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Pseudogap in cuprate and organic superconductors

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We study the pseudogap present in cuprate and organic superconductors. We use the dynamical cluster approximation (DCA), treating a cluster embedded in a bath. As the Coulomb interaction is increased, cluster-bath Kondo states are destroyed and bound cluster states formed. We show that this leads to a pseudogap. Due to weaker coupling to the bath for the anti-nodal point, this happens first for this point, explaining the $\mathbf{k}$ -dependence of the pseudogap. The pseudogap can be understood in terms of preformed d -wave pairs, but it does not prove the existence of such pairs.

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Experiments show a pseudogap at the Fermi energy in the normal phase in both cuprate[1] and organic[2] superconductors under certain circumstances. For cuprates the pseudogap forms for $\mathbf{k} = (\pi, 0)$ while for $\mathbf{k} = (\pi/2, \pi/2)$ the spectrum has a peak[3]. It is crucial for the understanding of these systems to trace the origin of the pseudogap.

Calculations using embedded cluster methods[4], e.g., the dynamical cluster approximation (DCA) reproduce a $\mathbf{k}$ -dependent pseudogap for the Hubbard model[6–13]. Ferrero *et al.*[14] discussed small embedded clusters in terms of a transition between a state where the cluster orbitals form Kondo-states with the bath and a state where the cluster forms a bound state and a pseudogap. However, the strong $\mathbf{k}$ -dependence of the pseudogap in cuprates was not discussed. More recently, the pseudogap has been interpreted as a momentum-selective metal-insulator transition[11]. There has been much work relating the pseudogap to preformed superconducting pairs[15], which have have not reached phase coherence and superconductivity at the temperature T studied. On the other hand it has been argued that the pseudogap and superconductivity phases compete[16].

The DCA treats a cluster of N_c atoms embedded in a bath. Guided by the smallest ($N_c = 4$) cluster giving a pseudogap, we construct a very simple two-site two-orbital model. For a small Coulomb interaction U , the cluster orbitals form Kondo states with the bath. As U is increased, the Kondo states are destroyed and a bound state is formed on the cluster. We show that if this state is nondegenerate, this leads to a pseudogap. By comparing correlation functions, we find that a $N_c = 8$ DCA calculation behaves in a similar way. Due to the weak dispersion around $\mathbf{k} = (\pi, 0)$, the coupling of this cluster $\mathbf{k}$ -vector to the bath is much weaker than for $\mathbf{k} = (\pi/2, \pi/2)$. Then the Kondo state for $(\pi/2, \pi/2)$ is destroyed for a larger U , giving the $\mathbf{k}$ -dependent pseudogap. We show why the pseudogap is lost for a frustrated electron-doped cuprate. We find that the pseudogap can be interpreted in terms of d -wave supercon-

ducting pairs. The pseudogap hinders the propagation of pairs and hurts superconductivity.

Below we study the Hubbard model

$$H = \sum_{ij\sigma} t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.}) + U \sum_i n_{i\uparrow} n_{i\downarrow} - \mu \sum_{i\sigma} n_{i\sigma}, \quad (1)$$

where $c_{i\sigma}^\dagger$ creates an electron on site i with spin σ , $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$, $t_{ij} = t$ if i and j are nearest neighbors, t' if they are second nearest neighbors and zero otherwise. Here we use $t = -0.4$ eV and $t' = 0$ or $-0.3t$. U is a Coulomb integral and μ the chemical potential.

We solve this model using DCA, and discuss the cluster in terms of its N_c $\mathbf{K}$ -states. With $N_c = 8$ there are both nodal [$\mathbf{K} = (\pm\pi/2, \pm\pi/2)$] and antinodal [$(\pi, 0), (0, \pi)$] states. For *isolated* clusters, the nodal and antinodal spectra are identical for $N_c = 8$ and 16. This is due to a symmetry for these values of N_c [5]. Since DCA calculations with $N_c = 8$ and 16 give a pseudogap for the antinodal point, this must then be due to the coupling to the bath. To test this we have switched the baths. The pseudogap then indeed appears at the nodal point instead of the antinodal point. An important clue is that the coupling of the bath to the antinodal point is weaker by a factor of three to four, due to the weaker dispersion of the band around this point. For a self-consistent DCA calculation, we find that the bath also tends to have a pseudogap. Is then the pseudogap in the spectrum mainly due to the pseudogap in the bath or to cluster properties? We find that performing only one iteration, with a fully metallic bath, the pseudogap is reproduced, although for a larger U value. Below we then focus on the first iteration. The key issue is then to understand the associated multi-orbital quantum impurity. We assign the pseudogap to the weakness of the antinodal coupling and internal electronic structure of the cluster.

The smallest embedded cluster with a pseudogap is $N_c = 4$. $\mathbf{K} = (0, 0)$ is mainly occupied and $\mathbf{K} = (\pi, \pi)$ mainly unoccupied. We can then focus on $\mathbf{K} = (\pi, 0)$ and $(0, \pi)$, which mainly determine the dynamics. For simplicity, we first only couple each $\mathbf{K}$ -state to one bath

state. This leads to a two-site model with a two-fold orbital degeneracy. This model has the essential features of a pseudogap and we can study the origin in detail. We introduce a hopping integral V between each cluster orbital, labeled $1c$ or $2c$, and its bath site orbital, labeled $1b$ or $2b$. On the cluster site c there is a direct Coulomb integral $U_{xx} \equiv U - \Delta U$ between spin up and down states of the same orbital, $U_{xy} \equiv U + \Delta U$ between two different orbitals and an exchange integral K . The lowest $S_z = 0$ states on the isolated c site are

$$|1\mp\rangle = \frac{1}{\sqrt{2}}(c_{1c\uparrow}^\dagger c_{1c\downarrow}^\dagger \mp c_{2c\uparrow}^\dagger c_{2c\downarrow}^\dagger); \quad E_{1\mp} = U_{xx} \mp K \quad (2)$$

$$|2\mp\rangle = \frac{1}{\sqrt{2}}(c_{1c\uparrow}^\dagger c_{2c\downarrow}^\dagger \mp c_{2c\uparrow}^\dagger c_{1c\downarrow}^\dagger); \quad E_{2\mp} = U_{xy} \mp K,$$

where $c_{ic\uparrow}^\dagger$ creates a spin up electron in level i on site c and $E_{i\mp}$ are the energies of the states. The state $|2-\rangle$ is a triplet and the other states are singlets. To simulate the lowest three states of an isolated cluster with $N_c = 4$, we identify $1c$ with $(\pi, 0)$ and $2c$ with $(0, \pi)$ and put $\Delta U = 0.03U$ and $K = 0.1U$. The lowest state in the $N_c = 4$ cluster and the model cluster are both singlets. $U_{xx} < U_{xy}$ may seem surprising, but is related to the influence of the $(0, 0)$ and (π, π) orbitals, neglected in the model cluster. Alternatively, we can choose $U_{xx} > U_{xy}$. Then the lowest model cluster state is a triplet and, we will see, the physics is completely changed.

For the two-level, two-orbital model we have calculated the correlation function $S_{1c1b} = \langle \mathbf{S}_{1c} \cdot \mathbf{S}_{1b} \rangle$, between the spins in orbitals $1c$ and $1b$, on the cluster and in the bath. We have $-3/4 \leq S_{1c1b}$. This is shown in Fig. 1 as a function of U (full curve). For moderate values of U there is a strong negative correlation, which increases with U . The correlation function $C_{1c\uparrow 1c\downarrow} = \langle n_{1c\uparrow} n_{1c\downarrow} \rangle - 1/4$ is also shown. It becomes negative as U is increased, implying a reduced double occupancy of orbital $1c$ and a spin $1/2$ state starts to form on $1c$. Both these results are consistent with levels $1c$ and $1b$ forming a Kondo-like state. Further increase of U , however, leads to a rapid reduction of $|S_{1c1b}|$ and $C_{1c\uparrow 1c\downarrow}$ grows positive. The Kondo singlets are then broken, and instead a bound state is formed on the cluster. Increasing U reduces the Kondo energy, while the gain from correlating the electrons on the cluster increases. In the lowest cluster state $C_{1c\uparrow 1c\downarrow}$ takes the value 0.25 , which is approached in Fig. 1.

Fig. 1 also shows weights of the spectral function on the photoemission side for the two-orbital, two-site model. For $U = 0$ all weight is in the leading peak. As U is increased, the weight of the Hubbard peak increases, corresponding to final state with primarily one electron less on the cluster site. At the same time three peaks close to the leading peak get more weight, summed up in the curve “Satellites”. These peaks correspond to final states with substantial weights in excited neutral cluster states, while the leading peak corresponds to a final state with a large weight of the lowest neutral state. For $U/|V| \sim 11$

FIG. 1. Weight of the correlation functions S_{1c1b} and $n_{1c\uparrow}n_{1c\downarrow}$ as well as spectral weights for the leading peak, satellites forming the peaks surrounding the the pseudogap and the Hubbard peak for the two-level two-orbital model.

the satellites acquire more weight than the leading peak and a pseudogap starts to develop. This happens when the Kondo-like effect starts to be lost and a bound state on the cluster starts to form.

To address the pseudogap, we study a general cluster with $U \gg |V|$. Assume an integer number of electrons per cluster, n_0 , and project out the piece, $|\Phi_{n_0}\rangle$, of the ground-state, $|\Phi\rangle$, that has exactly n_0 electrons. We study photoemission processes, removing an electron with the quantum number ν and energy ε . We split $|\Phi_{n_0}\rangle$ as

$$|\Phi_{n_0}\rangle = |0\rangle + |1\rangle, \quad (3)$$

where $c_\nu|0\rangle = 0$ and $c_\nu^\dagger c_\nu|1\rangle = |1\rangle$. Let $H_0 = \sum_{\nu\varepsilon} (V_\nu c_\nu^\dagger c_{\nu\varepsilon} + \text{h.c.})$ give the hopping into the cluster. We then write the part of the ground-state corresponding to $n_0 + 1$ electrons as

$$|\Phi_{n_0+1}\rangle = - \sum_{\nu'} \sum_{\varepsilon} \frac{V_{\nu'\varepsilon}}{\Delta E_+ - \varepsilon} c_{\nu'}^\dagger c_{\nu'\varepsilon} |\Phi_{n_0}\rangle, \quad (4)$$

where we have approximated the energy difference between cluster states with one extra electron (hole) and the lowest neutral state as ΔE_+ (ΔE_-). If the lowest state of the isolated cluster is nondegenerate, this is (non-degenerate) perturbation theory. However, if the lowest state is degenerate, e.g., for the Kondo problem, $|0\rangle$ and $|1\rangle$ contain much more information than can be obtained from perturbation theory, and the treatment goes beyond perturbation theory. For an infinite system, we construct a state $|\nu\varepsilon_F\rangle$ with a hole in the bath at the Fermi energy ε_F . We form $c_{\nu\varepsilon_F}|\Phi_{n_0}\rangle$ and allow hopping to states with $n_0 - 1$ electrons on the cluster. The corresponding am-

FIG. 2. Initial and final states for a process removing an electron with quantum number ν . There is a negative interference between two channels for reaching the final state.

plitude is then

$$\begin{aligned} \langle \nu \varepsilon_F | c_\nu | \Phi \rangle = & \sum_{\varepsilon} V_{\nu\varepsilon} \left(\frac{\langle 1 | c_{\nu\varepsilon_F}^\dagger c_{\nu\varepsilon} | 1 \rangle}{\Delta E_- + \varepsilon} - \frac{\langle 0 | c_{\nu\varepsilon_F}^\dagger c_{\nu\varepsilon} | 0 \rangle}{\Delta E_+ - \varepsilon} \right) \\ & + \sum_{\nu'(\neq\nu)\varepsilon} V_{\nu'\varepsilon} \left(\frac{\langle 0 | c_{\nu'}^\dagger c_{\nu} c_{\nu\varepsilon_F}^\dagger c_{\nu'\varepsilon} | 1 \rangle}{\Delta E_- + \varepsilon} + \frac{\langle 0 | c_{\nu'}^\dagger c_{\nu} c_{\nu\varepsilon_F}^\dagger c_{\nu'\varepsilon} | 1 \rangle}{\Delta E_+ - \varepsilon} \right). \end{aligned}$$

If the isolated cluster ground-state is nondegenerate and $\Delta E_{\pm}$ are large, the number of holes in the bath for the states $|0\rangle$ and $|1\rangle$ is small. Then the third and fourth terms are also small, while the expectation values in the first and second terms are large and tend to cancel. If the orbital ν is half-filled the cancellation becomes perfect. This cancellation is illustrated schematically in Fig. 2. For, e.g., $U = 1.0$ and $V = -0.02$ in the model above, the four terms are -0.0117 , 0.0121 , 0.0020 , 0.0020 . This illustrates the strong cancellation between the first two terms and the small magnitude of the next two terms.

If the lowest cluster state is degenerate, the situation is quite different. The system may then form a Kondo-like state even for large U . The first term in the amplitude above is then smaller and does not cancel the second term. Furthermore, the “spin-flip” terms three and four can be substantial. As a result, a pseudogap may not develop. In the model above, a negative ΔU gives a triplet ground-state for the model cluster. In the limit of a large U , there are then triplet states on both the c and b site, coupling to a total singlet. The leading peak has more weight than each of the neighboring satellite for the U range shown in Fig. 1, and a pseudogap does not form. For an infinite system, these results can also be discussed in terms of the phase shift at the Fermi energy[14].

We now study $N_c = 8$ at half-filling. Fig. 3 shows correlation functions of the type $C_{(\pi,0)\uparrow,(\pi,0)\downarrow} = \langle n_{(\pi,0)\uparrow} n_{(\pi,0)\downarrow} \rangle - \langle n_{(\pi,0)\uparrow} \rangle \langle n_{(\pi,0)\downarrow} \rangle$. This function behaves in a very similar way as $C_{1c\uparrow 1c\downarrow}$ in Fig. 1. As U is increased it first turns negative, indicating the formation of a Kondo-like state. Then it turns positive, indicating the formation of a bound state on the cluster in the $(\pi, 0)$, $(0, \pi)$ -space similar as for the two-site two-orbital model. This is also supported by the strong negative correlation between $(\pi, 0)$ and $(0, \pi)$ as is also found for $|1-\rangle$ in Eq. (2). At the same time a pseudogap develops

FIG. 3. Correlation functions as a function of U for an embedded eight site cluster in the first iteration. The parameters are $t = -0.4$ eV and $T = 290$ K.

TABLE I. Bath-cluster spin correlations and hybridization energies from ED calculations. The parameters are $t = -0.4$ eV, $V(\pi, 0) = 0.08$ eV and $V(\pi/2, \pi/2) = 0.21$ eV.

			$\langle S^z s^z \rangle$		$\langle V \rangle$	
U	μ	n	$(\pi, 0)$	$(\frac{\pi}{2}, \frac{\pi}{2})$	$(\pi, 0)$	$(\frac{\pi}{2}, \frac{\pi}{2})$
2.5	1.25	1.0	-0.0020	-0.0051	-0.021	-0.097
3.2	1.6	1.0	-0.0012	-0.0030	-0.014	-0.066

in the $(\pi, 0)$ spectrum, as in Fig. 1. $C_{(\pi/2,\pi/2)\uparrow,(\pi/2,\pi/2)\downarrow}$ behaves in a similar way, but it stays Kondo-like up to larger values of U before the $(\pm\pi/2, \pm\pi/2)$ -space is also used to form a bound state on the cluster. Therefore the $(\pm\pi/2, \pm\pi/2)$ pseudogap forms for larger U . The reason is that the coupling to the bath is a factor of three to four stronger for $(\pi/2, \pi/2)$ and it is favorable to keep the Kondo-like state up to a larger U .

To further support this, we have performed exact diagonalization (ED) calculations for $N_c = 8$ and one bath level at the chemical potential with the coupling $V(\mathbf{K})$ to the cluster $\mathbf{K}$ -state. We study the correlation $\langle S^z s^z \rangle$ between spins on the cluster and the bath and the hopping energy $\langle V \rangle$ between the cluster and the bath for a certain $\mathbf{K}$. Table I shows that both quantities are much larger for $(\pi/2, \pi/2)$ and that both decrease rapidly with U .

The energy gain when all electrons of the isolated cluster correlate is much larger than the sum of the energies gained when correlation is only allowed in the $(\pi, 0)$, $(0, \pi)$ - or $(\pm\pi/2, \pm\pi/2)$ -spaces. This favors a simultaneous switch of both spaces from the Kondo-like states to a correlated cluster state. However, the difference in the coupling to the bath is so large that the switch, nevertheless, happens for different U in Fig. 3. This can be different for the frustrated case.

We now consider the frustrated case, $t' = -0.3t$, for

TABLE II. Bath-cluster spin correlations and hybridization energies from ED calculations on $N_c = 8$ frustrated clusters with $U = 2.5$ eV, $t = -0.4$, $t' = -0.3t$, $V(\pi, 0) = 0.08$ eV and $V(\pi/2, \pi/2) = 0.21$ eV. We also show $J(\mathbf{K})$ [Eq. (5)].

μ	n	$\langle S^z s^z \rangle$		$\langle V \rangle$		J/V^2	
		$(\pi, 0)$	$(\frac{\pi}{2}, \frac{\pi}{2})$	$(\pi, 0)$	$(\frac{\pi}{2}, \frac{\pi}{2})$	$(\pi, 0)$	$(\frac{\pi}{2}, \frac{\pi}{2})$
0.85	0.96	-0.0042	-0.0068	-0.036	-0.12	1.61	1.78
1.05	0.98	-0.0038	-0.0063	-0.035	-0.11	1.69	1.64
1.25	1.0	-0.0048	-0.0065	-0.043	-0.11	1.88	1.60
1.35	1.02	-0.0064	-0.0063	-0.047	-0.12	2.04	1.61
1.45	1.04	-0.0084	-0.0071	-0.055	-0.13	2.27	1.64

different fillings and $U = 8|t|$ in the first iteration. For moderate hole doping the $N_c = 8$ calculation shows a clear pseudogap. On the other hand, for electron doping we find no pseudogap. We first study this using ED as above. The coupling to the bath is kept fixed, and n is varied by varying μ . Table II shows how both the cluster-bath spin correlation and hopping energy increase for $(\pi, 0)$ as we go from hole- to electron-doping, while the change is small for $(\pi/2, \pi/2)$. To understand this, we study the $J(\mathbf{K})$ obtained from a Schrieffer-Wolf transformation

$$J(\mathbf{K}) = |V(\mathbf{K})|^2 \left(\frac{1}{\epsilon(\mathbf{K}) + U - \mu} + \frac{1}{\mu - \epsilon(\mathbf{K})} \right). \quad (5)$$

Due to the shift of μ with n , $J(\mathbf{K})$ increases for $(\pi, 0)$, while the levels for $(\pi/2, \pi/2)$ are located in such a way that the changes of $J(\mathbf{K})$ are relatively small.

In the ED calculation we assumed a doping independent coupling. The coupling is related to $\text{Im } 1/G_0(\mathbf{K}, \omega_n)$, where G_0 is the Green's function of the bath. For $t' = -0.3t$ and moderate hole doping, both $\text{Re } G_0[(\pi, 0), \omega_n]$ and $\text{Im } G_0[(\pi, 0), \omega_n]$ are large for small imaginary frequencies ω_n . Then $\text{Im } 1/G_0[(\pi, 0), \omega_n]$ is small and the coupling relatively weak. As n is increased, μ shifts so that for the electron doped system both the real and imaginary parts of G_0 are smaller and the coupling larger. This increase and the increase of $J(\mathbf{K})$ with n favor a Kondo effect for $(\pi, 0)$ for the electron-doped system. The Kondo effect then tends to be lost simultaneously in the $(\pi, 0)$ - and $(\pm\pi/2, \pm\pi/2)$ -spaces. This result depends crucially on the sign of t'/t . For the organic superconductors, with a triangular lattice and at half-filling, we find that the pseudogap goes away at $t' \sim 0.6t$, in agreement with experiment[2], since $J(\pi, 0)$ has a minimum at $t' = 0$ for half-filling. An additional effect is that when the system is doped, the ground-state has a substantial mixture of a cluster state with one electron more or less. This state is degenerate, which is unfavorable for a pseudogap and in agreement with the lack of a pseudogap for large doping.

To understand the character of the pseudogap, we have calculated $P_d = \sum_{ij} \langle \Delta_i \Delta_j^\dagger \rangle / N_c$, where Δ_i is the operator

TABLE III. Energy E , spin S , degeneracy Deg. and d -wave superconductivity correlation P_d for low-lying states with $S_z = 0$ for $N_c = 8$. Deg. refers to the degeneracy of $S_z = 0$ states. The parameters are $U = 3$, $t = -0.324$ eV and $t' = 0$.

E	-1.116	-1.062	-0.941	-0.897	-0.764
S	0	1	2	1	0
Deg.	1	1	1	6	9
P_d	0.37	0.28	0.10	0.15	0.10

for d -wave superconductivity. A factorized term was subtracted so that $P_d = 0$ for $U = 0$. For the ground-state of the isolated cluster P_d is rather large (see Table III). This state dominates the ground-state of the embedded cluster for relatively large U . The peaks surrounding the pseudogap correspond primarily to final states with a large weight of excited neutral states with $S = 0$ or 1, for which P_d is substantially smaller. We can then think of the pseudogap as corresponding to the break up of a d -wave singlet pair.

This relation of the pseudogap to d -wave pairs does not contradict the observation that the pseudogap and superconductivity compete[16]. The occurrence of superconductivity is determined by the Bethe-Salpeter equation, and depends on the matrix $(1 - \bar{\chi}^0 \Gamma_c)$ becoming singular[4]. Here Γ_c is the two-particle irreducible vertex, representing the interaction, and $\bar{\chi}^0$ consists of products of two dressed Green's functions, describing propagation of pairs. $\bar{\chi}^0$ is closely related to the density of states at the Fermi energy. As U is increased, Γ_c grows and $\bar{\chi}^0$ is reduced. This reduction becomes very rapid as a pseudogap forms[17]. This hurts the onset of superconductivity. A similar competition between $\bar{\chi}^0$ and Γ_c was observed for alkali-doped fullerenes[18].

From the discussion below Eq. (4) we may expect to see a pseudogap if the lowest state of the cluster is non-degenerate, independently of P_d . To test this, we have applied an antiferromagnetic (AF) field $\sum_i \delta_i (n_{i\uparrow} - n_{i\downarrow})$, where $\delta_i = \delta$ on one sublattice and $-\delta$ on the other. This term is only applied to the cluster and not to the bath. One iteration is performed. This term favors an AF state on the cluster and reduces P_d . Fig. 4 shows that there is, nevertheless, a pseudogap for $\mathbf{K} = (\pi, 0)$.

In conclusion we have provided and analyzed a very simple model, showing pseudogap formation. As U is increased, the system switches from a case where the cluster orbitals form a Kondo state with the bath to a case where a nondegenerate bound state is formed on the cluster. This leads to a pseudogap. We showed that this analysis also applies to larger clusters. It is crucial that the coupling to the bath is much weaker for $\mathbf{K} = (\pi, 0)$ than for $(\pi/2, \pi/2)$, which leads to the strong $\mathbf{K}$ -dependence. We have found that the pseudogap is lost for the electron doped system, due to an increase in the effective J and the cluster-bath coupling for $\mathbf{K} = (\pi, 0)$. The pseudogap

FIG. 4. Spectrum averaged over $\mathbf{k}$ around $\mathbf{K} = (\pi, 0)$ and $(\pi/2, \pi/2)$ when an AF field δ is applied to the cluster.

can be interpreted in terms of d -wave superconducting pairs. It strongly reduces $\bar{\chi}^0$, describing how the propagation of pairs is hindered, leading to a competition with superconductivity.

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