12 K. After rotate the magnetic field by 10 degrees (figure 4.8 b) the same area was measured again. In the image, several PV magnetic profiles are elongated in the direction of the magnetic field. This behaviour is better seen after changing the angle to 45° (figure 4.8 c), 70° (figure 4.8 d) and 90° (figure 4.8 e). PV profiles became more elongated as the angle between the slow scan axis and the in-plane magnetic field decreases, always following the direction of the magnetic field. The elongation is maximum when the magnetic field is aligned with the slow axis of the MFM. Figure 4.8 f, summarizes the evolution of the PV elongation as a function of the angle between the fast scan axis and the magnetic field.

As it was presented in the introduction of this chapter, such vortex elongated profile was ascribed to vortex movement in previous works in YBCO [84, 87, 157]. The straight field of the MFM tip exerts a given lateral force, F_{lat} , on the vortices. The lateral force can be decomposed in two components, $F_{lat,s}$ and $F_{lat,f}$ in the directions of the slow and fast scan axis respectively. $F_{lat,f}$ shakes the vortices, moving them back and forth on its well potential while $F_{lat,s}$ can be used to drag vortices along the scan axis if the gain in energy of the vortex due to $F_{lat,f}$ is enough [84, 87, 157] (figure 4.9 a).

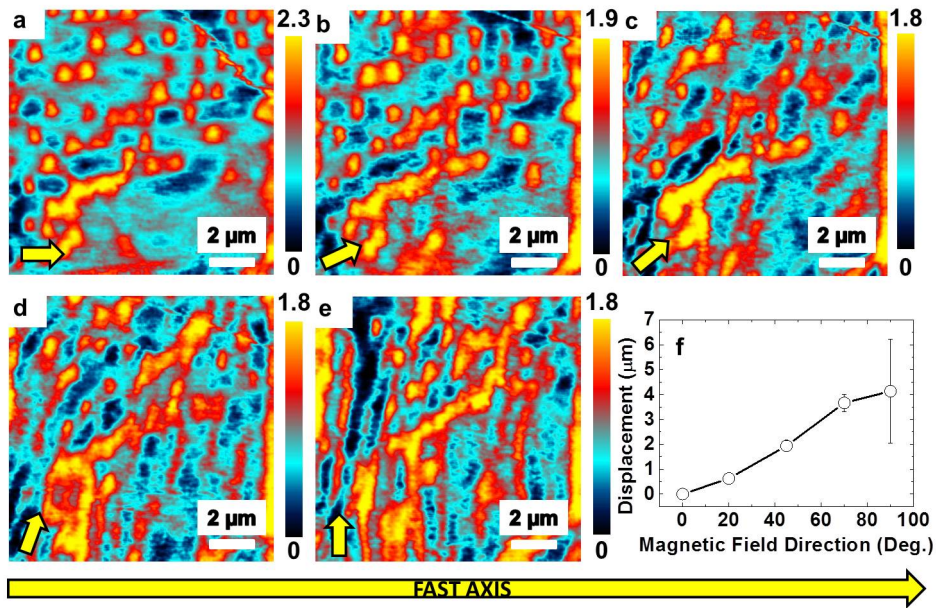


Figure 4.8: Motion of PVs by the combined action of the MFM tip and the rotating magnetic field. We show the evolution of a set of PVs when changing the direction of $B_{||} = 200$ Oe (marked as a yellow arrow in the images). In a, b, c, d and e, MFM images measured with an angle of $B_{||}$ with respect the X axis of 0° , 10° , 45° , 70° and 90° respectively. Some PVs presents elongated magnetic profiles in the direction of $B_{||}$ as the angle increases. The direction of the fast scan axis is represented by a yellow arrow at the bottom. In f, we show the average PVs displacement vs the angle, the line is a guide to the eye.

In addition, a parallel magnetic field applied to Bi-2112, will enter the material in form of JVs. As a consequence, superconducting currents flow on the CuO planes perpendicular to the direction of $B_{||}$. These currents exerts a Lorentz force, F_L , on the PVs parallel to the direction of $B_{||}$.

Combination of the two previous forces was used to manipulate PVs. From figure 4.8, it is clear that there is not movement when both forces are perpendicular and maximum when they are parallel. The process is schematized in figure 4.9. When $F_{lat,s}$ and F_L are perpendicular, no vortex movement was found, suggesting that $F_{lat,s}$ was not strong enough to manipulate the PVs. When the angle, Θ , between $F_{lat,s}$ and F_L is different from 90° , PVs movement was measured in the direction of B_{\parallel} . The movement, in this case is due to the sum of F_L and the projection of F_{ts} in the direction of F_L , as is shown in figure 4.9 c. PVs movement is stronger when $\Theta = 0$ as the total forces becomes maximum and PVs can be dragged far from their equilibrium positions (figure 4.9 d).

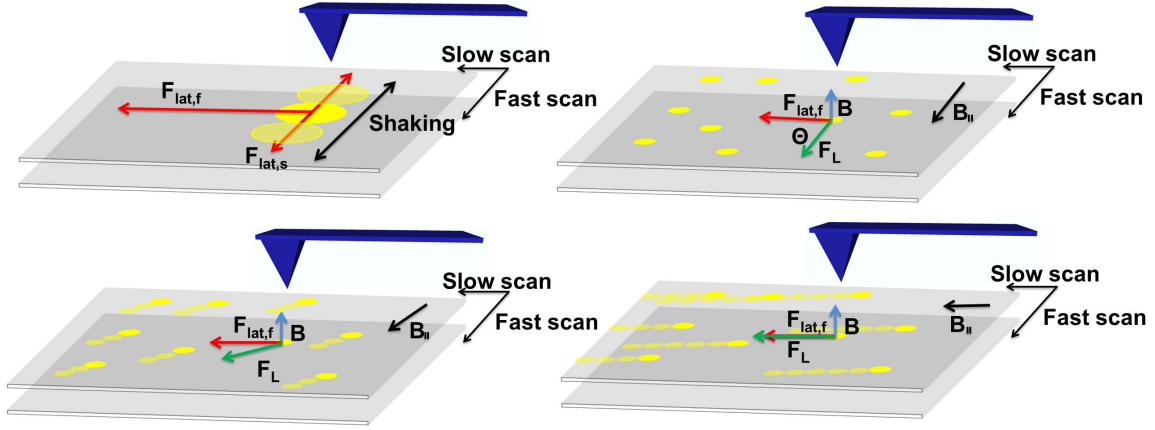


Figure 4.9: Scheme of the motion of PVs by the combined action of the MFM tip and the rotating magnetic field. The MFM tip is represented as a blue triangle, the CuO layers as grey planes and the PVs as yellow circles. We use transparent yellow circles to represent the PVs movement. In a, the force of the MFM tip acting on a PV is schematized. The force is decomposed in two components, $F_{lat,s}$ and $F_{lat,f}$ in the directions of the slow and fast scan axis respectively. $F_{lat,s}$ shakes the PV back and forth and $F_{lat,f}$ drags the PV. In b, the Lorentz force, F_L , acting on the PVs and the slow scan axis are perpendicular. They are not strong enough to move the PVs. In c, the the parallel magnetic field form an angle $\Theta \neq 90$ with the slow axis and the sum of both forces becomes strong enough to drag PVs a short distance in the direction of F_L . In d, both forces are parallel and the PVs move a bigger distance.

We have estimated the value of Lorentz force acting on a PV due to the JV supercurrents of about 50 pN. Pinning force of PVs is therefore stronger as F_L by itself is not enough to drag PVs. We have calculated the force exerted by the MFM probe

on a PV by using the equation 4.5. For our calculation we have use typical values of $\alpha = 0.35$, $h_0 = 250$ nm and $m = 30$ nAm following [157, 171, 172], and the experimental tip-sample separation of 120 nm, obtaining $F_{lat}^{max} = 80$ pN. PVs movement was achieved by the combination of the force of the MFM tip and the Lorentz force. Thus, we estimate the force needed to drag isolated PVs of ≈ 130 pN.

4.2.4.2 Manipulation of PVs on top of JVs

Figure 4.10 a, shows JVs decorated with PVs at 5.3 K. In the image, the in plane component of the magnetic field is aligned with the slow axis of the scan. PVs are well localized on top of the JVs with some clusters in between, without any signature of vortex movement. After change the direction of the scan by 90° and increase the temperature up to 12 K, the same area was measured again. Result are presented in figures 4.10 b and c. In these images, PVs are not well localized. They present an elongated profile in the direction of the slow scan axis, visible in the images as straight lines of magnetic flux connecting different JVs.

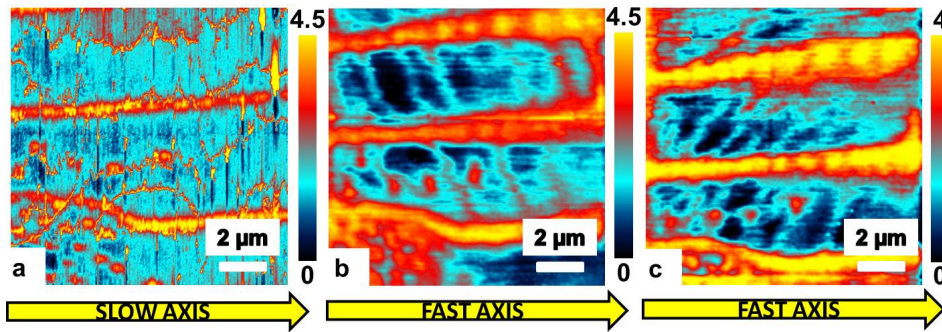


Figure 4.10: Triggering motion of PVs between JVs by the MFM tip. In a, we show an image measured at 5.4 K and $B_x = 200$ Oe with the slow scan axis parallel to B_x . In b and c, we show the same field of view at 12 K, changing the scanning direction (marked by the yellow arrow) with respect to a. Between b and c we change the direction over which the tip is scanned during imaging, from left to right and bottom to top in b and to right to left and top to bottom in c. Note that, in addition to the signal on top of the JVs, we observe strikes in between JVs.

Figure 4.10 b, was obtained by scanning from the bottom to the top and from right to left of the image while 4.10 c, was obtained by scanning from the top to

the bottom and from right to left. From visual inspection is clear that the straight trajectories of the PVs follow the direction of the tip during the scan.

In this case PVs are not randomly arranged on the surface but pinned on JVs forming rows. In this particular configuration we have found PVs movement in the direction of the scan. In this case, the force on the PVs from the tip is strong enough to depin them from the JVs, moving them from one JV to another. The process is schematized in figure 4.11, where the trajectories of three PV are shown, full yellow circles represents their equilibrium positions on the JVs and empty yellow circles their positions during the scan as a result of the force of the MFM tip.

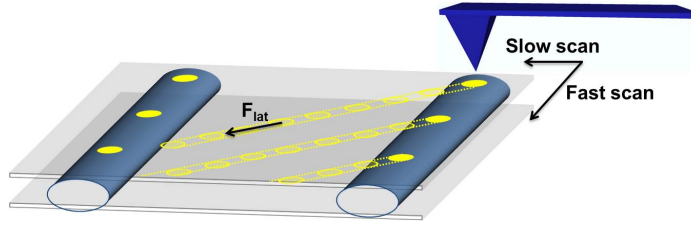


Figure 4.11: Scheme of the motion of PVs between JVs by the MFM tip. The MFM tip is represented as a blue pyramid, the JVs as blue cylinders, the PVs as yellow circles and the CuO layers as grey planes. The movement of the PVs is schematized by transparent yellow circles. The MFM tip exerts a given force, F_{lat} on the PVs in the direction of the scan.

We have calculated the attractive force per unit length along the c -axis, between a PV stack and a JV stack following equation 4.1. We have used $\gamma = 250$ and $a_z = 15.4\text{nm}$ calculated in previous sections, obtaining $f_x = 2.28 \cdot 10^{-7}\text{N/m}$. Assuming a sample width of about 0.5mm , the attraction force will be $f_x \approx 114\text{pN}$, smaller than the estimated force to drag isolated PVs and comparable to the force of the MFM tip on the PVs.

4.2.5 Crossing JVs

Previous works have demonstrated that the JV-PV interaction is sufficiently strong to indirectly pin JVs stacks at the location of pinned PVs [173]. We have used this behaviour to generate and keep a JV in a fixed direction and then cross other JVs with it by rotating the in-plane magnetic field.

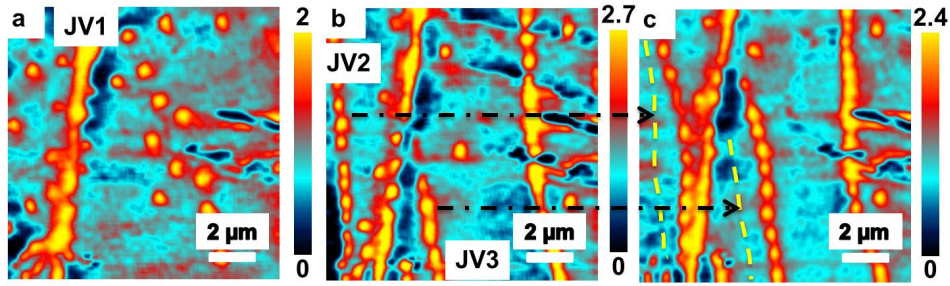


Figure 4.12: Crossing JVs. In a, a JV (denoted by JV1) pinned at a topographic feature at 5.3 K and $B_{\parallel} = 200$ Oe. In b, the same area, measure after heat the sample up to 20 K and modify the direction of B_{\parallel} by -5° with respect to the Y axis. Three new JVs appears in the image, two of them (denoted as JV2 and JV3) cross JV1. In c, after heat and cool the sample again, JV2 and JV3 have changed slightly their position. We mark the previous position of JV1 and JV2 by dashed yellow lines. Remarkably, JV2 is attracted to JV1 and JV3 cut JV1.

After localized an area with a longitudinal topographic feature, we have applied a tilted magnetic field with B_{\parallel} , parallel to the topographic feature, generating a series of decorated JVs at 5.3K. As a result, one JV and several PVs were pinned to the topographic feature. Then, we have heated the sample up to 20 K and cooled down to 5.3 K quickly. After that, we have measured the same area again. All PVs that were pinned on the JVs where depinned, except the ones on the JV on the topographic feature labelled as JV1 (figure 4.12 a). The pancake intervortex distances within the feature are consistent with the presence of a JV, indicating that the original JV is still pinned to the feature. Then, the angle of the in-plane magnetic field was changed by -5 degrees generating a new JV lattice tilted 5° with respect to the topographic feature. The new JV lattice crosses the topographic feature in two points (figure 4.12 b). Finally, the sample was heated up to 20 K and cooled down again to 5.5K to favour the JV lattice movement. After scanning the area again, we have found that the JVs have moved a few microns crossing the topographic feature at new points (figure 4.12 c). In figure 4.12 c) the JV labelled as JV2 approaches to the topographic feature in a asymptotic way while the JV labelled as JV3 form a kink with the feature.

A similar situation was previously reported by [66]. In this work, they were able to split the PVs row on top of a JV in two ‘forks’ by quickly changing the direction of the magnetic field. In their work, the double row of PVs relaxed back to a single

chain after a few minutes. This suggests that the JV was not split in two branches. Instead, the most possible scenario is that when changing the direction of magnetic field and therefore the direction of the JV, some of the PVs are dragged with it and others are not. Finally the PVs that were not dragged are attracted again to the JV forming a single row again.

Our case is completely different as the JV configuration was stable during all the experiment (several hours). Thus, we suggest that we are in a crossing flux configuration, where we have successfully crossed three JVs. The MFM does not allow to determine the direction of the JVs at the crossing point, but a twist, crossing and reconnection of the magnetic field inside the material is the most likely scenario as it was previously suggested by [174, 175].

4.2.6 Pinning of the crossing lattice at low temperatures

A previous work, has reported the possibility of manipulate JVs and PVs by changing the azimuthal angle of the applied magnetic field at high temperatures (80 K), where the pinning potential is weak [68]. Authors in [68], argue that at 80 K the pinning force acting on the PVs in their crystal is three times smaller than the attractive force between JVs and PVs. Thus, changing the direction of B_{\parallel} modifies the direction of the JVs and drags the PVs with them.

We have measured the evolution JVs decorated by PVs at low temperatures (5.5 K and 10 K) for different azimuthal angles. We have rotated the magnetic field in the XY plane up to 120° without finding any movement of the crossing lattice. JVs and PVs remain pinned at their original positions and do not change with the direction of the magnetic field. At 10 K only a increment of the PV profiles was measured. Results are presented in figure 4.13.

Our results prove that pinning of PVs on the JV lattice at low temperatures is big enough to avoid any JV movement with the magnetic field.

The effect of pinning in our crystal is also seen when raising B_{\perp} from 0 Oe to

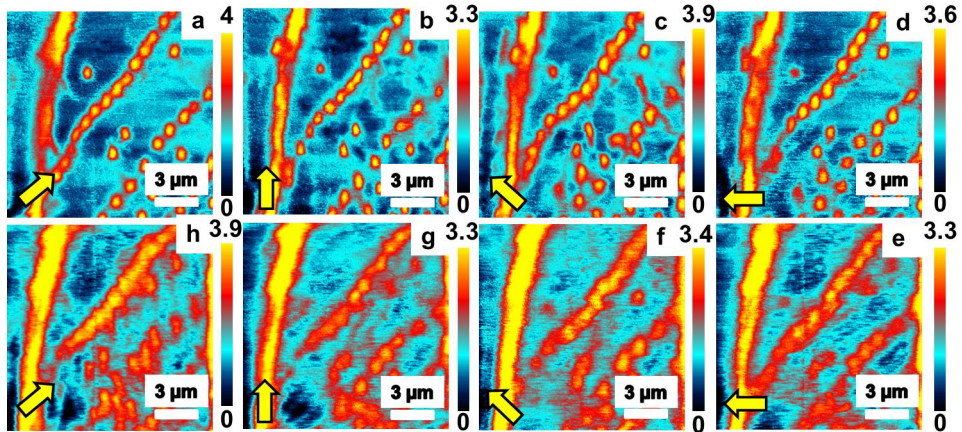


Figure 4.13: Pinned crossing lattice in rotating magnetic fields. In the figure we show the same field of view when changing the direction of $B_{\parallel}=200$ Oe. The direction of B_{\parallel} is marked by a yellow arrow at the images. a, b, c and d were measured at 5.3 K and e, f, g and h at 10 K. Clearly, JVs and PVs remain pinned, in spite of the varying direction of the magnetic field at both temperatures. The yellow stripe on the left is a feature due to the electrostatic interaction with an step.

2000 Oe as is presented in figure 4.14. First, we have obtained two JVs decorated with PVs at 5.5 K. Then we have increased the perpendicular magnetic field from 0 Oe to 2000 Oe in several steps, measuring the surface at each step. Surprisingly, the vortex distribution almost does not change until 2000 Oe where the flux distribution becomes homogeneous. Then, the magnetic field was decreased to 50 Oe and the hexagonal vortex lattice was recovered (Figure 4.14 i).

This indicates that the magnetic flux does not penetrate to the centre of the sample until the field reaches a trigger value (2000 Oe in our case), accumulating flux elsewhere.

The vortex distribution at low fields in High- T_C superconductors results from the competition of pinning and geometrical barriers (GBs) [176]. The GB is formed by the interplay between the vortex line tension and the Lorentz force that is induced by the circulating Meissner currents [176–182]. When B_{\perp} is increased, vortices entering through the edges are swept by the Meissner currents toward the center, where they accumulate, giving rise to a dome-shaped induction profile. If the effect of pinning is weak, the GB potential will dominate and PVs will accumulate at the centre of

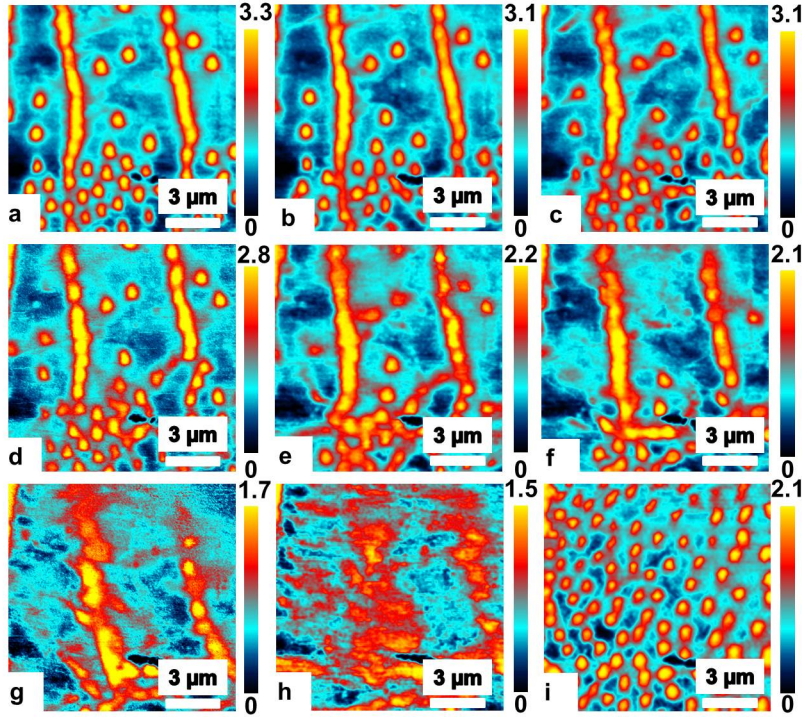


Figure 4.14: Ramping the Z field in crossed lattices. We show the evolution of the crossing lattice with the perpendicular component of the magnetic field (B_{\perp}) at constant temperature (5.3 K). B_{\parallel} remained constant at 200 Oe in the Y direction and the perpendicular component was 0 Oe (a), 100 Oe (b), 300 Oe (c), 500 Oe (d), 750 Oe (e), 1000 Oe (f), 1500 Oe (g), 2000 Oe (h). Clearly, the crossed lattices remain roughly at the same position below $B_{\perp} = 1500$ Oe. At $B_{\perp} = 1500$ Oe, no vortices are resolved. When decreasing B_{\perp} down to 50 Oe, we observe again the lattice of PVs.

the sample, if not, PVs will accumulate elsewhere depending on the pinning potential landscape on the surface. It has been also demonstrated that JVs in Bi-2212 serve as narrow channels for easy vortex entry and exit through the geometrical barrier (GB) [183], in this case, PVs still accumulate at the centre of the sample but with some of them decorating JVs outside the central dome.

We have found that there is not vortex entry to the centre of the samples at low fields. PVs only reach the centre of the sample at fields above 2000 Oe, which is a much bigger value than previous reported values for the entry of PV in Bi-2212 crystals with low pinning (≈ 5 Oe) [183]. From our experiment, we conclude that at low temperatures, the effect of pinning on PVs determines the flux entry on the

sample.

4.2.7 Evolution of the PV lattice with the polar angle of the magnetic field

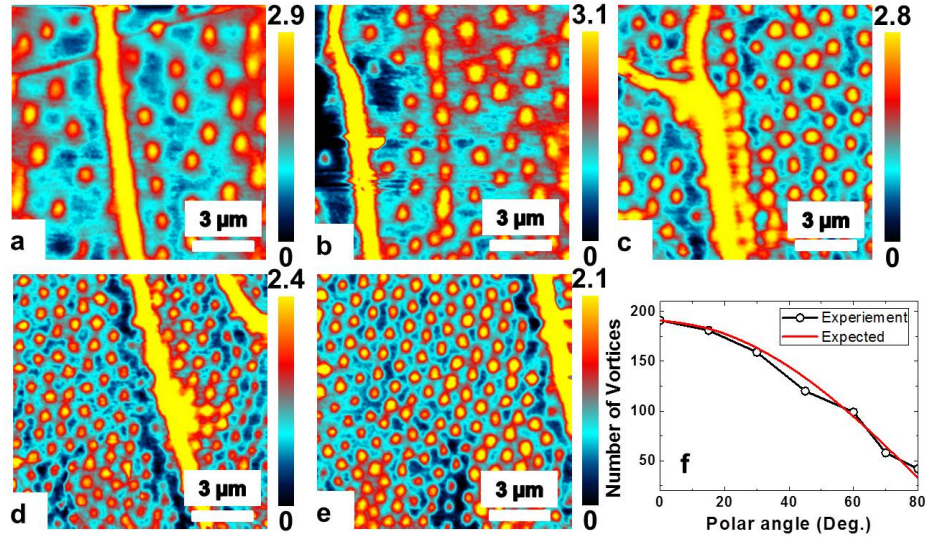


Figure 4.15: Evolution of the PV lattice with the polar angle. We show a set of images measured at constant magnetic field (27.5 Oe) and azimuthal angle ($\phi = 0$) but different polar angle. All are taken at 5.3 K in FC conditions, with the magnetic field applied above $T_C = 88$ K. Polar angle is 80° in a, 70° in b, 60° in c, 15° in d and 0° in e. The field of view is different in each image. In f, we compare the measured vortex density with the expected value within Ginzburg-Landau theory.

We have investigated the influence of B_\perp from a different approach. To avoid the pinning of the PVs, each measurement was taken at FC conditions. The sample was heated above its critical temperature and a constant magnetic field of 27.5 Oe was applied for different polar angles. Then, the sample was cooled down to 5.5 K. This procedure avoids the effect of pinning and allows to measure the evolution of the lattice at different angles, but does not allow to measure a specific area due to the thermal drift of the sample. Results are summarized in Figure 4.15.

All images of $12 \times 12 \mu\text{m}^2$ were measured at different positions of a bigger area about $30 \times 30 \mu\text{m}^2$. The number of vortices at the area, follows the expected behaviour (figure 4.15 f), indicating that at high temperatures where the pinning potential is

smaller, PVs enter to the centre of the sample with an homogeneous distribution.

4.3 Conclusions

In this chapter, we have studied the crossing lattice of Bi-2212 single crystal at low temperatures and low fields. We have successfully manipulated an arbitrary arrangement of PVs and the crossing lattice of JVs and PVs. We have measured the dependence of the PV movement with the angle between the in-plane magnetic field and the scan of the MFM. We have measured the necessary force to manipulate isolated PVs and PVs trapped on JVs. We were able to cross three JVs. We have also demonstrated that pinning determines the entry of PVs from the edges to the centre of the sample at low temperatures.

To our knowledge, ours is the first work showing manipulation of the crossing lattice in a superconductor.

Strain induced magneto-structural and superconducting transitions in $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$

Tuning parameters are an essential tool in the study of materials, since they can be used to promote a specific interaction. As an example, unconventional superconductivity often emerges around the point where antiferromagnetic order is suppressed by hydrostatic pressure [184]. Strain has been occasionally used as a tuning parameter [185–188], but is less employed than pressure. Additionally, strain has been employed to probe the nematic susceptibility of iron-based superconductors [189–192].

Iron-based superconductors have a huge and complex tunable interplay between antiferromagnetism, orthorhombic to tetragonal distortion and superconductivity. There are numerous tuning parameters in iron-based superconductors, including: chemical substitution [193], hydrostatic pressure [194, 195], epitaxial strains in thin films [196, 197], uniaxial strain in CaFe_2As_2 and BaFe_2As_2 [198–203] and biaxial pressure in $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ [9].

5.1 Previous studies in the parent compound CaFe_2As_2

The parent compound, CaFe_2As_2 , presents tetragonal structure together with paramagnetism at ambient conditions and transits to either an antiferromagnetic/orthorhombic (AFM/ORTH) phase or a paramagnetic/collapsed tetragonal (PM/CT) phase when decreasing the temperature, depending on the hydrostatic pressure [204]. The AFM/ORTH transition is also present on other compounds of the same family like BaFe_2As_2 , but CaFe_2As_2 is extremely sensitive to the pressure, for example it transits from orthorhombic to a collapsed tetragonal phase under 0.35 GPa at 33 K [204], which is a much more moderated pressure than BaFe_2As_2 (29 GPa [205]).

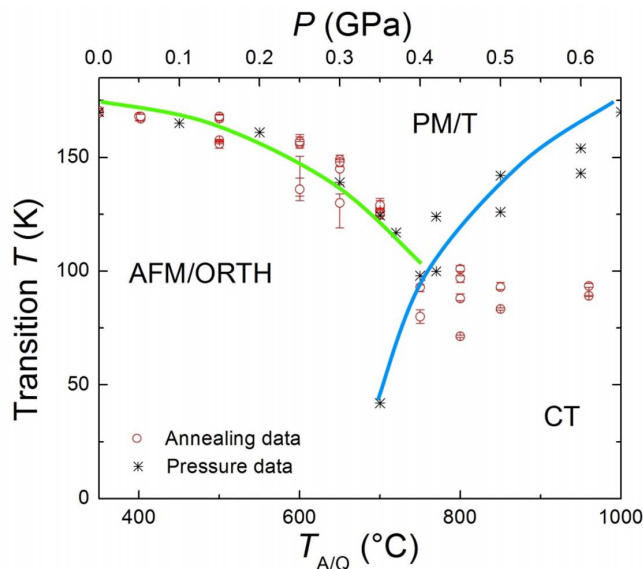


Figure 5.1: Phase diagram of CaFe_2As_2 as a function of the post growth annealing temperature and the hydrostatic pressure. In the lower x-axis, the annealing temperature and in the upper x-axis, the hydrostatic pressure. Black asterisks are the pressure data measured in [204] and red circles are the data obtained from the annealing treatment in [206]. Green and blue lines are a guide to the eye. Adapted from [207].

The possibility of stabilizing the PM/CT ground state at ambient pressure was also proved by [206] using a post growth annealing treatment. They argued that the changes in the internal strains due to the formation of FeAs nanoparticles in the sample are the cause of the change in the ground state as a function of the annealing temperature. Changing the annealing temperature will modify the size

of the FeAs precipitates and therefore the internal strain. The combination of the phase diagrams obtained in [204] and [206] is shown in figure 5.1. From the plot the equivalence between pressure and annealing temperature is clear.

5.1.1 Structural domains at low temperatures

Studies of polarized light microscopy have shown the formation of structural domains below the tetragonal to orthorhombic transition in iron-arsenide superconductors, AFe_2As_2 , $\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$ [208]. The authors of this work have associated the contrast of the optical images to the rotation of the polarization plane between neighbouring domains in twin boundaries of orthorhombic domains. Therefore, proportional to the degree of orthorhombic distortion

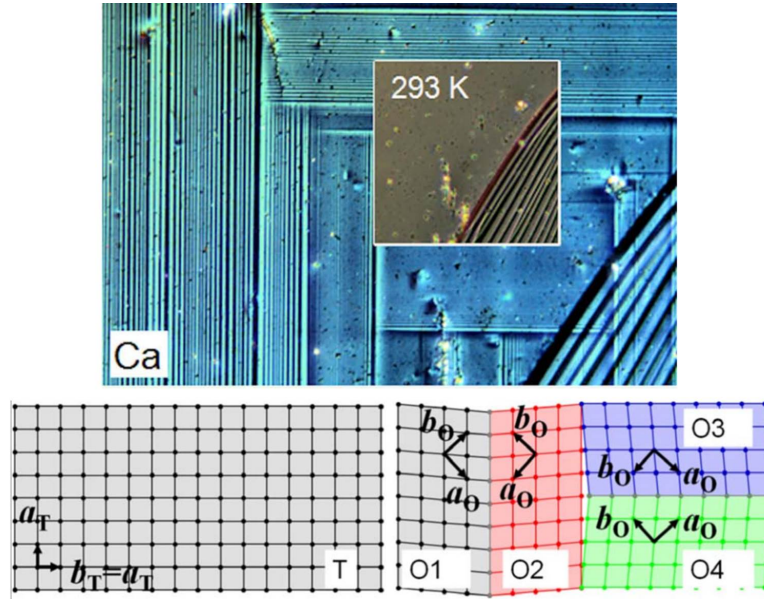


Figure 5.2: In the upper panel, a white light optical image measured in a polarization microscope showing a pattern of structural domains in CaFe_2As_2 , at $T \approx 5$ K. The characteristic spacing between the lines is about $10 \mu\text{m}$ and the contrast in optical images follows the magnitude of orthorhombic distortion in the compound. The inset shows terraces on the crystal at room temperature at the same area. The c axis is perpendicular to the surface. In the lower panel, in the left, an scheme of the atomic positions in the tetragonal lattice. In the right, an scheme of the orthorhombic distortion and formation of domain walls at low temperatures. Different colours are used to different domains. Adapted from [208]

In figure 5.2 we show an optical image of a CaFe_2As_2 single crystal at 5 K, well below the tetragonal to orthorhombic transition. A regular pattern of domain boundaries oriented in two orthogonal directions is clearly visible. A typical domain width is about $10 \mu\text{m}$. Over large areas, sometimes covering the whole surface of the crystal, domains form stacks of parallel plates. In some areas perpendicular domain sets interpenetrate. The crystal under study has terraces on the sample surface running at an angle to the figure and shown in inset at RT, with a step size of the order of $20 \mu\text{m}$ axis. On crossing the terraces, the domain lines perfectly match at different levels. This clearly shows that the domain walls are extended along the c axis.

A later work have shown the possibility of mechanical detwinning single crystals of CaFe_2As_2 via uniaxial strain [202]. Proving the huge dependence on the strain of the of structural domains in the compound.

5.2 Previous studies in $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$

A different approach was made by authors in [209], they combined the effect of cobalt substitution and post growth annealing to characterize the 3D phase diagram of $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ as a function of these two tuning parameters. In $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, substitution of Co for Fe suppresses a coupled first-order magnetostructural transition at $T_{s,N}$ and induces superconductivity with a maximum T_c of 16 K [209]. Authors in [209] probed that the ground state of $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ can be tuned to two new states, one superconducting, paramagnetic and tetragonal (SC/PM/T) state and another normal, paramagnetic and tetragonal (N/PM/T) state. The phase diagram obtained via resistivity, susceptibility and specific heat measurements is shown in figure 5.3. It is important to note that the studies from [209] were performed on free standing samples, only fixed using soft glues.

$\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ is also exceptionally pressure sensitive. Authors in [210] found a large rate of suppression of $T_{s,N}$ with hydrostatic pressure in the compound with $x=0.028$, $dT_{s,N}/dp \approx -1100 \text{ K/GPa}$.

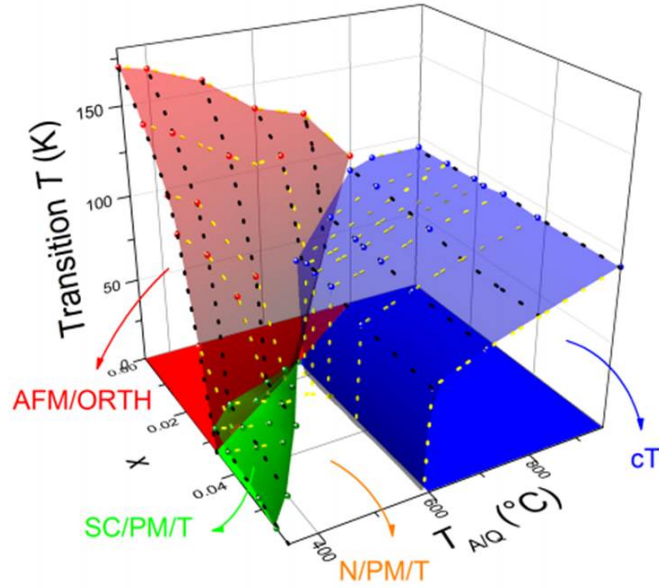


Figure 5.3: 3D phase diagram of $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. x is the substitution level and $T_{A/Q}$ the annealing/quenching temperature. Four phases are observed, in red the antiferromagnetic/orthorhombic (AFM/ORTH), in green the superconducting/paramagnetic/tetragonal (SC/PM/T), in white the non superconducting/paramagnetic/tetragonal (N/PM/T) and in blue the collapsed tetragonal (CT) state. Adapted from [207].

5.2.1 Effect of biaxial strain

A recent work has focused in the effect of biaxial strain on the doped compound $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ [9]. The authors have studied the effect of biaxial strain by making use of the different thermal expansion between the sample and a rigid substrate where the sample was glued. They measured a series of samples with different Co concentrations, first in free standing conditions and then glued to a rigid substrate. With a combination of high energy x-ray diffraction (XRD) and capacitance dilatometry techniques, they compared the evolution of the lattice parameters of both, free standing and glued samples, finding that the effect of biaxial strain induced by the difference between substrate and sample thermal expansion coefficients, modifies the sample state. The different expansion coefficients, causes strain in the a-b plane of the sample affecting the c/a ratio. They found that samples that do not show AFM/ORTH transition when free standing, show an structural transition when glued to a substrate, proving that the c/a ratio is a suitable tuning parameter in $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ [9].

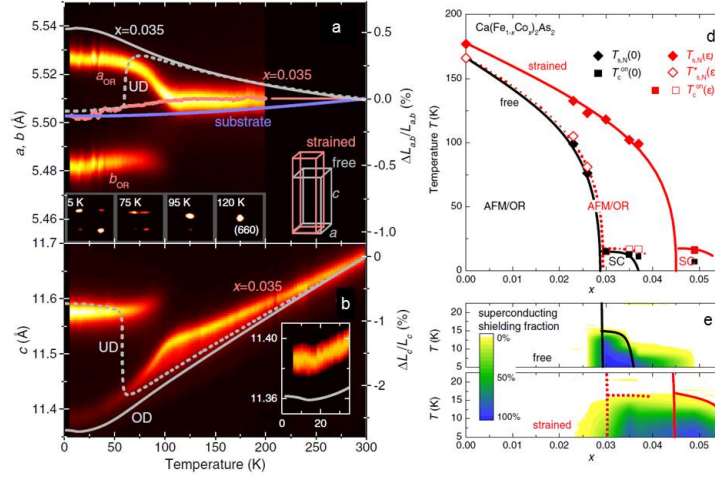


Figure 5.4: In the left panel, the in plane and c axis structural data for $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ $x=0.35$ is presented. The colour code intensity maps represents the lattice parameters measured by x-rays diffraction when warming the sample. Lines indicate uniaxial fractional length changes, $\Delta L_i = L_i$ ($i=c$, c axis and $i=a, b$, in-plane average), of free overdoped (OD samples, $x=0.035$ in a and $x=0.029$ in b) and of a representative underdoped (UD) $x=0.027$ sample obtained by capacitance dilatometry. The blue line shows the substrate thermal expansion and the red line indicates the average in-plane length of strained $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ inferred from the diffraction data. The right inset in a shows an scheme of the deformation of the unit cell due to the strain. The row of insets in a, show the diffraction pattern close to the tetragonal (660) reflection revealing orthorhombic domains. The inset in b presents the data on expanded scales. In the right panel, in c, the phase diagram of $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ in the free (black) and strained (red) state. The AFM/ORTH transition at $T_{s,N}(\epsilon)$ is only gradual. Red open symbols and dashed lines correspond to the remaining phase fraction within the strained sample. In e, the superconducting shielding fraction of free and strained samples, respectively. Lines are a guide to the eye. Adapted from [9].

In the figure 5.4 a, the in plane and c axis structural data for samples with $x=0.35$ is presented in combination with the data for free standing samples obtained by capacitance dilatometry. The substrate thermal expansion is also shown. In figure 5.4 c, the phase diagram of $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ in the free and strained state is presented. In the diagram is shown how the AFM/ORTH transition in the strained sample takes place at higher Co concentration than in the free standing samples, as the temperature decreases, the c/a ratio is modified due to the strain on the sample,

favouring the nucleation of orthorhombic domains in certain regions of the sample, splitting the samples in orthorhombic and tetragonal domains. In the under doped (UD) samples ($x < 0.28$) the samples have a strain induced orthorhombic structural transition at a temperature above the structural/magnetic transition of free samples. Below this temperature the sample is split into tetragonal and orthorhombic domains and below a temperature close to the orthorhombic transition of free samples, the remaining tetragonal domains of the UD samples transition to the orthorhombic phase. For overdoped (OD) samples with cobalt concentrations between 0.28 and 0.49, the strain produces the coexistence of tetragonal and orthorhombic domains in the sample below $T_{s,N}(\epsilon)$ that persists until lower temperatures. For those samples, when the temperature is decreased below the T_C of free standing samples, the strained ones, present a superconducting transition associated to the tetragonal domains. Above concentrations of 0.49, there is no structural transition associated with strain while the superconducting transition is still present below T_C .

5.2.1.1 Domain boundaries in strained samples

The samples that present coexistence between orthorhombic and tetragonal domains, have domain boundaries separating both phases and different orientations of the same phase [9]. In the orthorhombic phase, the small axis, b_{ORTH} , is equal to the lattice parameter of the tetragonal phase, a_{TET} and therefore, the tetragonal/orthorhombic domain boundary will occur along the crystallographic direction determined by b_{ORTH} . Note in particular that the interface between these domains has no stress within the plane, because in-plane lattice constants coincide along the interface [9]. Domain boundaries between orthorhombic domains are by contrast, oriented at 45° with respect to the crystallographic axis, forming a twin boundary similarly to the case of the parent compound $CaFe_2As_2$ described before [208]. In figure 5.5 two examples of the orthorhombic/tetragonal and orthorhombic/orthorhombic domain walls are presented

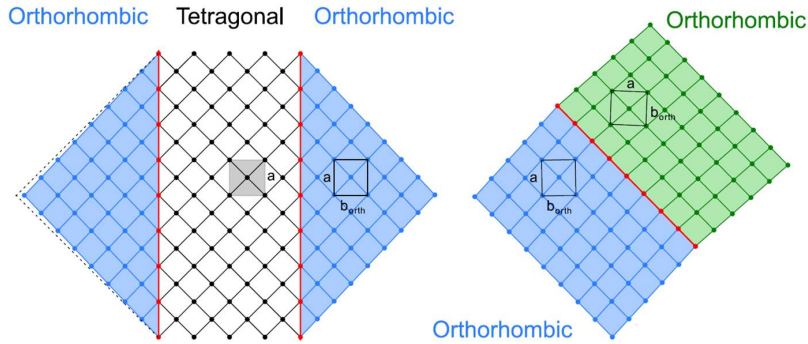


Figure 5.5: Example of the structural domain boundaries in strained $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. In the left panel, two domain boundaries between two orthorhombic and a tetragonal domain, the domain boundary develops along the crystallographic direction determined by b_{ORTH} . In the right panel, the boundary between two orthorhombic domains, forming 45° with respect to the crystallographic axis. Different colours are used to different crystallographic domains. The domain wall is represented as a red line. Adapted from [75, 208].

5.3 AFM/MFM studies in $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$

Previous works have studied the coexistence of tetragonal and orthorhombic domains in strained $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ single crystals from a macroscopic point of view [9]. Prior to our work there was not microscopic evidence of the distribution of those domains or the geometry of their boundaries. Moreover, the interplay between tetragonal and orthorhombic domains in the superconducting phase remains unclear. This was our main motivation and the starting point of our study in this compound.

AFM/MFM measurements were performed in the set up of our lab described in section 2.1 in a $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ single crystal doped with a 3.5% of Co and annealed at 350° . The crystal was grown by the Group of Prof. Paul Canfield at *Ames Laboratory* in Iowa, following the procedure described in [207].

Before the AFM/MFM measurements, the crystal was glued on a copper substrate with low temperature silver epoxy to apply a biaxial strain on it at low temperatures, similarly to the experiment in [9]. The same sample was also measured with STM at the *Laboratorio de Bajas Temperaturas de la Universidad Autonoma de Madrid* (LBTUAM) by Dr. Anton Fente in a set-up similar to the one described in [211].

Some of his STM result will be presented together with the AFM/MFM measurement to complement them, but the specifics of the STM experiments will not be given here as they are beyond the scope of this thesis. It is important to emphasize, that the sample was never unglued from the copper substrate, neither between AFM/MFM experiments, neither to perform the STM measurement. Therefore, the strain on the sample should be the same or at least comparable in the AFM/MFM and the STM measurements.

5.3.1 Topographic characterization

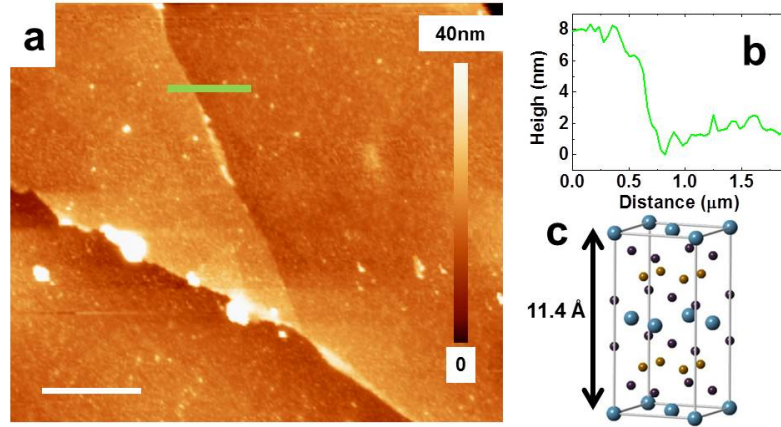


Figure 5.6: In a, a topographic AFM image of the sample at 100 K. The image shows large flat areas separated by steps of few nanometres high. Scale bar is $2\mu\text{m}$. In b, the height of the profile marked as a green line on the topography is presented. In c, the unit cell of the sample. Ca atoms are represented in blue, Fe atoms in yellow and As atoms in purple.

Topographic measurements were taken using the AFM dynamic mode described in section 2.1.3.7.1 with typical tip-sample separation around 10 nm.

Very flat surfaces were easily found after cleaving the sample, making possible topographic and magnetic images at 2 K up to $\approx 15 \times 15\mu\text{m}^2$ with height differences of tens of nanometres. The cleaving of the surface is expected to occur in the *c*-axis [75]. An example of the crystal surface is presented in figure 5.6 together with a topographic profile, and the unit cell of the crystal.

5.3.2 Tetragonal to orthorhombic structural transition

As it was presented at the introduction of this chapter, a partial tetragonal to orthorhombic structural transition is expected for strained $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$ samples at $T_{s,N}(\epsilon) \approx 100$ K.

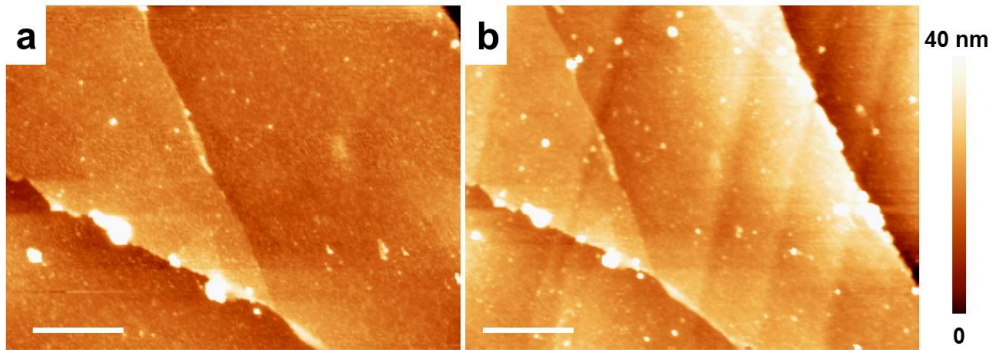


Figure 5.7: Topography of the strained $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$ sample above and below $T_{s,N}(\epsilon)$. In a, a topographic image measured at 100 K. In b, the same area measured at 32 K. At 100 K the image shows atomically flat terraces with steps of few nanometres in between. At 32 K longitudinal stripes at an angle with respect to the image are visible. The scale bar represents $2\mu\text{m}$.

We have performed AFM measurements above and below $T_{s,N}(\epsilon)$ in different areas of the sample, finding radically different topographies. Above $T_{s,N}(\epsilon)$, in the tetragonal phase, flat terraces and small steps of few nanometres were found (figure 5.7 a). Slightly below the structural transition expected from [9], we have observed clear stripes in the topographic images (figure 5.7 b). The stripes are separated by flat regions, few microns wide and are straight and parallel on the hole field of view. The origin of the stripes will be discussed in the following.

5.3.2.1 Origin of the topographic stripes

The corrugation observed reminds AFM measurements below the tetragonal to orthorhombic transition in BaTiO_3 [212] and STM measurement below the Verwey transition in FeO_3 [213]. In both material, the corrugation in the surface was associated to the reorientation of the structural domains due to the changes in the lattice parameters in the transition. We believe that our images can be explained in the

same way. We now will introduce a simple model originally developed to explain the corrugation in $BaTiO_3$ single crystals[214].

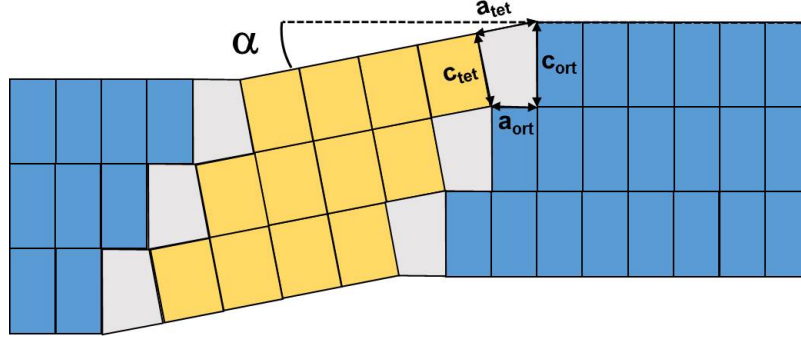


Figure 5.8: Scheme of the corrugation below the structural transition. The orthorhombic and tetragonal unit cells are represented in blue and yellow rectangles respectively. The unit cell at the interface is represented as a grey polygon.

A condition to the formation of domain walls, is the matching and continuity of the lattice at the wall [214]. Due to the differences in the c lattice parameter between the two phases below $T_{s,N}$ in our crystal, this condition is fulfilled by the accommodation of the tetragonal and orthorhombic domains schematically showed in figure 5.8. The angle formed by the perpendicular and in-plane axis of both lattices at each side of the wall is not exactly 90° . It differs from 90° by an angle $\alpha = \arctan(c_{tet}/a_{tet}) + \arctan(a_{ort}/c_{ort})$ [214]. We have calculated the expected angle of the corrugation at the tet/orth domain wall using the lattice parameters obtained for strained samples from [9]. We have found a corrugation angle of $\alpha \approx 0.55^\circ$.

Topographic features in form of stripes were also measured in STM experiments in the same sample (figure 5.9). In this case, the stripes are separated by tens of nanometres and are few Angstroms high. The STM measurements also show the 2×1 reconstruction of Ca atoms expected for $CaFe_2As_2$ [75, 215] covering most of the surface. The two main axis of the Ca reconstruction (corresponding to the two main axis of the Ca sublattice) form 45° with the crystallographic axis [153]. As is shown in figure 5.9, the reconstruction is found forming 45° with the topographic stripes (vertical lines in the figure), thus the stripes are oriented with the direction of the crystallographic axis. As it was presented in the introduction of the chapter, the domain boundary compatible with a direction of the crystallographic axis is the

tetragonal/orthorhombic domain wall, with the domain parallel to the a_{TET} and b_{ORTH} axis. This support the idea that stripes are related with the tet/orth domain boundaries

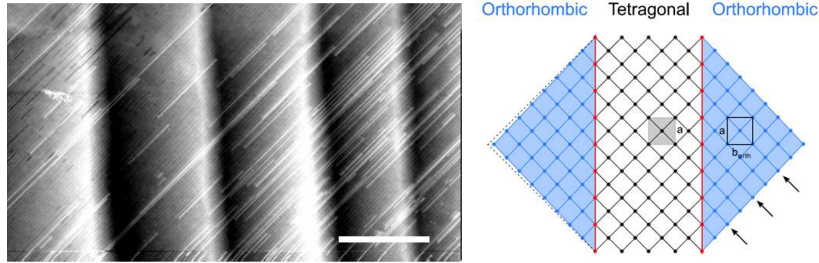


Figure 5.9: In the left panel, an STM topographic image taken at $T < 4.2K$. The image display parallel elongated stripes, forming 45° with the 2×1 Ca reconstruction. The white scale bar represents 100 nm. In the right panel, a schematized tetragonal/orthorhombic domain wall. The 2×1 Ca reconstruction is pointed by black arrows and different domains are represented by different colours.

We have calculated the angle between domains at both sides of the stripes in AFM and STM images, finding that it remains almost constant in all the stripes with a value between 0.8 and 1.3° . A few STM and AFM selected images are shown together with their topographic profiles and the measured angle at the stripes in figure 5.10. It is noticeable that images with such different scales, present the same angle between domains.

The value of the angle measured at the stripes greater the calculated angle for this system ($\approx 0.55^\circ$). But, it is important to note that the distortion of the tetragonal and orthorhombic lattice is dependent on the strain and therefore on the substrate where the sample is glued. We have followed a procedure similar to the one in reference [9] but we can not measure the magnitude of the strain in our samples. Thus, we can not unambiguously determine the magnitude of the distortion of the unit cells in our crystal. Moreover, the model used to calculate the distortion of the lattice at the boundary proposed in [214] is very simple. It suppose that the unit cell at the boundary is deformed, presenting exactly the same cell parameters of the tetragonal and orthorhombic phases at each side of the boundary, which do not have to be exactly true. For example, small deviation of $\approx 0.5\%$ from the values of the cell parameters measured by [9], leads to a different $\alpha \approx 1^\circ$.

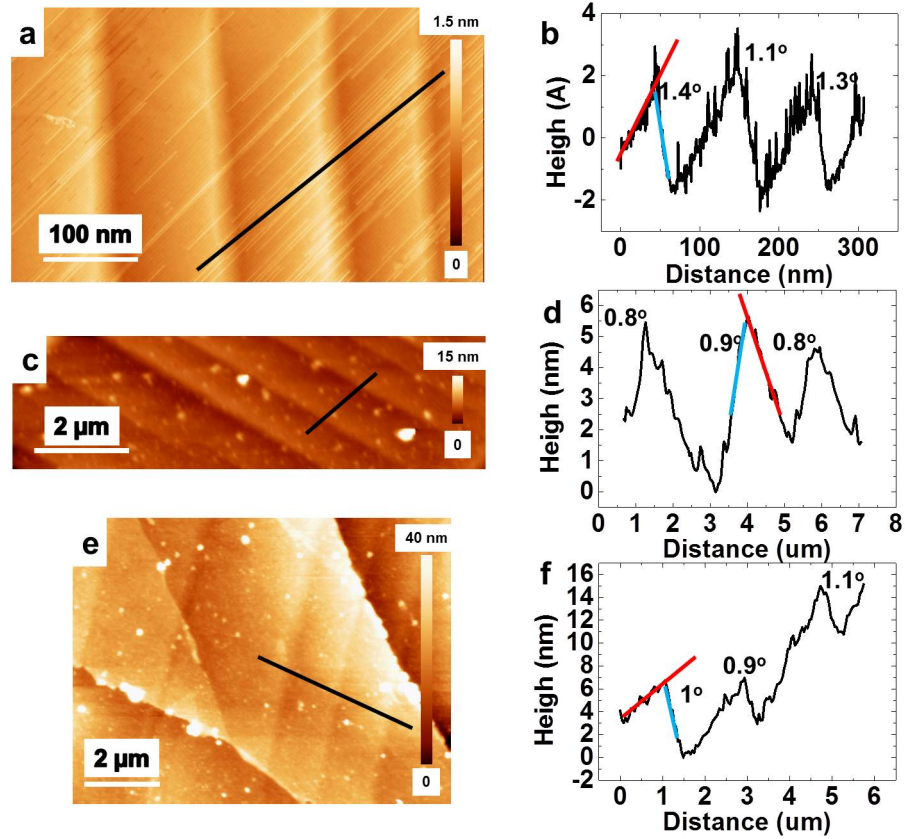


Figure 5.10: In the left column, the topographic images measured by STM (a) and AFM (c and e). The images, show step-like features associated to the tetragonal-orthorhombic domain boundaries. In the right column, three topographic profiles measured black lines on the topographic images. All the step-like features showed in the profiles present an angle of $\approx 1^\circ$ between the tetragonal and the orthorhombic domains. The surface of the tetragonal and orthorhombic domains is highlighted in the profiles using a blue and a red line respectively.

We believe that, given the simple approximations made, the agreement is remarkable and provides a simple but successfully explanation to the corrugation on the topography in our samples.

5.3.2.2 Evolution of the corrugation on the surface

We have measured the evolution of the stripes with the temperature at the same area. The results are shown in figure 5.11.

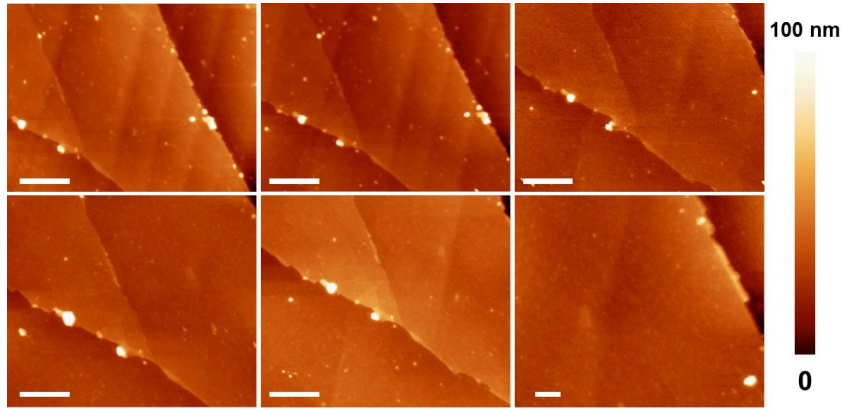


Figure 5.11: Evolution of the stripes on the surface with the temperature. Images measured when heating the sample at 17 K (a), 32 K (b), 55 K (c) and 68 K (d), e and f where measured after cooled the sample again at 55 K from 68 K. The stripes on the images become less visible at 55 K and they are not present at 68 K, they reappear at the same positions after decrease the temperature again. Scale bars represents $2\mu\text{m}$.

Images a, b, c and d were taken at 17 K, 32 K 55 K and 68 K respectively. In them, the stripes remain at the same positions until the temperature reaches 55 K where some of the stripes start to vanish to be completely lost at 68 K. This temperature is close to the expected $T_{s,N}(\epsilon)$ [9]. Then, the sample was cooled down and measured again at 55 K (figure 5.11 e and f), obtaining the same position for the stripes that in the previous case. This result is in perfect agreement with our assumptions that the topographic stripes are domain walls formed below $T_{s,N}(\epsilon)$.

It is important to clarify, that the value of $T_{s,N}(\epsilon)$ can not be unambiguously determined by the measurements of the stripes on the surface. Domain boundaries can nucleate first in another area of the surface at higher temperatures and extend to our field of view at lower temperatures. Therefore the measured value of $T_{s,N}(\epsilon) \approx 68$ K for our sample could be smaller than the real value for the magneto structural transition. Also, the transition measured in [9] is very broad, beginning at $T_{s,N}(\epsilon) \approx 100$ K and extending down to almost 50 K, which suggests that the formation of the orthorhombic domains is continuous when decreasing the temperature.

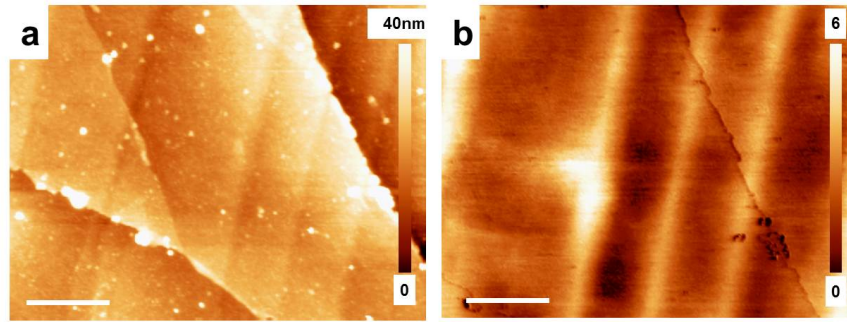


Figure 5.12: In a and b the topographic and magnetic image of the same area measured at 4.2 K and 1360 Oe. The magnetic image show elongated stripes that perfectly matches the topographic features originated when the sample is cooled bellow $T_{s,N}(\epsilon)$. White scale bars represents $2\mu\text{m}$.

5.3.3 Superconducting transition

To further understand the behaviour of the tetragonal and orthorhombic domains on the sample and they interplay in the superconducting properties, we have measured the same area presented in figure 5.11, in the MFM mode at 4 K. After FC the sample at 1360 Oe bellow the superconducting critical temperature of free standing samples ($T_C \approx 16$ K), we have performed MFM measurements, finding that magnetic images show elongated an alternative paramagnetic and diamagnetic domains that exactly coincide with the topographic stripes observed in the AFM images (figure 5.12). Similar stripes where found in SQUID microscopy and MFM measurements in the compound of the same family, $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, by Kalisky et al [216, 217]. In this case, they have associated the alternative stripes to an enhanced superfluid density on twin boundaries. By contrast, in our case, the magnetic domains seems to be related to the orthorhombic phase which exhibits antiferromagnetism and the tetragonal phase which exhibits superconductivity at low temperatures. We have not found any difference with the topography at higher temperatures.

5.3.3.1 Evolution with the Temperature

To clarify if the magnetic signal is related or not with the superconducting transition, we have imaged the same area after FC the sample at 230 Oe, at different temper-

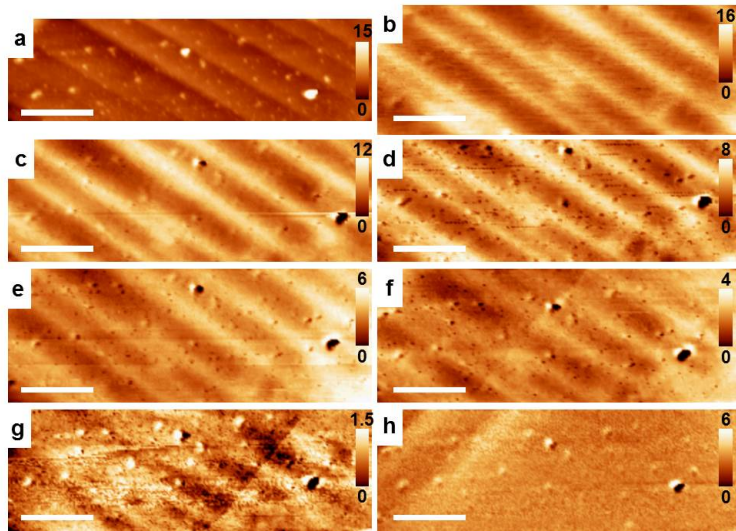


Figure 5.13: Evolution of the magnetic stripes with the temperature. In a, the topographic image of the area where the magnetic images were measured. In b-h, the magnetic images measured at 3.7 K, 4.8 K, 7.8K, 11.2K, 12.6K, 14 K and 16 K respectively. The contrast of the superconducting domains (white and light yellow), become softer as the temperature increases, until they are not visible at 16 K in agreement with the expected T_C for the superconducting transition. All measurements were done with an applied field of 230 Oe. Scale bar is $2\mu\text{m}$.

atures, keeping the magnetic field constant. We have found that the diamagnetic domains become broader and less intense as the temperature increases and they are completely gone at temperatures above 16 K, which is the expected T_C for the sample [9].

We have combined the information obtained in the MFM measurements with the STM data. In STM experiments, conductance maps at zero magnetic field near the stripes also saw a GAP opening that matches the expected GAP for the material ($\Delta = 1.78K_B T_C \approx 2.3$ mV) [72, 75]. STM conductance maps at 6 T also show vortex images at the tetragonal domains with intervortex distance corresponding to the expected one as is presented in figure 5.14 [75].

Therefore we can conclude that the diamagnetic domains observed in the MFM measurements are the result of the superconducting transition of the tetragonal domains of the sample that are still present at the sample bellow $T_{s,N}(\epsilon)$. The paramagnetic domains are related with the orthorhombic domains. It is interesting to note

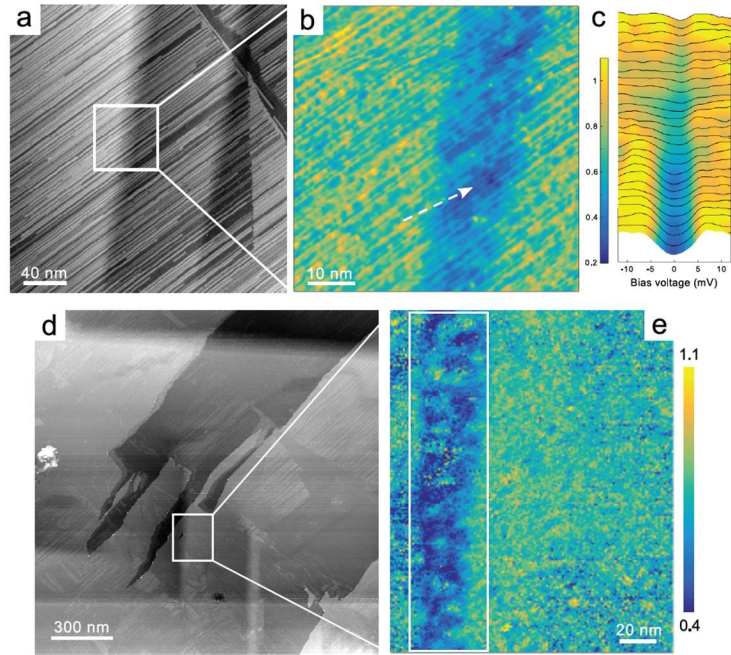


Figure 5.14: In a, a topographic STM image measured at 2 K. Surface shows a step-like feature similar to those founded in the AFM measurement. In b, zero bias normalized conductance map of the area in the white square in a at zero magnetic field. In c, normalized conductance curves along the line in b, showing a superconducting GAP in the expected energy range opening in the tetragonal domain. In d, an STM topographic image measured at 2 K. In e, zero bias normalized conductance map measured in the area of the white rectangle in d at $H = 6$ T, showing superconducting vortices (green spots on the blue area). Adapted from [75].

that the sample is split in two different phases normal/superconductor related with structural domains.

5.3.3.2 Evolution with the magnetic field

MFM measurements were taken at different applied magnetic fields at 4 K to characterize the evolution of the tetragonal-superconductor domains. The same area was mapped from perpendicular fields of 25 Oe to 1360 Oe after FC at 25 Oe. The results are shown in figure 5.15. The superconducting domains become thinner as the magnetic field increases. At the lower fields, there are some small domains perpendicular to the topographic stripes. When increasing the magnetic field we observe that the overall difference between large and small magnetization decreases and that

the perpendicular domains become normal.

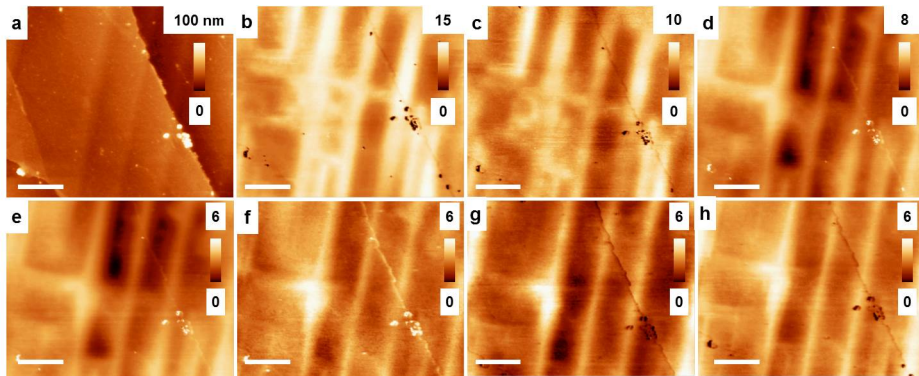


Figure 5.15: Evolution with the applied magnetic field of the magnetic stripes. In a, the topographic image of the area were the MFM images were measured. In b-h the magnetic images measured at 25, 70, 300, 430, 700, 1160 and 1360 Oe respectively. The superconducting domains (white and light yellow) become thinner as the magnetic field increases and some small domains perpendicular to bigger domains along the vertical dimension are visible at the lower field and are not visible at higher fields. Scale bar is 2 μm .

The AFM resolution do not allow to determine if there are smaller stripes associated to this domains. The possible origin of this orthogonal domains will be discussed in the following section.

5.3.4 Origin of the perpendicular domains

In the previous section, it was shown that there are some superconducting domains that are perpendicular to the topographic stripes and seems not to be related with any feature in the topographic image. This is nicely seen in images at bigger areas as the one presented in figure 5.16.

Figure 5.16 shows intersection of superconducting domains, always forming angles of $\approx 90^\circ$. This may be explained by the formation of tetragonal-orthorhombic domain walls at each side of the twin boundaries between two orthorhombic domains as is schematized in figure 5.16. Two orthorhombic domains in a twin boundary, form an angle of $\approx 2^\circ$ between them due to the orthorhombic distortion [72, 75]. As a result, the angle between two tetragonal domains at both sides of a twin boundary should

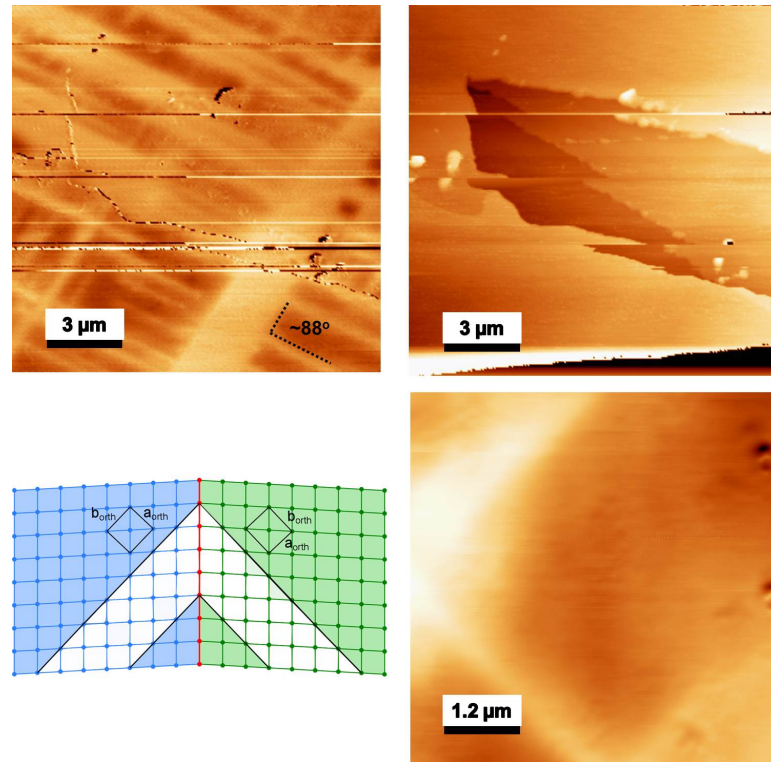


Figure 5.16: In the upper panel, two MFM and AFM images measured at 4K and 100 Oe. In the left, in the MFM image, elongated diamagnetic stripes associated to the tetragonal domains are observed. The stripes presents two main directions with an angle between them that differs from 90° by a few degrees. In the right, the topography image of the same area. In the lower panel, in the left, an schematic representation of two tetragonal domains at both sides of an orthorhombic twin boundary. Orthorhombic domains are represented in blue and green and tetragonal domains in white. The twin boundary is represented as a red line. In the right, a closer MFM image to the intersection of three tetragonal domains.

differ from 90° by this small angle. This is compatible with the MFM images, where the angle between the stripes gives values that differs from 90° by a few degrees, the same distortion that was found in the STM measurements for the 2×1 Ca reconstruction [75]. However, that would also result in some surface corrugation, which we do not observe. Another possibility is that fluctuations induce superconducting correlations in some parts of the orthorhombic phase. In both cases, this result shows that superconductivity in the tetragonal linear domains can be connected with each other.

In the topographic image of the same area, a huge step of several tens of nanome-

tres is shown at the centre of the image. On crossing the big step, the domain lines perfectly match. This shows that the domain walls are extended along the c axis.

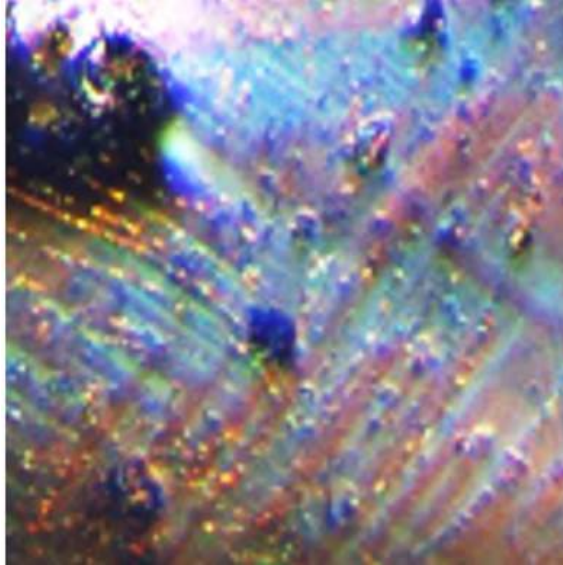


Figure 5.17: An optical image taken with polarized light in a different $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$, revealing orthogonal domains on the whole field of view.

Optical images taken with polarized light by the group of Prof. Paul Canfield at 5 K in another strained sample also shows perpendicular domains covering the hole field of view (figure 5.17). The optical image compares nicely with our MFM measurements but, in it, is not clear if the contrast is formed by the orthorhombic distortion at the twin boundaries as in the case of the parent compound presented in the introduction of this chapter or due to the tetragonal/orthorhombic domains as in the MFM images.

5.4 Conclusions

In this chapter we have studied the effect of strain in a $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$ single crystal from the microscopic point of view. We have imaged the coexistence of tetragonal/orthorhombic domain walls bellow the strains mediated transition at $T_{s,N}(\epsilon)$.

Bellow the superconducting critical temperature of free standing samples, we have measured the formation of diamagnetic domains coinciding with the tetragonal do-

mains. We have associated the diamagnetic domains with the superconducting transition of the remaining tetragonal phase. We have characterized their evolution with the applied magnetic field and the temperature. STM images are consistent with our results, showing the opening of a superconducting GAP and the existence of vortices in the tetragonal domains bellow T_C .

Quite likely, the size of the domains can be modified by applying uniaxial stress to the substrate, either perpendicular or parallel to the stripes. Or simply by changing the substrate. For instance, the thermal expansion of glass is of -0.1% which should result in a differential thermal expansion of 0.6% between sample and substrate and eventually lead to modified length scales in the domain size and distribution. Thus, strain might be used as a control parameter to produce novel kinds of superconducting systems, such as intrinsic Josephson junction arrays or to use the domain structure to improve vortex pinning. At very low magnetic fields we observe sometimes linear diamagnetic structures in the orthorhombic phase that might join elongated tetragonal domains, suggesting that such a coupling between elongated domains can indeed happen in some parts of the sample.

To our knowledge, ours is the first experimental work showing phase separation associated to strain bellow T_C in pnictides. The likely absence of magnetic order in the tetragonal domains, having in close spatial proximity a magnetically ordered domain, suggests that magnetic and superconducting order are both antagonistic, although they are probably fed by the same fluctuations.

Exfoliation and characterization of layered superconductors and graphene/superconductor heterostructures

6.1 Introduction

The pioneering work published by Novoselov et al. [218] gave rise to the isolation of single layers of graphene. They reported the repeated peeling of high oriented pyrolytic graphite (HOPG) on a photoresist layer and the final release of the resulting thin flakes in acetone. This method was later improved with the *dry* exfoliation of several layered materials by simply pushing the surface of crystalline samples against different surfaces [219]. This basic methodology gave access to large surface area flakes of atomically-thin graphene and also to flakes of certain transition metals dichalcogenides TMDCs like the superconducting NbSe₂ or MoS₂ among others [12–15]. The technique was implemented in a variety of different ways under the generic name of micromechanical cleavage also known more informally as the *Scotch tape method*.

During the last decade a great variety of different mechanical cleavage methods were developed for the clean deposition of 2D materials on different surfaces. Among them, we can highlight micromechanical exfoliation techniques based on the use of silicone stamps that do not present any glue on their surfaces that could contaminate

the sample as in the case of the scotch tape method [220, 221].

2D superconductivity in thin films and crystals flakes has attracted the attention of many researchers in the last decade [10–18]. For example, superconducting crystals like BSCCO or TaS₂ have been successfully exfoliated down to a single layer and deposited in a substrate in the past [19–21]. In addition, a lot of work has been done trying to induce superconductivity in graphene in contact with a superconductor due to the proximity effect [10, 11, 22–26].

During the thesis, we have used both, the silicone stamp method and the scotch tape method to prepare thin flakes of several superconductors and graphene. The main method employed was the silicon stamp method due to its great simplicity, cleanness and relatively high efficiency. Unfortunately, from our attempts to exfoliate BSCCO crystals with the stamp method, it appeared to be less easily exfoliated by silicone stamps and we had to exfoliate them with the regular scotch tape procedure, obtaining high quality flakes but with some contaminants attached to their surfaces. Finally, we have also transferred a hole sheet of graphene grown on a copper foil on top of a β -Bi₂Pd crystal using a PMMA as a wetting layer. We have investigated several flakes of this superconductors and superconductor/graphene heterostructures with the goal of establish the basis for future experiments to induce superconductivity in graphene via proximity effects.

6.2 Micromechanical exfoliation

In order to transfer microscopic flakes of a macroscopic crystal on top of a substrate we have used Polydimethylsiloxane (PDMS) stamps, a well known viscoelastic material used to exfoliate and transfer crystal flakes in the last years [220, 221].

In this method, first, the PDMS stamp is gently placed on top of the crystal and an small pressure is applied by hand (figure 6.1 a and b). Then, the stamp is removed from the crystal surface with some flakes of the crystal attached to it (figure 6.1 c). Next, the PDMS stamp is placed on top of a the desired substrate applying again an small pressure by hand (figure 6.1 d). Finally, the stamp is removed, leaving small

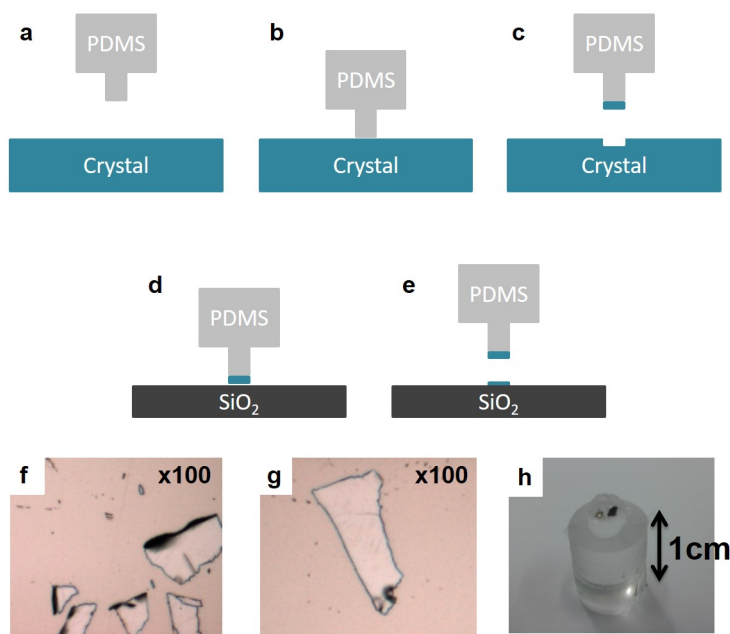


Figure 6.1: In a, b and c, we show the exfoliation process using a PDMS stamp. First, the stamp is gently pushed against the clean crystal surface and then they are separated. Some crystals flakes are attached to the stamp after separate it from the crystal. In d and e, we schematically show the deposition of the crystal flakes on top of a substrate. The PDMS stamp is pushed against the substrate and few flakes are deposited on top of it. In f and g, we show two optical images of arbitrary β -Bi₂Pd flakes deposited on a SiO₂ substrate. In h, we show a real picture of the PDMS stamp with some crystal flakes attached on top.

flakes of the crystal deposited on the substrate (figure 6.1 e). The PDMS stamp facilitates the accommodation of the crystal to the substrate when they are put in contact, thanks to its viscoelastic properties. It is important to note that the PDMS stamps do not have any glue on their surfaces, the crystals remains attached to the stamp due to its viscoelastic properties. Thus, the crystal flakes deposited with this technique are free of the contaminants that typically appear with the regular scotch tape method [222]. An schematic representation of the exfoliation-transfer method is represented in figure 6.1 together with some pictures of the deposited flakes and the stamp.

After deposit the crystal flakes on top of a substrate, they are localized using an optical microscope using a combination of x10, x50 and x100 zoom lenses (figure 6.2 a and b). Then, the sample is moved to our room temperature AFM (RT-AFM) and

the same area is localized using another optical microscope attached to the RT-AFM (figure 6.2 c). Finally, the flakes are measured with the RT-AFM to determine their height.

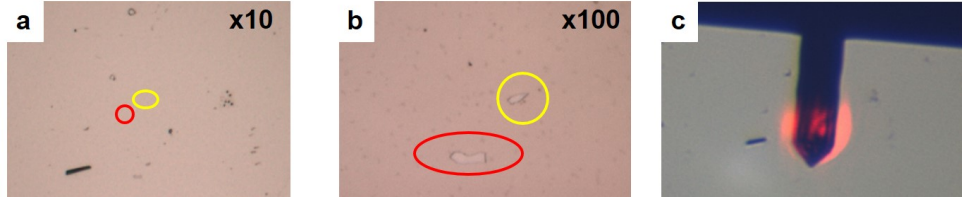


Figure 6.2: In a and b, we show two pictures of two β -Bi₂Pd flakes deposited on top of a SiO₂ substrate with zooms x10 and x100 respectively. In both images the position of the flakes are highlighted with yellow and red circles or ellipses. In c, the same area imaged with an optical microscope attached to our RT-AFM showing the AFM cantilever above the area where the crystal flakes are deposited.

6.2.1 BSCCO on top of SiO₂

We have exfoliated BSCCO crystals down to a few layers and deposited them in a Si substrate with a SiO₂ layer of 300 nm on top, following the same procedure described in the previous section. We have successfully localized and measured a 25 nm thick flake at low temperatures under different magnetic applied fields in our LT-MFM.

6.2.1.1 Moderate magnetic fields

First, we have localized the flake at 10 K and measured its topography and magnetic profile under an applied magnetic field of 100 Oe in the out of plane direction, as is shown in figure 6.3. We have found that the magnetic profile was homogeneous in the whole field of view. This homogeneity can be understood as an increment in the size of the superconducting vortices in very thick samples proposed by Pearl in 1964 [223].

The mayor problem of magnetic image in thin superconducting material is the evolution of the penetration depth as the thickness of the superconductor decreases. The thin-film problem differs from the behavior of currents and vortices in bulk

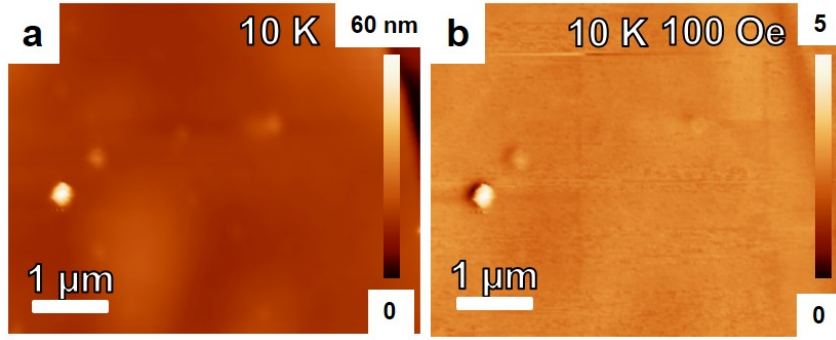


Figure 6.3: In a, we show the topographic image measured at 10 K of a BSCCO flake deposited on a SiO₂ substrate. In b, we show the corresponding magnetic image measured under an applied magnetic field of 100 Oe at 10 K. From the images is clear that the magnetic profile is homogeneous at the BSCCO flake surface.

superconductors by the dominating role of the magnetic stray field outside the film. The interaction between vortices occurs mainly by this stray field, while in bulk superconductors the vortex currents and the vortex interaction are screened and thus decrease exponentially over the length λ [159, 223, 224].

If we consider one vortex in the center of a large circular film with infinite radius and in the limit of zero λ . The point vortex behaves like a magnetic dipole, composed of two magnetic monopoles: one above and another below the film. The magnetic field lines of this point-vortex are straight radial lines, all passing through this point. The magnitude of this magnetic stray field is $\phi_0/2\pi r^2$ above and $-\phi_0/2\pi r^2$ below the film. This jump of the magnetic field component parallel to the film is caused by a sheet current that circulates around the vortex and equals this field difference in size, $J = \phi_0/\mu_0\pi r^2$. This result differs from the strong decay of screening currents of bulk superconductors, $J = \frac{\phi_0}{2\pi\mu_0\lambda^3}K_1(r/\lambda)$ [29], where K_1 is the first order modified Bessel function. In his original paper, Pearl found an effective penetration depth for superconductors when $\lambda < d$ with d the thickness of the superconductor, is equal to $\Lambda = 2\lambda^2/d$. Where Λ is known as the *Pearl penetration depth*.

The expected penetration depth for a BSCCO crystal is ≈ 200 nm [164, 165, 167], in good agreement with our LT-MFM measurements in chapter 4. Therefore, for a flake 25 nm thick, a Pearl penetration depth of $\approx 5 \mu\text{m}$ is expected. From equation 1.16, the intervortex distance at a magnetic field of 100 Oe is expected to be ≈ 500

nm. Thus, superconducting vortices will present a huge overlapping among them at this field, therefore, the magnetic profile on the superconductor is expected to become homogeneous, in good agreement with our measurements.

6.2.1.2 Very low magnetic fields

At very low fields, for example 1 Oe, the intervortex distance is expected to be $\approx 5 \mu\text{m}$. This corresponds to the value of the Pearl penetration depth for our flake. Thus, at 1 Oe, the magnetic profiles of the superconducting vortices will not have a huge overlapping and some corrugation in the magnetic profile of the superconducting flake should be measurable.

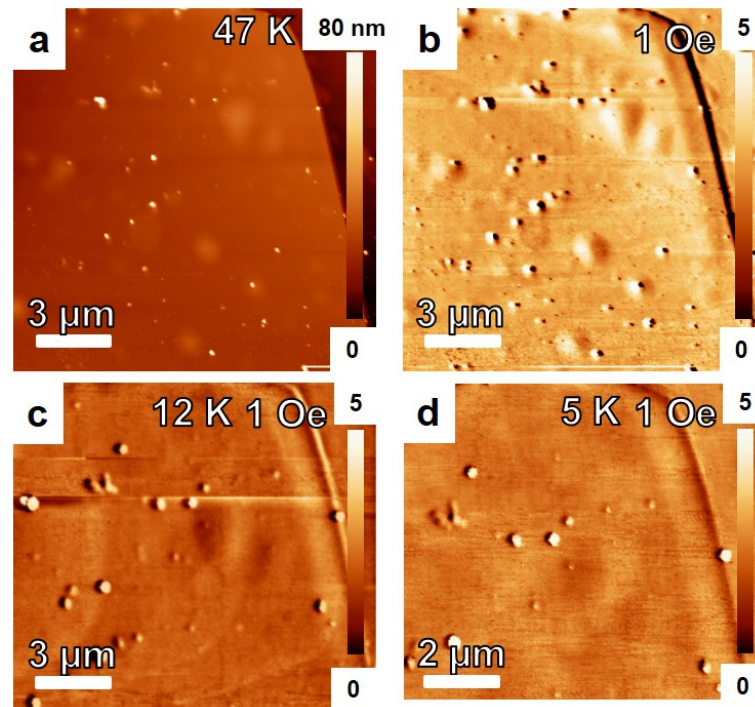


Figure 6.4: In a, we show the topographic image measured at 47 K on a BSCCO flake deposited on a SiO_2 substrate, showing some contamination deposited on the flake during the deposition. In b, c and d, the corresponding magnetic images measured under an applied magnetic field of 1 Oe at 47, 12 and 5 K respectively. The magnetic profile is homogeneous at the BSCCO surface at the three temperatures. There are only few inhomogeneities at the positions of the contaminations, probably related with the electrostatic long range interaction between them and the tip.

For these reasons, we have imaged again the magnetic profile of the same flake at very low applied magnetic fields (1 Oe) and different temperatures. Our results are summarized in figure 6.4 and 6.5. In figure 6.4, we show three magnetic images with a magnetic field of 1 Oe at 47, 12 and 5 K together with the corresponding topographic image. As it is clear from the images, the magnetic profiles are homogeneous in the whole field of view at the three temperatures, presenting only small corrugations related with the contamination deposited on top of the flake during the exfoliation. The homogeneity of the magnetic profiles point that the overlap between vortices is strong enough to avoid single vortex resolution with our LT-MFM even at this very low fields. Another possibility is that the electrostatic interaction with the contamination deposited on the crystal dominates the interaction with the tip and therefore no vortex resolution can be achieved.

We have also measured a region where the edge of the BSCCO flake and the SiO₂ substrate are clearly visible to determine if there is any measurable screening of the magnetic field in the superconducting flake. Our results are presented in figure 6.5. The magnetic image in figure 6.5 shows that the magnetic profile is homogeneous in the whole field of view. There is not visible difference between the region occupied by the superconductor with respect to the substrate. There is only some contrast in the magnetic image at the positions of the edge of the BSCCO flake and of a longitudinal topographic feature in the right part of the image, due to the electrostatic interaction with the AFM tip at this locations.

Unfortunately, the screening of the magnetic field in the superconducting flake with respect to the substrate is below our experimental resolution at this thickness. Thus, we were not able to measure the existence of Pear vortices or another superconducting effect related with the thickness in our experiments on BSCCO. Nevertheless, we were able to deposit small flakes of this material and localize them at low temperatures opening the possibility for further experiment in BSCCO flakes of different thicknesses in the future.

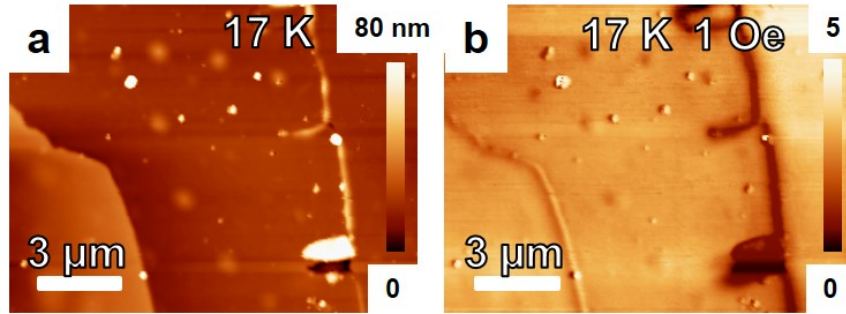


Figure 6.5: In a, we show the topographic image measured at 17 K of the edge of the BSCCO flake (left) and the SiO₂ substrate (right). In b, we show the corresponding magnetic image measured under an applied magnetic fields of 1 Oe at 17 K. From the image is clear that the magnetic profile is homogeneous at the BSCCO flake and at the substrate, indicating that there is not a measurable difference in the magnetic field between the two places. There is only contrast at the edge of the flake and at the position of longitudinal topographic feature on the right of the image due to the electrostatic interaction with the tip.

6.2.2 β -Bi₂Pd on top of SiO₂

As it was presented in chapter 3, β -Bi₂Pd is a layered compound that can be easily exfoliated using the regular scotch tape method. In contrast with another layered crystals that were successfully exfoliated down to a single monolayer, the layers of β -Bi₂Pd are not weakly coupled via Van de Waals interactions but strongly coupled via covalent bonds [140]. Nevertheless, a recent theoretical work has establish that the bonds between Bi layers in the crystal are much weaker that the Bi-Pd or Pd-Pd bonds and therefore, the crystal is expected to cleave in this planes. This theoretical calculation was demonstrated experimentally by [6] and [7] and also corroborated in this thesis in chapter 3. We have tried to exfoliate our β -Bi₂Pd crystals to the minimum possible thickness to open the possibility of studying superconductivity in the 2D limit in this non VdW superconductors.

6.2.2.1 Exfoliation down to few tens of nanometers

Using the PDMS stamp method described before, we have successfully deposited several β -Bi₂Pd flakes of different thickness on SiO₂ substrates, as is shown in figure 6.6.

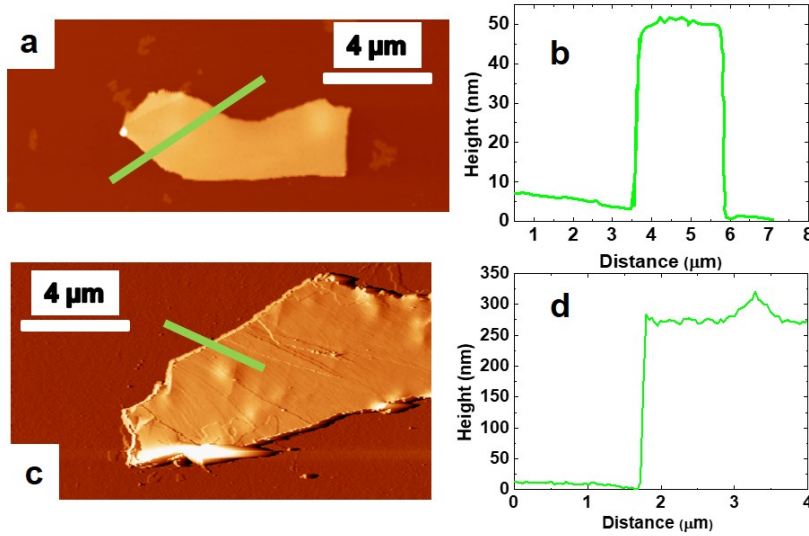


Figure 6.6: In the left panel, two AFM images of two β -Bi₂Pd flakes deposited on top of a SiO₂ substrate. In the right panel, the corresponding topographic profiles marked as green lines on the images in the left. The images show the possibility of deposit flake of β -Bi₂Pd of a great variate of thicknesses.

In the image, two examples of β -Bi₂Pd flakes are presented, one thick flake of several hundreds of nanometers and a thin flake of just some tens of nanometers. The thin flake reveals that it is possible to exfoliate a β -Bi₂Pd single crystal down to very small thicknesses where changes in the superconducting behavior are expected, for example, the formation of Pearl vortices introduced in the previous sections. Unfortunately, the density of thin flakes of β -Bi₂Pd that we achieved with the stamp method was not enough to allow us to localize one of these flakes at low temperatures. The thermal drift of our LT-MFM has prevented us from locating a β -Bi₂Pd flake at low temperature despite all our efforts. More work is needed in this direction to investigate the superconducting behavior of β -Bi₂Pd thin flakes. In particular, improve the deposition technique to increase the ratio of success and the density of flakes is of particular interest in this system.

6.2.3 Graphene on top of β -Bi₂Pd

We have successfully exfoliated several graphene and few-layers-graphene (FLG) flakes and deposited them on top of a single crystal of the superconductor β -Bi₂Pd. The flakes were characterized with our RT-AFM after localize them with an optical microscope. An example is shown in figure 6.7, where three pictures at different zooms are presented together with a topographic AFM image. The AFM topographic measurements show a combination of several FLG flakes at the edge of the big graphite flake.

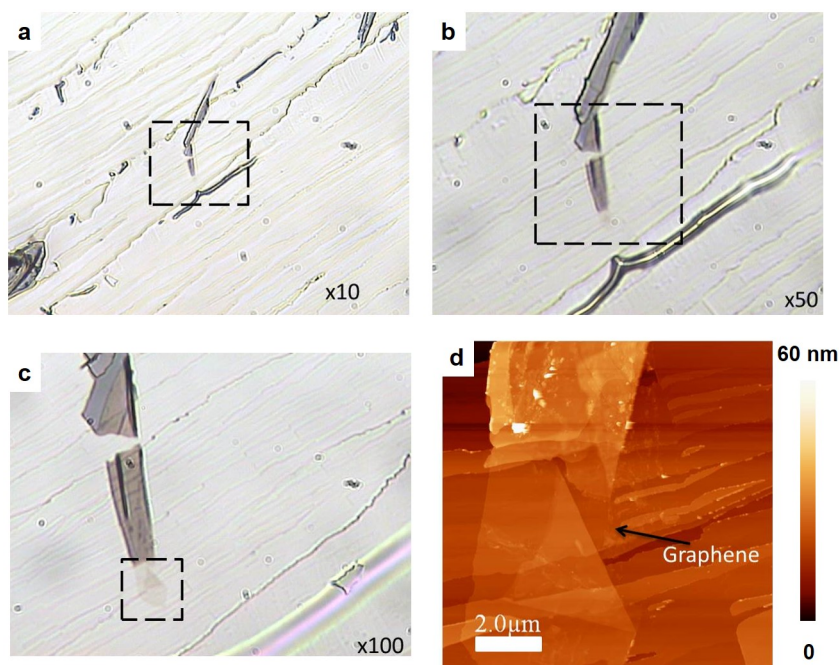


Figure 6.7: In a b and c, three optical images with zooms x10, x50 and x100, used to localize the graphene flakes deposited on to of the β -Bi₂Pd crystal. In d, the corresponding topographic image of the area highlighted with a black square in c. It shows flakes with different thicknesses with a graphene flake signaled with a black arrow

6.2.3.1 Friction measurements

It is know that graphene has lubricant properties [225]. This, allows to improve the localization of the flakes via friction images, as the contrast in friction images between graphene, FLG and the substrates is often huge [226]. For this reason, we

have performed a combination of topographic and friction measurements at the same flake to localize the different graphene or FLG flakes.

The physical basics of the friction measurements in AFM are as follows. When scanning in the contact mode at a constant force, besides the cantilever's deflection in the normal direction, an additional torsion bending of the cantilever takes place. When moving over a flat surface with zones of different friction factors, the angle of torsion will be changing in every new zone. This allows measuring of the local friction force. If the surface is not absolutely flat, such an interpretation is complicated. To distinguish zones of different friction and relief influence one can utilize second pass on the same line in opposite direction (for a detail study of friction measurements in AFM see reference [98]).

In figure 6.8 b, there are several FLG flakes on the right of the image that are not easily localized in the topographic image. They appear as a clear dark contrast in the friction image. In our case, we have not found mayor differences between friction images on graphene and FLG on top of the β -Bi₂Pd crystal, probably due to surface contamination during the exfoliation-transfer method. Friction images do not allow us to distinguish between graphene or FLG flakes on top of β -Bi₂Pd but is still the best technique to quickly localize FLG flakes on top of β -Bi₂Pd that are not visible in the topographic image.

6.2.3.2 Kelvin Probe Microscopy (KPM) measurements

We have used a different approach to establish a experimental procedure that unambiguously distinguish between graphene and FLG flakes deposited on β -Bi₂Pd. We have performed Kelvin Probe Microscopy (KPM) measurements on the same flake to characterize the surface potential difference between the β -Bi₂Pd substrate, graphene and FLG flakes. KPM is a tool that enables nanometer-scale imaging of the surface potential of a broad range of materials. KPM measures the Surface Potential Difference (SPD) between a conducting tip and the sample.

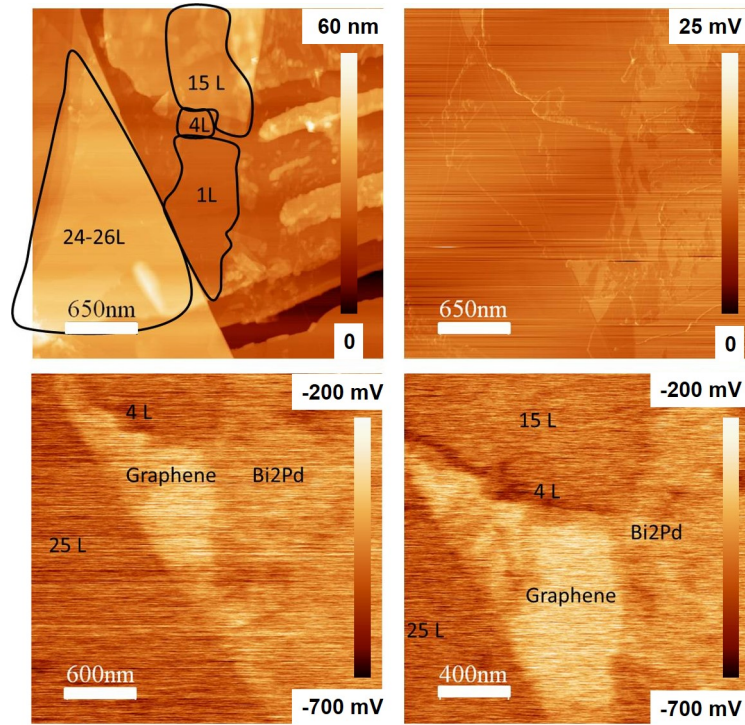


Figure 6.8: In a, a topographic image of some graphene flakes with different thicknesses, from 25 to 1 layers. The area occupied by different flakes is approximately signaled with black lines. In b, a friction map measured with our RT-AFM showing two different regions, one on top of the graphene flakes and another corresponding to the β -Bi₂Pd crystal. In c and d two KPM images revealing a clear contrast between the β -Bi₂Pd crystal, the single layer graphene flake and thicker graphene flakes.

$$V_{SPD} = \frac{\phi_{tip} - \phi_{sample}}{-e} \quad (6.1)$$

Where ϕ_{tip} and ϕ_{sample} are the work functions of the tip and the sample (for a more detailed study of the KPM see reference [98]).

In figure 6.8 c and d, KPM maps at the same areas that the topographic and friction images are shown. In this case, is clear that the difference in surface potential between graphene, different FLG flakes and β -Bi₂Pd is measurable. More important, the surface potential in a single layer graphene is smaller that the surface potential of the β -Bi₂Pd crystal and the surface potential of thicker flakes is higher that in the β -Bi₂Pd. This allows to unambiguously distinguish between single layer graphene and thicker flakes by simply comparing the value of the surface potential of a flake

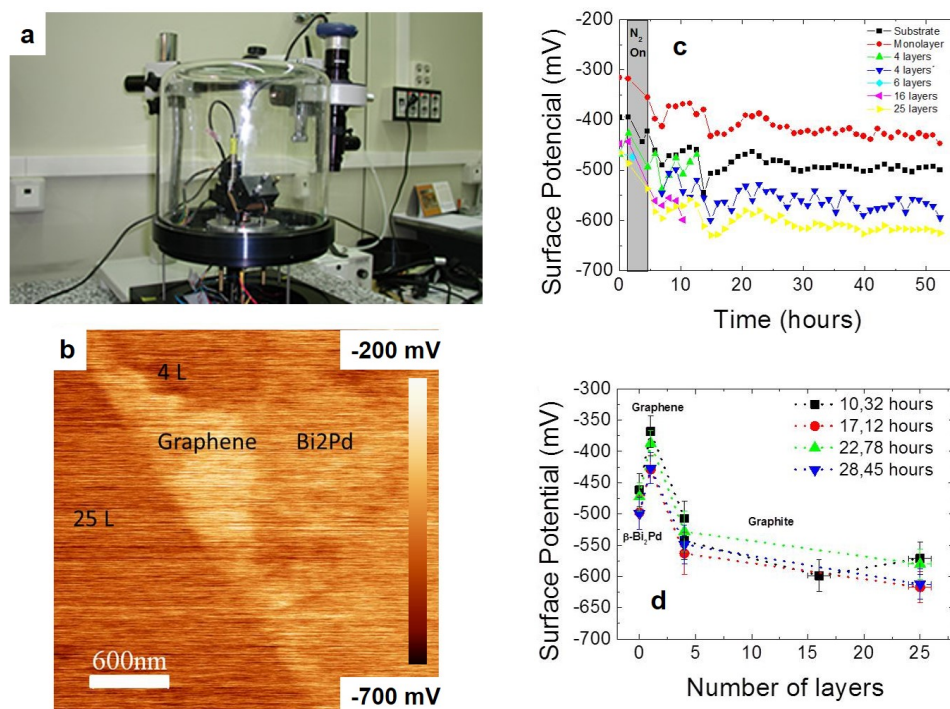


Figure 6.9: In a, our RT-AFM with the crystal chamber used to control the ambient humidity with the N₂ flux. In b, a KPM image measured during the experiment as an example. In c, the evolution of the surface potential for the substrate (β -Bi₂Pd), graphene and FLG after decrease the humidity of the sample chamber by applying a constant current of N₂, in the image. The moment when the N₂ flux was turned on is signaled in gray. In d, the evolution of the surface potential of the same system as a function of the number of layers of graphene for four different times after decrease the humidity. Both plots show that the surface potential difference between the β -Bi₂Pd, the graphene and the FLG remains almost constant with the time.

and the β -Bi₂Pd substrate.

KPM surface potential measurement are often affected by adsorbates on the surfaces of study. As Oxygen, Hydrogen and another adsorbates present in the atmosphere attached to the graphene and β -Bi₂Pd surfaces. For this reason, we have sealed our AFM in a crystal chamber with a continuous flux of Nitrogen for several hours. Nitrogen flux is expected to dramatically decrease the humidity inside the crystal chamber and partially remove the adsorbates from the sample. We have maintained the Nitrogen flux for 50 hours, performing several KPM measurement during the process. We have found that the Surface potential of all FLG, graphene

and β -Bi₂Pd decreases with the time and approaches an stable value after turn on the N₂ flux. After reach a stable value, the difference between the surface potential of the different flakes and the β -Bi₂Pd substrate, is almost the same than before, showing that this technique allow to localize graphene flakes even at ambient conditions with contaminants deposited on the surface. The results of this experiment are summarized in figure 6.9.

Our results shows that KPM is the best experimental microscopic technique to individual localize graphene flakes on top of the superconductor β -Bi₂Pd. By contrast friction maps have probed to be a valuable tool to localize graphene and FLG flakes on top of β -Bi₂Pd but no dot allow to unambiguously determine the number of graphene layers.

6.3 Electrochemical transfer of graphene on top of β -Bi₂Pd

We have transfered a graphene sheet of $\approx 1 \text{ cm} \times 1 \text{ cm}$ area on a β -Bi₂Pd substrate. The graphene sheet was grown in high vacuum on a copper foil by Jon Azpeitia at the laboratory of Prof. Jose Martin Gago (for a detailed description of the graphene growth see reference [227]).

To transfer the graphene sheet from the copper foil to the β -Bi₂Pd substrate we have use the common electrochemical transfer method described in [229, 230]. In this method, first, the graphene is covered by a PMMA layer via spin coating (figure 6.10 a). Then, the coper foil with the graphene and the PMMA is immersed in a solution of potassium chloride at a rate of 1 mm/s (figure 6.10 b). The copper is negatively polarized up to 5 V with respect to a carbon anode. When the graphene/copper cathode is negatively polarized, hydrogen bubbles appear at the graphene/copper interface due to the reduction of water molecules and allow graphene to gently detach (figure 6.10 c). Then, the graphene/PMMA layer is placed on top of the substrate (figure 6.10 d) and the PMMA layer is dissolved with acetone to obtain free graphene deposit on top of the substrate (figure 6.10 e and f).

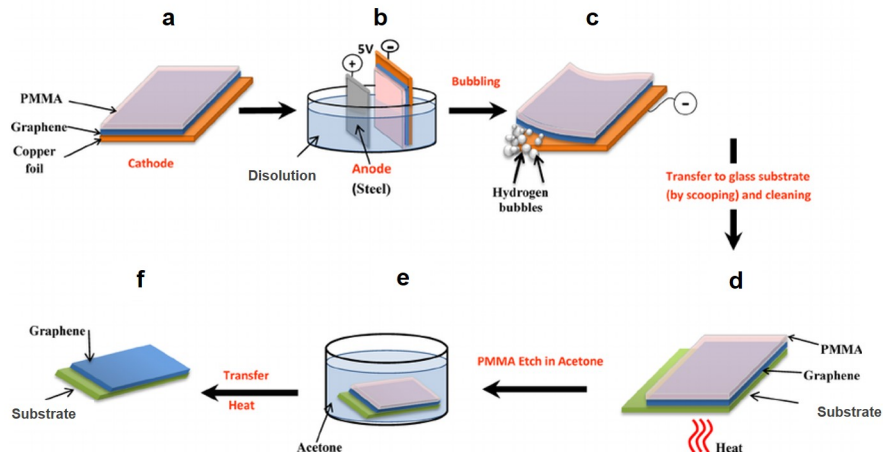


Figure 6.10: Schematic representation of the graphene sheet transfer method. The PMMA layer is presented in light pink, the graphene in blue, the copper foil in orange and the substrate in green. First, the graphene is covered by a PMMA layer via spin coating (a). Then, the copper foil with the graphene and the PMMA is immersed in a solution of potassium chloride at a rate of 1 mm/s (b). The copper is negatively polarized with respect to a carbon anode. When the cathode is negatively polarized, hydrogen bubbles appear at the graphene/copper interface due to the reduction of water molecules and allow graphene to gently detach (c). Then, the graphene/PMMA layer is placed on top of the substrate (d) and the PMMA layer is dissolved with acetone and heated up to 70 ° to obtain free graphene deposit on top of the substrate (e and f) Adapted from [228].

6.3.1 Characterization at room temperature

After transfer the graphene on the β -Bi₂Pd crystal, we have measured it in our RT-AFM to ensure the success of the transfer. We have found that the whole surface of the crystal is covered by graphene showing its characteristic wrinkles, pointing the big success of the transfer (figure 6.11).

6.3.2 Characterization at low temperatures

We have measured the vortex lattice in the same sample at low temperatures in or LT-AFM. Figure 6.12 d shows the topographic image of sample at 2 K. It reveals the characteristic wrinkles of the graphene layers and some steps and terraces of the β -Bi₂Pd crystal. Figure 6.12 e shows the magnetic image measured at the same area

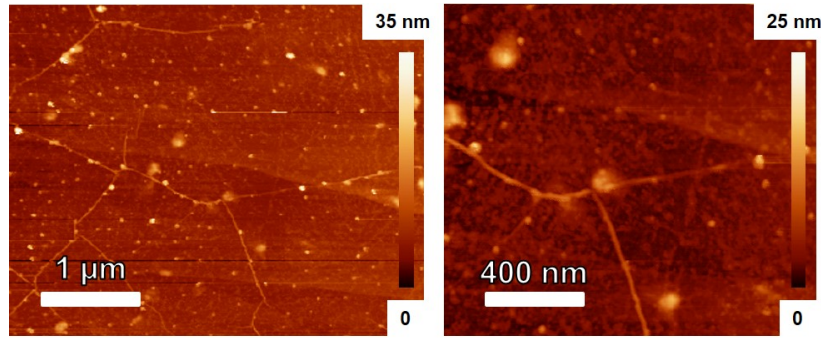


Figure 6.11: In the figure, we show two topographic images measured at RT with our RT-AFM in a sample with graphene deposited on top of a β -Bi₂Pd crystal. In both images are visible the characteristic wrinkles of the graphene pointing the success of the transfer.

at 2 K and under an applied magnetic field of 200 Oe in the out of plane direction. The image shows an hexagonal vortex lattice in the whole field of view, revealing that it is possible to observe the vortex lattice even when graphene is present. We have not found any any visible effect due to pinning or screening of the superconducting vortices on the graphene layer.

Further experiments are needed to determine if there is induced superconductivity or not on the graphene due to the proximity effect as it was previously reported by [11] in a Pr_{2-x}Ce_xCuO₄ graphene heterostructure. In particular STM measurements in the same β -Bi₂Pd/graphene sample are of particular interest to reveal if there is a superconducting gap opening at the graphene.

6.4 Conclusions

In conclusion, we have successfully exfoliated several superconducting crystals and graphene and deposit them in different substrates using a combination of PDMS silicon stamps and the regular scotch tape method. We have investigated three different system with this method, BSCCO on SiO₂, β -Bi₂Pd on SiO₂ and graphene and FLG on β -Bi₂Pd.

In the case of BSCCO flakes deposited in SiO₂, we were able to measure one thick

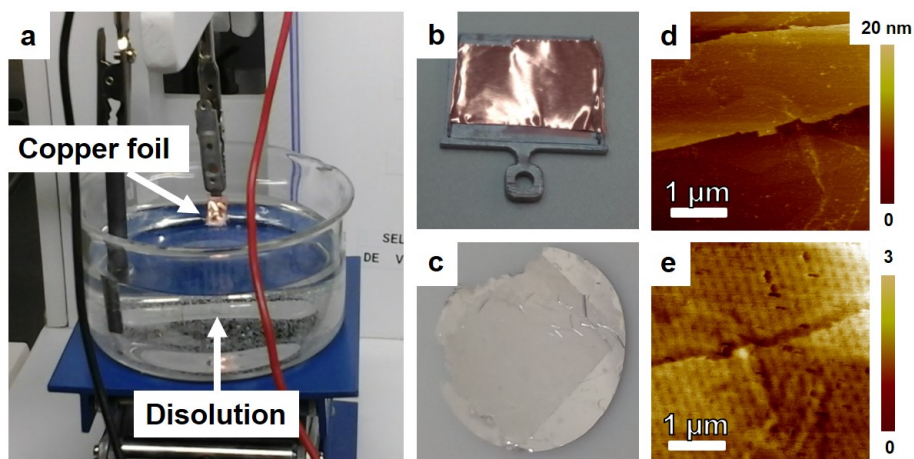


Figure 6.12: In a, the experimental set-up to transfer the PMMA/graphene layer to an arbitrary substrate. In b, a photograph of the copper foil with the graphene deposited on top. In c, a photograph of the β -Bi₂Pd crystal with the graphene/PMMA layer deposited on top. In d and e, topographic and magnetic images of the transferred graphene layer on top of the β -Bi₂Pd crystal measured at the same area at 2 K and an applied magnetic field of 200 Oe perpendicular to the surface. The topographic image reveal the characteristic wrinkles of the graphene sheet and the steps and terraces of the β -Bi₂Pd crystal. The magnetic image shows the ordered hexagonal vortex lattice even with the presence of the graphene layer.

flake at low temperatures and characterize its magnetic profile in the superconducting state at different magnetic fields and temperatures. We have found that the screening of the magnetic field in these flakes is below our experimental resolution at moderate (100 Oe) and very low fields (1 Oe).

For β -Bi₂Pd flakes deposited on SiO₂, we were able to exfoliate and deposit flakes of this system for the very first time, down to some tens of nanometers. This result opens the possibility to study the superconducting behavior in the 2D limit in this system in the future.

We have also transferred graphene and FLG flakes on top of a β -Bi₂Pd single crystal and developed an experimental procedure to unambiguously localize graphene and FLG flakes on top of β -Bi₂Pd using a combination of friction and KPM measurements with an AFM. We have also probed that this method allows to localize the flakes at ambient and at low humidity conditions.

Finally, we have transferred a $\approx 1 \text{ cm} \times 1 \text{ cm}$ graphene sheet on top of a $\beta\text{-Bi}_2\text{Pd}$ single crystal with the electromechanical transfer method and measured the vortex lattice of the $\beta\text{-Bi}_2\text{Pd}$ crystal at the superconducting state at 2K through the graphene layer. This experiment, opens the possibility of characterize this heterostructure in future experiments to determine if there is a gap opening in the graphene in this situation.

General conclusions

In this project we have used magnetic force microscopy at low temperatures to study three different superconducting compounds. We have focused our research in the study of superconducting vortices and their manipulation. We have also studied the decomposition in superconducting and ferromagnetic domains in a $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ single crystal and the exfoliation and deposition of several 2D systems. Regarding the experimental system, we have used a set-up that allows us to perform AFM-MFM measurements between 1.8 K and 300 K, applying magnetic fields in any direction of the space up to 5 T in the Z direction and 1.2 T in the X and Y directions.

With this set up, we have characterized for the first time the hysteresis cycle of MFM commercial probes as a function of the temperature from 1.8 to 300 K. We have also found that the MFM images of superconducting vortices show star-like features at vortex positions when the MFM tip is magnetized below its coercive field. Interestingly, at some magnetization fields, below the coercive field of the tips, the lateral resolution is improved with respect to tips magnetized above their coercive field.

We have observed two different regimes in the patterns of pinned superconducting vortices at low magnetic fields in the single gap superconductor $\beta\text{-Bi}_2\text{Pd}$. We have shown that lines of vortices form at defects due to pinning at very low magnetic fields, while at higher fields the vortex lattice acts as a whole, leaving a regular hexag-

onal lattice even below H_{c1} . Crystalline strain close to defects determines the vortex arrangements at low fields and leads to sizable modifications of the local superconducting screening properties, as shown by the measured increase in the penetration depth λ close to defects.

We have studied the crossing lattice in a Bi-2212 single crystal at low temperatures and low fields. We have successfully manipulated an arbitrary arrangement of PVs and the crossing lattice of JVs and PVs. We have measured the dependence of the PV movement with the angle between the in-plane magnetic field and the scan of the MFM. We have measured the necessary force to manipulate isolated PVs and PVs trapped on JVs. We were able to cross three JVs. To our knowledge, ours is the first work showing manipulation of the crossing lattice in a superconductor.

We have studied the effect of strain in a $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.35})_2\text{As}_2$ single crystal from the microscopic point of view. We have imaged the coexistence of tetragonal/orthorhombic domain walls below the strains mediated transition at $T_{s,N}(\epsilon)$. Below the superconducting critical temperature of free standing samples, we have measured the formation of diamagnetic domains coinciding with the tetragonal domains. We have associated the diamagnetic domains with the superconducting transition of the remaining tetragonal phase. We have characterized their evolution with the applied magnetic field and the temperature. At very low magnetic fields we observe sometimes linear diamagnetic structures in the orthorhombic phase that might join elongated tetragonal domains, suggesting that such a coupling between elongated domains can indeed happen in some parts of the sample. To our knowledge, ours is the first experimental work showing phase separation associated to strain below T_C in pnictides.

We have successfully exfoliated several superconducting crystals and graphene and deposit them in different substrates using a combination of PDMS silicon stamps and the regular scotch tape method. We have investigated three different system with this method, BSCCO on SiO_2 , $\beta\text{-Bi}_2\text{Pd}$ on SiO_2 and graphene and FLG on $\beta\text{-Bi}_2\text{Pd}$.

In the case of BSCCO flakes deposited in SiO_2 , we were able to measure one thick flake at low temperatures and characterize its magnetic profile in the superconducting

state at different magnetic fields and temperatures.

For β -Bi₂Pd flakes deposited on SiO₂, we were able to exfoliate and deposit flakes of this system for the very first time, down to some tens of nanometers. This results open the possibility to study the superconducting behavior in the 2D limit in this system in the future.

We have also transferred graphene and FLG flakes on top of a β -Bi₂Pd single crystal and developed an experimental procedure to unambiguously localize graphene and FLG flakes on top of β -Bi₂Pd using a combination of friction and KPM measurements with an AFM. We have also probed that this method allows to localize the flakes at ambient and at low humidity conditions.

Finally, we have transferred a $\approx 1 \text{ cm} \times 1 \text{ cm}$ graphene sheet on top of a β -Bi₂Pd single crystal with the electromechanical transfer method and measured the vortex lattice of the β -Bi₂Pd crystal at the superconducting state at 2K through the graphene layer. This experiment, opens the possibility of characterizing this heterostructure in future experiments to determine if there is a gap opening in the graphene in this situation.

Conclusiones generales

En este proyecto hemos usado la microscopía de fuerzas magnéticas a bajas temperaturas para estudiar tres compuestos superconductores distintos. Hemos centrado nuestra investigación en el estudio de los vórtices superconductores y en su manipulación. Hemos estudiado además, la descomposición en demonios superconductores y ferromagnéticos en un monocristal de $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ y la exfoliación de varios sistemas 2D. Acerca del sistema experimental, hemos usado un set-up que nos permite realizar medidas de AFM/MFM entre 1.8 K y 300 K, aplicando campos magnéticos en cualquier dirección del espacio hasta 5 T en la dirección Z y 1.2 T en las direcciones X e Y.

Con este set-up, hemos caracterizado por primera vez el ciclo de histéresis de una punta comercial de MFM en función de la temperatura, desde 1.8 K hasta 300 K. Además, hemos encontrado que las imágenes de MFM de vórtices superconductores muestran estructuras en forma de estrella en la posición de los mismos cuando la punta de MFM está magnetizada por debajo de su campo coercitivo. Es interesante que a algunos campos de magnetización, por debajo del campo coercitivo de la punta, la resolución lateral mejora con respecto a puntas magnetizadas por encima del campo coercitivo.

Hemos observado dos regimenes distintos en la distribución de los vórtices superconductores en $\beta\text{-Bi}_2\text{Pd}$. Hemos mostrado que a campos magnéticos muy bajos, estos

se distribuyen en forma de líneas en los defectos, debido al pinning, mientras que a campos más altos, la red de vórtices actúa como un todo, distribuyéndose en una red hexagonal incluso por debajo de H_{C1} . La tensión cristalográfica en los defectos determina la distribución a campos bajos y lleva a modificaciones de las propiedades superconductoras locales, como se muestra por el aumento medido en la longitud de penetración λ cerca de los defectos.

Hemos estudiado la red cruzada en un monocristal de Bi-2212 a bajas temperaturas y bajos campos magnéticos. Hemos conseguido manipular una distribución aleatoria de PVs y la red cruzada de JVs y PVs. Hemos medido la dependencia del movimiento de los PVs con el ángulo del campo magnético en el plano y el eje de medida el MFM. Hemos medido la fuerza necesaria para manipular PVs aislados y PVs decorando JVs. Hemos cruzado tres JVs. En nuestro conocimiento, este es el primer trabajo mostrando manipulación de la red cruzada en un superconductor.

Hemos estudiado el efecto de la tensión en un monocristal de $\text{Ca}(\text{Fe}_{0.965}\text{Co}_{0.035})_2\text{As}_2$ desde el punto de vista microscópico. Hemos medido la coexistencia de dominios tetragonales y ortorrómbicos por debajo de la transición producida por la presencia de tensión en el cristal, $T_{S,N}(\epsilon)$. Por debajo de la transición superconductor de muestras sin tensión externa, hemos medido la formación de dominios diamagnéticos coincidiendo con la localización de los dominios tetragonales. Hemos asociado los dominios diamagnéticos con la transición superconductor de la fracción de dominios tetragonales aún presente en la muestra. Hemos caracterizado su evolución con la temperatura y el campo magnético. A campos muy bajos, hemos observado que en algunos casos estructuras diamagnéticas unen distintos dominios diamagnéticos, sugiriendo que el acoplamiento entre dominios diamagnéticos puede ocurrir en algunas partes de la muestra. En nuestro conocimiento, el nuestro, es el primer trabajo experimental que muestra separación de fases asociada con tensión por debajo de T_C en superconductores basados en hierro.

Hemos exfoliado distintos cristales superconductores y grafeno y los hemos depositado en distintos sustratos usando una combinación de sellos de PDMS y cinta adhesiva. Hemos investigado tres sistemas diferentes con estos métodos: BSCCO en SiO_2 , $\beta\text{-Bi}_2\text{Pd}$ en SiO_2 y grafeno en $\beta\text{-Bi}_2\text{Pd}$.

En el caso de los flakes de BSCCO depositados en SiO_2 , fuimos capaces de medir un flake fino a bajas temperaturas y de caracterizar su perfil magnético en el estado superconductor a distintos campos magnéticos y temperaturas.

Para los flakes de $\beta\text{-Bi}_2\text{Pd}$ en SiO_2 , fuimos capaces de exfoliar y depositar flakes de este sistema por primera vez hasta espesores de decenas de nanómetros. Este resultado abre la posibilidad a estudiar el comportamiento superconductor en el límite 2D de este sistema en el futuro.

También hemos transferido grafeno y FLG flakes en un monocristal de $\beta\text{-Bi}_2\text{Pd}$ y desarrollado un método experimental para localizar sin ambigüedad grafeno y FLG depositados en $\beta\text{-Bi}_2\text{Pd}$, usando una combinación de medidas de fricción y KPM con AFM. Además, hemos probado que este método funciona tanto en condiciones ambientales normales como en condiciones de muy baja humedad.

Finalmente, hemos transferido una lámina de $1\text{ cm} \times 1\text{ cm}$ de grafeno en un monocristal de $\beta\text{-Bi}_2\text{Pd}$ mediante el método de transferencia electromecánica y hemos medido la red de vórtices superconductores del $\beta\text{-Bi}_2\text{Pd}$ en el estado superconductor a 2 K a través del grafeno. Este experimento abre la posibilidad de caracterizar esta heteroestructura en futuros experimentos y determinar si se induce un gap superconductor en el grafeno.

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