

Active Motion Induced by Random Electromagnetic Fields

Jorge Luis-Hita,* Juan José Sáenz, and Manuel I. Marqués*

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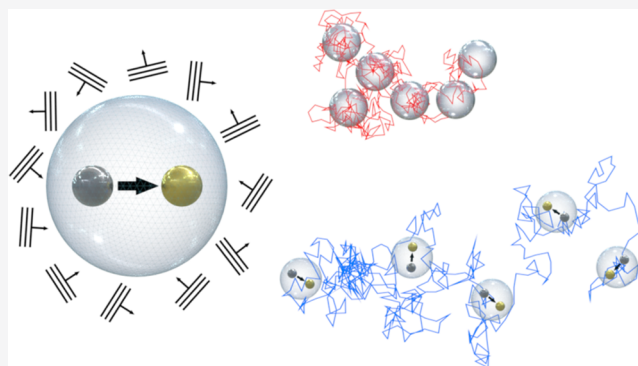
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ABSTRACT: Active particles are capable of extracting energy from their environment and converting it into direct motion. This direct motion is usually attained by asymmetries, which induce a net force in the particle. In this article we show how a dimer made up of two nanoparticles can obtain this energy supply from the nonreciprocal force induced by an homogeneous and isotropic random electromagnetic field. We explicitly obtain the value of the force and find it to be different from zero only for sets of different particles in which at least one of them is absorbent. In addition, we prove that the nonreciprocity of the force is the condition for the interaction dynamics to be nonconservative. We further show how this mechanism can be used to induce active Brownian motion with persistent random walks, effective diffusion coefficients, and noticeable Péclet numbers.

KEYWORDS: Optical forces, Active motion, Random fields, Nonreciprocal forces, Dipoles



Active Brownian particles have the ability to self-propel by converting the energy available in their surroundings into directed motion. A general feature of direct motion is that it is triggered by asymmetries that induce a nonreciprocal force.^{1–3} These nonreciprocal forces in colloidal systems have been intensely investigated in the context of nonequilibrium fluctuations or many-body Langevin dynamics^{4–6} and are found to be strongly related to systems of particles immersed in nonequilibrium environments.⁷ In a self-propelled particle, the nonreciprocal force induces a constant velocity along the particle's asymmetry axis that is coupled with its rotational diffusion. This coupling constantly randomizes the propulsion direction with a characteristic time scale inversely proportional to the rotational diffusive constant. The particle feeling the nonreciprocal force then has the propensity for performing straight paths with a persistence length proportional to the induced speed. This persistence length increases the diffusion constant of the Brownian particle. The aforementioned property has been verified experimentally,^{8,9} and various propulsion mechanisms to induce a net nonreciprocal force have been proposed, such as chemical reactions,^{10–12} external illumination,^{13–19} ultrasound,²⁰ and magnetic^{21–24} and electric fields.^{25,26} Speckle patterns have been used to promote out of equilibrium dynamics,^{27,28} and the use of optical forces, applied currently to promote binding,^{29,30} have been proposed in the literature to induce active motion.³¹ However, an analysis of an active propulsion driven by nonreciprocal optical forces in a isotropic radiation environment is still lacking.

In the field of optical forces, the nonreciprocity of the optical interaction in a dimer has been reported in a variety of

nonisotropic theoretical situations, such as two particles illuminated by a plane wave³² or a dimer moving through an optical lattice.³³ It has also been studied experimentally, with measurement of the interaction force between two particles illuminated by plane waves and confined to a pseudo-one-dimensional configuration.³⁴

However, optical forces between two particles in a dimer may be also induced in an isotropic radiation bath consisting of fluctuating electromagnetic waves with random orientations and polarizations. These optical interactions between nanoparticles in isotropic fluctuating fields have been investigated over the last few years.^{35,36} When they were calculated with the consideration of absorbing particles, they led to unexpected features, such as near- and far-range inverse-squared interactions³⁷ and the fulfillment of Kepler's laws.³⁸ In order to use this system to induce active motion, we must determine the asymmetry conditions needed to obtain a nonreciprocal force among the particles forming the dimer.

To ensure the conservativity of the particles' dynamics, specific conditions will be needed. From classic textbooks on analytical mechanics^{39,40} we know that the condition for a three-dimensional force acting on a pointlike particle to be conservative is the existence of a vanishing curl. In the special

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case of a central force the conservativity is assured due to the absence of tangential components of the force. Interestingly, the condition of vanishing curls will not be sufficient to ensure conservativity when the system is formed by more than one particle.

In this paper we will explicitly calculate the force felt by a dimer made up of two different electric dipoles and we will show how the condition to get a nonreciprocal force is to have at least one absorbing particle. After that, the conditions for the system to be conservative will be obtained and checked by numerical calculations. We will find that conservativity is determined by the reciprocity of the forces. Then, we will analyze the active Brownian dynamics induced by this isotropic force, coming solely from the optical interactions, and we will check our predictions for the effective diffusion constant using molecular dynamics simulations.

■ NONRECIPROCAL ISOTROPIC OPTICAL FORCES

The light-induced interaction force between two identical, absorbing nanoparticles separated along the z axis in a homogeneous, isotropic, random electromagnetic field is given in refs 35 and 37. Note that this field is random in space and in time, with fixed frequency and amplitude. Now we will allow particles A and B to have different polarizabilities.

The electrical fields on each particle A, B can be written as

$$\psi_A = \psi_A^0 + \mathbf{G}_{AB} \mathbf{T}_B \psi_B \quad (1)$$

$$\psi_B = \psi_B^0 + \mathbf{G}_{BA} \mathbf{T}_A \psi_A \quad (2)$$

where ψ^0 is the external illumination, $\mathbf{G}_{AB} = \mathbf{G}(r_A, r_B)$ is the Green dyadic function between positions r_A and r_B and $\mathbf{T}_{A,B}$ are matrices defined by

$$\mathbf{T}_{A,B} = k^2 \alpha_{A,B} \mathbf{I} \quad (3)$$

where \mathbf{I} is the identity matrix, k is the wavenumber, and $\alpha_{A,B}$ are the polarizabilities of particles A and B.

By defining the matrices

$$\mathbf{A} = \mathbf{T}_A^{-1} - \mathbf{G}_{AB} \mathbf{T}_B \mathbf{G}_{BA} \quad (4)$$

$$\mathbf{B} = \mathbf{T}_B^{-1} - \mathbf{G}_{BA} \mathbf{T}_A \mathbf{G}_{AB} \quad (5)$$

the solutions to the fields on each particle can be expressed as

$$\mathbf{T}_A \psi_A = \mathbf{A}^{-1} (\psi_A^0 + \mathbf{G}_{AB} \mathbf{T}_B \psi_B^0) \quad (6)$$

$$\mathbf{T}_B \psi_B = \mathbf{B}^{-1} (\psi_B^0 + \mathbf{G}_{BA} \mathbf{T}_A \psi_A^0) \quad (7)$$

The derivatives of the fields are obtained straightforwardly. In addition, the field–field correlations are given by⁴¹

$$\langle \psi_B^{0*} \otimes \psi_A^0 \rangle = \frac{4\pi}{\epsilon\epsilon_0 k} u_E(\omega) \text{Im}\{\mathbf{G}_{BA}\} \quad (8)$$

$$\langle [\partial_{z_B} \psi_B^{0*}] \otimes \psi_A^0 \rangle = \frac{4\pi}{\epsilon\epsilon_0 k} u_E(\omega) \text{Im}\{\partial_{z_B} \mathbf{G}_{BA}\} \quad (9)$$

where u_E is the energy density of the applied field.

Using these expressions, we can compute the time-averaged z component of the optical force on particle A:

$$\langle F_A^z \rangle = \frac{\epsilon\epsilon_0}{2} \langle \text{Re}\{\psi_A^t \alpha_A \cdot \partial_{z_A} \psi_A^*\} \rangle \quad (10)$$

To do so, we take into account that, in the case of absorbing particles, the following relation holds:

$$-\text{Im}\{\mathbf{G}_{AA}\} = \text{Im}\{\mathbf{T}_A^{-1}\} - \text{Im}\{\mathbf{K}_A^{-1}(\omega)\} \quad (11)$$

where

$$\mathbf{K}_A(\omega) = k^2 \alpha_{0A} \mathbf{I} \quad (12)$$

α_0 being the static polarizability. The derivation of these equations can be found in the [Supporting Information](#). Furthermore

$$\text{Im}\{\mathbf{K}^{-1}\} = -\frac{\sigma_{\text{abs}}}{k^3 |\alpha|^2} \mathbf{I} = -\frac{\sigma_{\text{abs}}/\sigma_{\text{scat}}}{3\lambda} \mathbf{I} \quad (13)$$

i.e., the imaginary part of the inverse of \mathbf{K} , is proportional to the ratio of the absorption cross section to the scattering cross section of the nanoparticles.

Using these relations, we find that the general force acting on particle A can be expressed as

$$\begin{aligned} F_z^A = & \frac{2\pi u_E(\omega)}{k^3} \text{Im}\{\text{Tr}[[\partial_{z_A} \mathbf{G}_{AB}] \mathbf{T}_B \mathbf{G}_{BA} \mathbf{A}^{-1}]]\} + \frac{2\pi u_E(\omega)}{k^3} \\ & \text{Re}\{\text{Tr}[[\partial_{z_A} \mathbf{G}_{AB}^*] \mathbf{B}^{-1*} \text{Im}\{\mathbf{K}_B^{-1}(\omega)\} \mathbf{T}_B \mathbf{G}_{BA} \mathbf{A}^{-1}]]\} \\ & + \frac{2\pi u_E(\omega)}{k^3} \text{Re}\{\text{Tr}[[\partial_{z_A} \mathbf{G}_{AB}^*] \mathbf{B}^{-1*} \mathbf{G}_{BA} \mathbf{T}_A^* \\ & \text{Im}\{\mathbf{K}_A^{-1}(\omega)\} \mathbf{A}^{-1}]]\} \end{aligned} \quad (14)$$

The force in B is obtained by changing A to B and vice versa. Adding both forces, we obtain the final result for the total force on the center of mass of the system ($F_z^{\text{cm}} = F_z^A + F_z^B$). Note how, for nonabsorbing particles, only the first term in eq 14 remains. However, since $\mathbf{A} \mathbf{T}_A = \mathbf{B} \mathbf{T}_B$ implies $\mathbf{T}_B \mathbf{G}_{BA} \mathbf{A}^{-1} = \mathbf{T}_A \mathbf{G}_{AB} \mathbf{B}^{-1}$ and $\partial_{z_A} \mathbf{G}_{AB} = -\partial_{z_B} \mathbf{G}_{BA}$, we obtain $F_z^A = -F_z^B$, resulting in $F_z^{\text{cm}} = 0$ for the case of particles with null absorption. However, for the general case, when both forces are added, we obtain

$$\begin{aligned} F_z^{\text{cm}} = & \frac{4\pi u_E(\omega)}{k^3} \text{Tr}\{\text{Im}(\partial_{z_A} \mathbf{G}_{AB}) [\text{Im}(\mathbf{K}_A^{-1}) \\ & \text{Im}(\mathbf{B}^{-1*} \mathbf{A}^{-1} \mathbf{T}_A^* \mathbf{G}_{AB}^*) \\ & + \text{Im}(\mathbf{K}_B^{-1}) \text{Im}(\mathbf{B}^{-1*} \mathbf{A}^{-1} \mathbf{T}_B \mathbf{G}_{AB})]\} \end{aligned} \quad (15)$$

Several noticeable consequences are derived from this equation. First, the interaction force between two nonabsorbing particles in an homogeneous, isotropic, random electromagnetic field is always reciprocal, even in the case of different particles. Second, in order to obtain a force in the center of mass different from zero, at least one of the particles must be absorbent. It is worth noticing that, as mentioned in refs 34 and 35, the total linear momentum of the system is still conserved, since part of the linear momentum is carried away by the scattered light. What is meant here by nonreciprocal interaction concerns the particle–particle system, which sees a net force acting on the center of mass. As we will show in the next section, this nonreciprocity leads to a nonconservative behavior for the force inducing the active motion. For that reason, absorption is not a sufficient condition to ensure nonconservation of the energy; asymmetry is also needed.

Next, we analyze the nonreciprocity of the interaction force between the two nanoparticles for a particular case. Let us consider that the polarizabilities of particles A and B correspond to those of gold and silver spherical nanoparticles, of $a = 30$ nm radius, in water. The gold and silver dielectric

functions can be obtained from the literature.⁴² The homogeneous, isotropic, random electromagnetic field is characterized by its wavelength, $\lambda = 514$ nm, corresponding to an argon laser. We consider a laser intensity of $I = 1 \times 10^{10}$ W/m².

The force on the center of mass of the gold–silver system is calculated with eq 15 (see Figure 1). The separation between

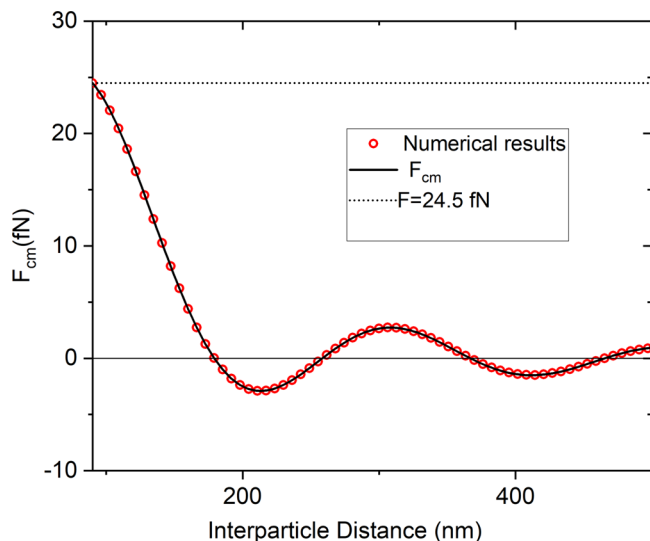


Figure 1. Force in the center of mass of the silver–gold dimer in water versus separation of the particles induced by a random electromagnetic field with an intensity of 10^{10} W/m². The force is obtained analytically (continuous line) using eq 15 and numerically (red circles) using a plane wave decomposition of the field. The dotted line indicates the value of the force when the interparticle separation distance is set to 3 times the radius (90 nm). The separation between particles is computed as the center to center distance. The minimum distance between particles is chosen so that the dipole approximation is valid in the entire separation range ($d_{\min} = 3a = 90$ nm).

particles is computed as the center to center distance. The minimum distance between particles is chosen so that the dipole approximation is valid in the entire separation range ($d_{\min} = 3a = 90$ nm). Interestingly, the force on the center of mass changes its sign as a function of the interparticle distance. This phenomenon is commented upon in the [Supporting Information](#). As a consequence, the dimer can be actively propelled by the homogeneous, isotropic, random electromagnetic field and the magnitude and direction of the propulsion are going to be determined by the separation between the particles and the frequency of the radiation.

To gain some knowledge on how this force on the center of mass behaves, a contour map is shown in Figure 2. In this map, F_0 is defined as $F_0 = 24.5$ fN. The $\lambda = 514$ nm line is explicitly shown. From this figure, it can be seen that the magnitude of the interaction force decreases with increasing wavelength. This is consistent with the decreasing value of the absorption terms for gold and silver nanoparticles at these wavelengths. Also, it can be seen that the modulation intervals increase with the wavelength.

To check the validity of eq 15, we have calculated the force on the center of mass numerically, explicitly considering the effect of 10^6 fluctuating plane waves (see the [Supporting Information](#)). The result of this calculation is shown in Figure 1. Note how the analytical result given by eq 15 coincides with

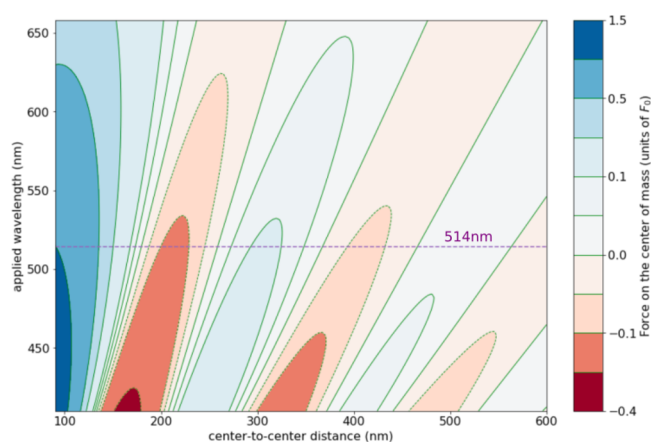


Figure 2. Contour plot of the force in the center of mass of the silver–gold dimer in water as a function of the center to center separation and the applied wavelength. The scale of the force is given by $F_0 = 24.5$ fN. The minimum interparticle distance is chosen to be $d_{\min} = 3a = 90$ nm to fulfill the dipole approximation. The line of $\lambda = 514$ nm is explicitly shown. It can be seen that the magnitude of the force decreases with increasing wavelengths.

the numerical simulation, demonstrating the existence of a nonreciprocal interaction.

We can fix the separation distance between both nanoparticles to be equal to 3 times the radius and obtain the value of the force (F_z^{cm}) for a field intensity (I):

$$F_z^{\text{cm}} = 2.45 \times 10^{-24} \left(\frac{Nm^2}{W} \right) I \quad (16)$$

NONCONSERVATIVITY

The conservativity of the interaction dynamics in the gold and silver nanoparticles system under random light illumination will now be determined. From the classic textbooks on analytical mechanics it is known that the force acting on a pointlike particle in a three-dimensional system is conservative if the line integral of the force along a closed path vanishes or, in an equivalent statement, if its curl is identically zero.^{39,40}

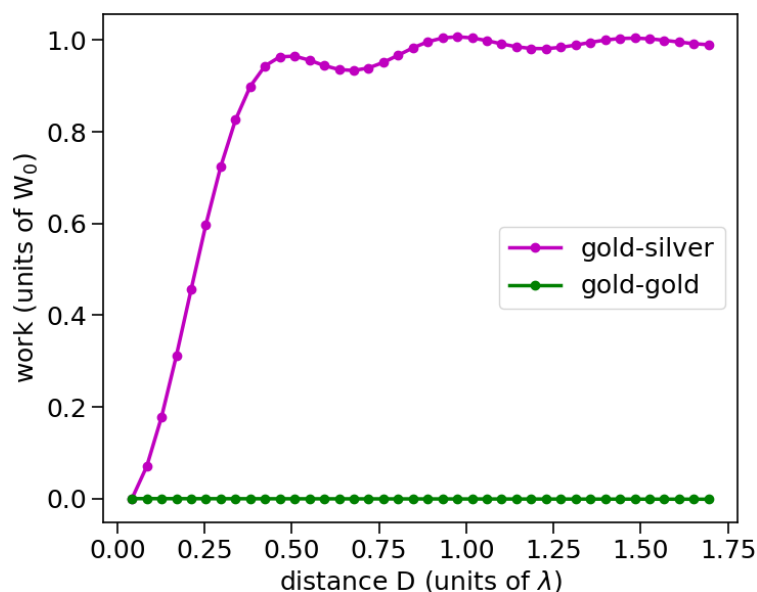
In the two-particle system, however, these two statements are not equivalent. We will show that it is possible to have a system in which $\nabla_A \times \mathbf{F}^A = \nabla_B \times \mathbf{F}^B = 0$ and still the line integral over a closed path does not vanish, indicating that the interaction dynamics is nonconservative.

From a mathematical point of view, the conditions on the interaction forces to produce a conservative interaction dynamics can be obtained from the approach of differential forms.⁴³ From this approach, it is known that a field $\mathbf{J} = (A_1, A_2, \dots, A_N)$ is conservative if all the conditions

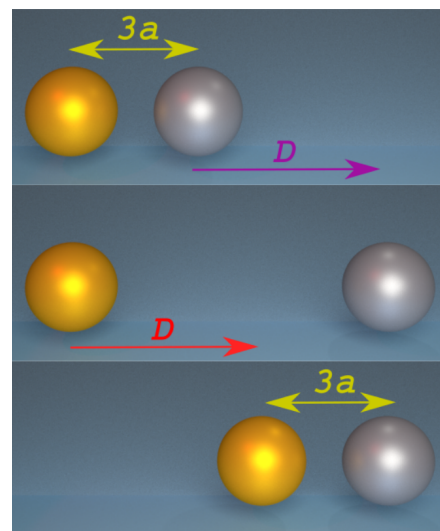
$$\frac{\partial A_i}{\partial \phi_j} = \frac{\partial A_j}{\partial \phi_i} \quad i, j = 1, \dots, N, \quad i \neq j \quad (17)$$

where $A_i = A_i(\phi_1, \phi_2, \dots, \phi_N)$, are satisfied. For our purposes, A_j represents the components of the forces on the system and ϕ_j represents the components of the positions of the particles. This rationale can be applied to systems of arbitrary number of particles.

In our system, for the six-dimensional field \mathbf{J} that combines the forces on both particles



(a) Work done by the system as a function of the distance D .



(b) Diagram of the displacement between two equivalent configurations.

Figure 3. Work done by the particle–particle system as a function of the distance D . (a) Results for the 5 nm radius gold–gold and gold–silver cases. It can be seen that the work is zero in the case of identical particles. (b) Diagram of the displacement between two equivalent configurations used to perform the line integral of the optical interaction force between the gold and the silver nanoparticles. Since the field is homogeneous and the tangential forces are zero, the path can be considered a closed path.

$$\mathbf{J} = (F_x^A, F_y^A, F_z^A, F_x^B, F_y^B, F_z^B) \quad (18)$$

the set of conditions (eq 17) reduces to

$$\frac{\partial F_z^A}{\partial z_B} = \frac{\partial F_z^B}{\partial z_A} \quad (19)$$

since there are no tangential forces and $F_z^{A,B}$ only depends on $z_{A,B}$. It is worth noting that, by taking the derivatives with respect to the separation distance $d = z_A - z_B$, this condition can be written as

$$F_z^A = -F_z^B + \text{const} \quad (20)$$

Note that, since interactions are expected to decay at long distances, the only physically admissible value of the constant is zero. Therefore, for a system of two nanoparticles in a fluctuating, random electromagnetic field, the reciprocity of the interaction forces is the condition for the interaction dynamics to be conservative.

In a more physical picture, it can be seen that in the one-dimensional problem we are considering (particles separated in the z direction) the condition $\nabla_A \times \mathbf{F}^A = \nabla_B \times \mathbf{F}^B = 0$ indicates that both forces can be expressed in terms of a potential energy: $F_z^A = -\partial U_A(r_A)/\partial r_A$ and $F_z^B = -\partial U_B(r_B)/\partial r_B$ with $r_A = z_A - z_B$ and $r_B = z_B - z_A$. The time variation of the kinetic and potential energy of both particles is given by

$$\dot{T}_i = -\partial U_i(z_i)\dot{z}_i \quad (21)$$

$$\dot{U}_i = \partial U_i/\partial r_i(\dot{z}_i - \dot{z}_j) \quad (22)$$

with $i, j = A, B$.

From the above expressions, if $\dot{z}_j = 0$, then $\dot{T}_i + \dot{U}_i = 0$, which is the usual energy conservation law expected in gradient-derived forces. However, if both particles move, the

energy is not necessarily conserved. If both particles are then free to move, the condition to have a conservative system is to have a reciprocal force. Actually if $F_z^A = -F_z^B$, by calculating the time derivative of $U_A + T_A + U_B + T_B$ it is easy to show that $\dot{U}_i = \dot{U}_j = 0$ and $\dot{T}_i + \dot{T}_j + \dot{U}_i = 0$, which is the condition for the conservation of the total mechanical energy on the system. However, if the forces are not reciprocal, $F_z^A + F_z^B \neq 0$, then the mechanical energy is no longer conserved. Therefore, for a system of two nanoparticles in a random field, the reciprocity of the interaction force is, again, the condition for the interaction dynamics to be conservative.

Applying this statement, in a system of two nonabsorbing nanoparticles the dynamics will always be conservative, even in the case of different polarizabilities since, as it was shown in the previous section, the interaction forces are always reciprocal. In the same manner, the interaction dynamics of two identical absorbing nanoparticles will always be conservative under fluctuating, random light illumination, even though absorption means that electromagnetic energy is constantly transformed into thermal energy at some pace.

In the system formed by the gold and silver nanoparticles, however, the reciprocity of the interaction forces is not fulfilled, resulting in a nonconservative interaction dynamics.

This result can be checked by performing the line integral of the interaction force along a path that connects two equivalent configurations of the system, as is shown in Figure 3b. We start with the gold and silver nanoparticles separated by a distance $3a$. Then, the silver nanoparticle is displaced along the z axis a certain distance D . Finally, the gold nanoparticle is displaced the same distance D along the z axis. Since the random electromagnetic field is homogeneous and isotropic, the initial and final configurations are equivalent and the line along which the integral is performed describes a closed path.

The results of the computed line integrals as a function of the distance D are shown in Figure 3a. In the case of a gold–silver nanoparticle system, it can be seen from the figure that there is always work performed by (on) the system along the described path. This indicates a nonconservative interaction dynamics. However, when a system of identical absorbing particles is considered, the performed work always vanishes. To show this result, the same line integral has been performed in the case of the gold–gold nanoparticle system. It can be seen that this work vanishes for every value of the distance D .

It should be stressed that, since there are no tangential forces in the system and the $F_z^{A,B}$ components only depend on $z_{A,B}$, the curls of the forces always vanish, $\nabla_A \times \mathbf{F}^A = \nabla_B \times \mathbf{F}^B = 0$, even in the case of nonidentical absorbing particles, where the interaction dynamics has been shown to be nonconservative.

ACTIVE MOTION

Now, we will analyze the active motion induced by the fluctuating isotropic field in the particular case of a 30 nm radius gold–silver dimer with a separation distance fixed to 90 nm. Both nanoparticles are embedded in a water-refractive index matching polymer with spherical shape: $R = 5 \mu\text{m}$ radius and a density of 1500 kg/m^3 .

This particle is nonisotropic and has a preferential axle given by the dimer's axis. The nonreciprocal force induced by the random fluctuating field always points in the direction of this preference axis and, in conjunction with the stochastic motion, promotes an active Brownian movement and a persistent random walk with an effective diffusion constant given by^{44–46}

$$D_{\text{eff}} = D + \frac{v_0^2}{6D_r} \quad (23)$$

In this equation $D = k_B T / (6\pi\eta R)$ is the diffusion constant for the translation of the sphere, η and T being the water's viscosity and temperature, respectively. $D_r = k_B T / (8\pi\eta R^3)$ is the rotational diffusion constant and $v_0 = F_{\text{cm}}/\gamma$ the velocity induced by the electromagnetic field, with $\gamma = 6\pi\eta R$ being the friction coefficient. We can write the effective diffusion constant of the persistent random walk as a function of the isotropic optical force

$$D_{\text{eff}} = \frac{1}{\gamma} \left(k_B T + \frac{2F_{\text{cm}}^2 R^2}{9k_B T} \right) \quad (24)$$

which using eq 16 can be written, for the particular case analyzed in this paper, as a function of the laser's intensity:

$$D_{\text{eff}} = \frac{1}{\gamma} \left(k_B T + \frac{1334 \times 10^{-48} \left[\frac{N^2 m^4}{W^2} \right] I^2 R^2}{k_B T} \right) \quad (25)$$

We can test this result by performing Langevin molecular dynamics simulations of the rotating sphere⁴⁷ with the force given by the isotropic radiation field. The mean-squared displacement is expected to fulfill^{44–46}

$$\langle r^2(t) \rangle = 6D_{\text{eff}}t - \frac{F_{\text{cm}}^2}{2\gamma^2 D_r^2} (1 - e^{-t/2D_r}) \quad (26)$$

which, for times much larger than the orientation correlation time ($1/2D_r$), turns into

$$\langle r^2(t) \rangle = 6D_{\text{eff}}t \quad (27)$$

corresponding to a Brownian motion with an effective diffusion constant given by eq 25.

The result of the mean-squared displacement from the molecular dynamics simulation for different values of the intensity of the field is shown in Figure 4. Note how the

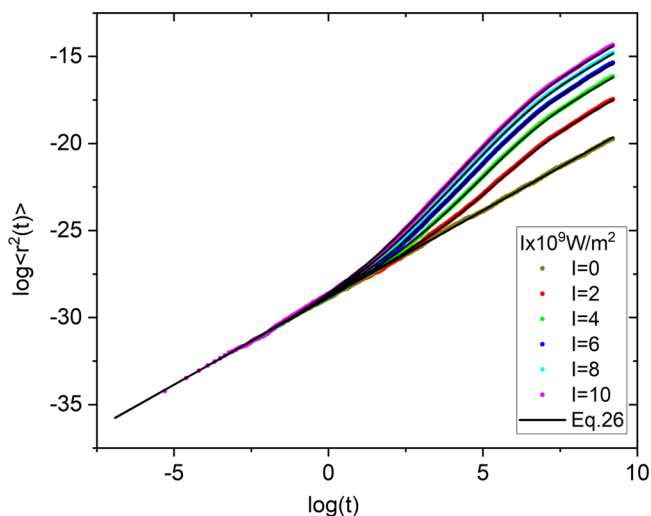


Figure 4. Logarithm of the mean-squared displacement versus logarithm of time for a gold–silver dimer embedded in a polymer sphere with a radius of $5 \mu\text{m}$. The particles are immersed in water at room temperature. The results from the Langevin molecular dynamics simulations for different values of the isotropic random field intensity are shown with colored circles. The continuous line is the analytical result from eq 26 with the effective diffusion constant given by eq 25 and the force on the center of mass given by eq 16.

isotropic random field induces an active motion with a well-defined effective diffusion constant given by eq 25. In Figure 5 we compare the trajectories corresponding with the pure Brownian motion ($I = 0$) and the activated Brownian motion ($I = 4 \times 10^9 \text{ W/m}^2$). The persistent random walk effect due to the activated motion is clearly visible.

Finally, we calculate the Péclet number Pe . The Péclet number compares the diffusive time scale given by $4R^2/D$ (time needed to diffuse the particle's size) with the ballistic time scale $2R\gamma/F_{\text{cm}}$ (time to move a distance equal to the particle's size propelled by the active force). The Péclet number is given by $Pe = 2RF_{\text{cm}}/D\gamma$. Clearly, the larger the Péclet number, the greater the active motion. Typical values of Pe , for spherical active colloids, range from 1 to 100. In Figure 6 we plot the Péclet number and the normalized effective diffusion constant (D_{eff}/D) for different values of the intensity of the electromagnetic radiation. Note how we reach values close to $Pe = 60$ and $D_{\text{eff}}/D = 200$ for an isotropic random field intensity of $I = 10^{10} \text{ W/m}^2$.

It is important to stress the difference between the previously reported random field-based mechanisms used to tune diffusion and those analyzed in this paper. In previous studies, several coherent random fields interfere, producing a speckle pattern intensity distribution.^{27,28} Random trapping of the particle in the large-intensity regions modifies its movement. The final diffusive behavior of the particle is due to the modification of the speckle pattern through time, due to the random fluctuation of the fields. However, in the mechanism analyzed in this paper, the fluctuation of the fields

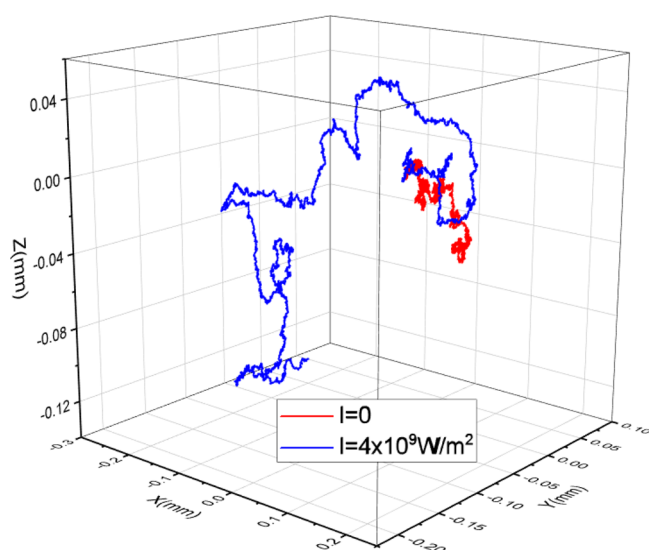


Figure 5. Langevin molecular dynamics simulation of the trajectory of the gold–silver dimer with a length of 90 nm, embedded in a polymer sphere with a radius of 5 μm , in water at room temperature. The results are shown for no illumination corresponding to the pure Brownian case (red line) and for a radiation intensity of $4 \times 10^9 \text{ W/m}^2$, corresponding with the activated Brownian motion (blue line). The total simulation time is 10^4 s .

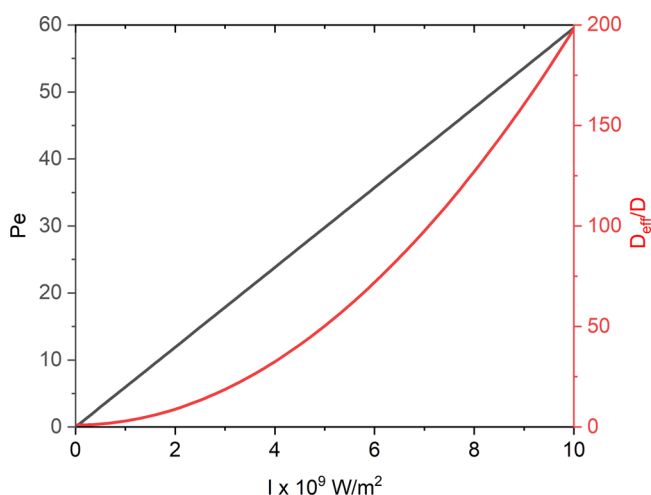


Figure 6. Péclet number (left axis) and normalized effective diffusion constant (right axis) versus isotropic random field intensity for a gold–silver dimer with a length of 90 nm embedded in a polymer sphere with a radius of 5 μm in water, at room temperature.

is fast enough to cancel out all interfering terms and no speckle pattern is formed. This is equivalent to considering that fluctuating waves interact independently with the particles, promoting a net nonreciprocal force responsible for the effective diffusion.

CONCLUSIONS

The binding force in the general case of a dimer made up of two arbitrary nanoparticles, illuminated with an isotropic random fluctuating field, has been calculated. This calculation shows that the interaction force is always reciprocal in the case of nonabsorbing particles, even when their polarizabilities are different and, in the case of two identical absorbing particles, for every value of the absorption term. However, if both

particles are different and at least one of them is absorbent, the forces are no longer reciprocal and the total force on the center of mass of the dimer is different from zero. Also, the condition for the interaction dynamics to be conservative has been obtained. It has been shown that, in the system analyzed, this condition is determined by the reciprocity of the interaction. Since the applied optical field is homogeneous and isotropic, this is an optical scheme that allows, for the first time, inducing an isotropic nonreciprocal and nonconservative force between two nanoparticles, holding for any orientation. This mechanism can be used to induce active Brownian motion with effective diffusion coefficients and remarkable Péclet numbers. All of the analytical predictions have been tested using a plane wave decomposition of the fluctuating field and rotational Langevin molecular dynamics simulations.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsp Photonics.1c01906>.

Numerical calculation of the nonreciprocal force, derivation of eqs 11 and 12, and zero values of the force on the center of mass of the dimer (PDF)

AUTHOR INFORMATION

Corresponding Authors

Jorge Luis-Hita – Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain; Present Address: Universidad Europea de Madrid, 28670 Villaviciosa de Odón, Madrid, Spain; Email: j.luis.hita@facultyue.es

Manuel I. Marqués – Departamento de Física de Materiales, IFIMAC and Instituto de Física de Materiales “Nicolás Cabrera” Universidad Autónoma de Madrid, 28049 Madrid, Spain; orcid.org/0000-0002-8191-743X; Email: manuel.marques@uam.es

Author

Juan José Sáenz – Donostia International Physics Center, 20018 Donostia-San Sebastián, Spain; IKERBASQUE, Basque Foundation for Science, 48013 Bilbao, Spain; orcid.org/0000-0002-1411-5648

Complete contact information is available at:

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Notes

The authors declare no competing financial interest.

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