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Plasmon—Assisted Resonance Energy Transfer Involving Electric and Magnetic Coupling

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Abstract: We develop a quantum theory based on macroscopic quantum electrodynamics to research the resonance energy transfer (RET) between a chiral donor and acceptor. It differs from the previous Green function approach which needs specific boundary conditions to obtain an analytical solution for calculating the RET rate. Our theory can combine the finite-difference time-domain (FDTD) method, which gives a simple and efficient semi-analytical approach, to evaluate the chiral RET rate in an arbitrary plasmonic nanosystem. Applying our theory to the systems of chiral molecules 3-methylcyclopentanone (3MCP) near the achiral/chiral plasmonic nanostructures, the RET process, which is divided into nondiscriminatory and discriminatory parts, is investigated. We find that plasmon will enhance both nondiscriminatory and discriminatory rates compared to the absence of plasmonic nanostructure, but the plasmon supported by chiral nanostructure contributes more to the discriminatory rate. The ratio of discriminatory to nondiscriminatory rates in the system consisting of 3MCP and chiral plasmonic structure is five-fold compared to the system consisting of 3MCP and achiral plasmonic structure. The phenomena can be attributed to the chiral electric-magnetic coupling. Our findings are important in understanding the achiral and chiral electric-magnetic interaction and designing chiral light-harvesting and sensing devices.

Keywords: energy transfer; plasmon; electric-magnetic coupling

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1. Introduction

Resonance energy transfer (RET) is a wide-ranging fundamental process for many natural and artificial phenomena and applications, including light harvesting [1,2], photovoltaics [3–5], and biosensing [6–8]. To understand the basic mechanism by which energy is transferred in biological, chemical, and physical systems, theoretical work has been carried out over the years. At first, the RET process usually occurs in common environments and can be approximated as homogeneous, nondispersive, and nonabsorptive media. Using the classic Förster theory [9] is enough to predict the rate of RET. During the past decade, artificial structures have developed rapidly. It has been reported that RET rates can be modified through different environments, including metal surfaces [10,11], nanoparticles [12–14], cavities [15–17] and other dimensionally constrained nanostructures [18–22], which motivate the theoretical studies. Several theories covering cavity-mediated energy transfer [23], polariton-assisted remote energy transfer [24–26], and plasmon-coupled resonance energy transfer [27] have been developed to investigate the mechanism of the new phenomena. Most of these methods are available to the particular system and cannot treat arbitrary environments.

In recent years, chirality in optics has attracted much attention due to the novel effects of fabricated nanostructures. For chiral molecules, both fluorescence and FRET exhibit discriminatory behavior with respect to optical and material handedness [28]. It has been demonstrated that resonance energy transfer can be used to discriminate between left- and right-handed molecules if the handedness of an object is known [29]. Many important

biological macromolecules and pharmaceuticals are both fluorescent and chiral. It seems to be necessary to investigate the chiral RET process. However, theoretical studies of chiral effects relevant to RET are rarely reported. They are mostly limited to quantum electrodynamical (QED) studies of simple energy transfer between chiral species [30,31] or within a chiral medium [32,33].

In this paper, we present a general computational scheme to investigate the RET process of chiral donor-acceptor pairs near the plasmonic structures. Firstly, the RET rate is described by a fully quantum electrodynamic expression, but its transition matrix elements can be calculated in terms of interactions between transition acceptor dipoles and the corresponding electromagnetic fields generated by the donor transition dipoles. The physical quantities can be obtained using the FDTD method, which gives a simple and useful method to investigate the chiral RET process modified by arbitrary plasmonic structures. Our results show good agreements with analytic solutions from quantum electrodynamics. Secondly, chiral donor-acceptor pairs with fixed dipole orientation are set near the achiral nanorod. Plasmon is found to enhance the pure electric, pure magnetic, and mixed electric-magnetic coupling. The contributions of different dipole terms are comparable when the transition electric and magnetic dipole moments are set uniformly, but magnetic dipole terms are insensitive to the effect of separation distance. Thirdly, choosing 3MCP as an example, the degree of discrimination which describes the ratio of discriminatory and nondiscriminatory parts is obtained as a function of separation distance. It is found that the discriminatory rate can be enhanced by achiral and chiral plasmonic structures. When the donor is near the chiral plasmonic, the degree of discrimination is five-fold compared to the degree without chiral plasmon in the far zone. The phenomena can be attributed to the active contribution of the chiral plasmon to the mixed electric-magnetic coupling. Our findings are important for the design of chiral light-harvesting and sensing devices.

2. Theory and Methods

We use a quantum electrodynamic (QED) theory to describe the interaction between two chiral molecules, chiral donor molecule D and acceptor A, in an arbitrary nanophotonic environment. The schematic illustration is shown in Figure 1. The Hamiltonian contains three components: $H = H_s + H_F + H_{\text{int}}$. Three parts are written as follows [34].

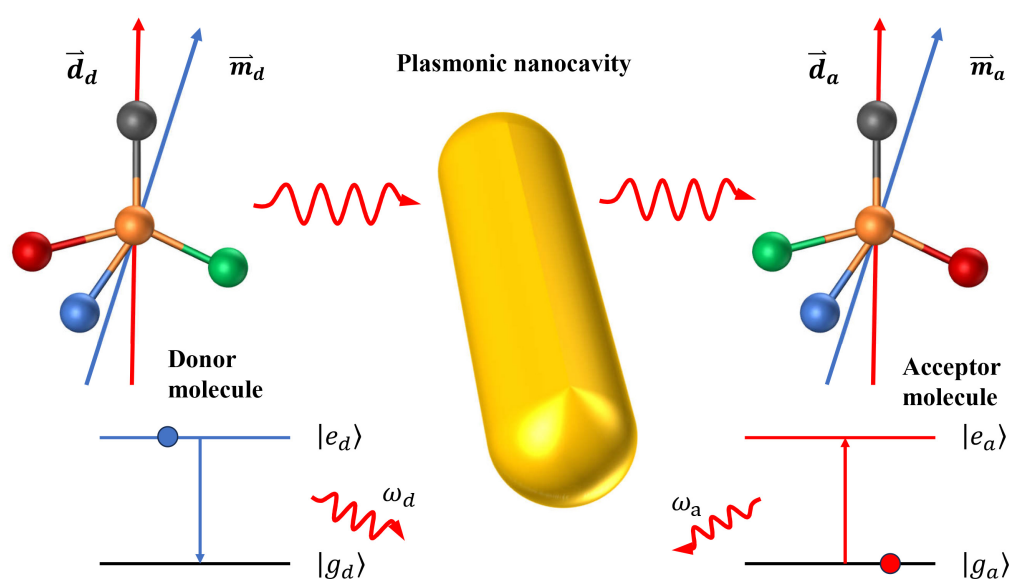


Figure 1. Schematic illustration of plasmon-assisted resonance energy transfer involving chiral molecules.

$$\begin{aligned}
H_s &= \sum_{i=a,d} \sum_{k=e,g} \hbar \omega_i^k |k_i\rangle \langle k_i| \\
H_F &= \sum_{\lambda=e,m} \int d^3r \int_0^\infty d\omega \hbar \omega \hat{f}_\lambda^\dagger(\mathbf{r}, \omega) \hat{f}_\lambda(\mathbf{r}, \omega) \\
H_{\text{int}} &= \sum_{i=a,d} -\hat{\mathbf{d}}_i \cdot \hat{\mathbf{E}}(\mathbf{r}_i) - \hat{\mathbf{m}}_i \cdot \hat{\mathbf{B}}(\mathbf{r}_i)
\end{aligned} \tag{1}$$

where H_s , H_F , H_{int} donate molecule, field, and molecule-field interaction Hamiltonians, respectively. The molecule is described as a two-level fermion with ground state $|g_i\rangle$ and excited state $|e_i\rangle$. H_s is written in terms of unperturbed molecular eigenenergies $\hbar \omega_i^k$ and eigenstates $|k_i\rangle$. H_F is the energy of the electromagnetic field in the medium. $\hat{f}_\lambda^\dagger(\mathbf{r}, \omega)/\hat{f}_\lambda(\mathbf{r}, \omega)$ represent the creation/annihilation operators of elementary electric ($\lambda = e$) or magnetic ($\lambda = m$) excitations of matter. The dynamical variable $\hat{f}_\lambda^\dagger(\mathbf{r}, \omega)$ describes the creation of a photon/polariton in medium, and $\hat{f}_\lambda(\mathbf{r}, \omega)$ describes the absorption of the dissipative system (or the reservoir). These bosonic operators obey the following commutation relations: $[\hat{f}_\lambda(\mathbf{r}, \omega), \hat{f}_{\lambda'}^\dagger(\mathbf{r}', \omega')] = \delta_{\lambda\lambda'} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega')$ and $[\hat{f}_\lambda(\mathbf{r}, \omega), \hat{f}_{\lambda'}(\mathbf{r}', \omega')] = [\hat{f}_\lambda^\dagger(\mathbf{r}, \omega), \hat{f}_{\lambda'}^\dagger(\mathbf{r}', \omega')] = 0$. For interaction term H_{int} , we assume each species to be optically active and ignore multipolar contributions under dipole approximation. $\hat{\mathbf{E}}(\mathbf{r}_i)/\hat{\mathbf{B}}(\mathbf{r}_i)$ is the electric field/magnetic field at the donor's or the acceptor's position $\mathbf{r}_i$. $\hat{\mathbf{d}}_i/\hat{\mathbf{m}}$ is the electric-magnetic dipole moment operator of the donor or acceptor. In dissipative quantum electrodynamics, the electric field can be defined with the Green tensor and bosonic operators as follows:

$$\hat{\mathbf{E}}(\mathbf{r}, \omega) = \sum_{\lambda=e,m} \int d^3r' \int_0^\infty d\omega' \mathbf{G}_\lambda(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{f}_\lambda(\mathbf{r}', \omega) \tag{2}$$

where $\mathbf{G}_e$ and $\mathbf{G}_m$ are the mode tensors introduced in terms of the Green tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ with relation. Written in terms of the inhomogeneous and frequency dependent permittivity ϵ and permeability μ , the forms are as follows [29]:

$$\begin{aligned}
\mathbf{G}_e(\mathbf{r}, \mathbf{r}', \omega) &= i \frac{\omega^2}{c^2} \sqrt{\frac{\hbar}{\pi \epsilon_0}} \text{Im} \epsilon(\mathbf{r}', \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\
\mathbf{G}_m(\mathbf{r}, \mathbf{r}', \omega) &= -i \frac{\omega}{c} \sqrt{\frac{\hbar}{\pi \epsilon_0}} \frac{\text{Im} \mu(\mathbf{r}', \omega)}{|\mu(\mathbf{r}', \omega)|^2} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \times \overleftarrow{\nabla}
\end{aligned} \tag{3}$$

where c is the vacuum speed of light, and $\overleftarrow{\nabla}$ denotes differentiation from the left. The dyadic tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ is the full electromagnetic Green's tensor characterizing the environment and fulfills the Helmholtz equation: $\nabla \times \boldsymbol{\mu}^{-1} \cdot \nabla \times \mathbf{G} = \delta(\mathbf{r} - \mathbf{r}')$. Magnetic fields are connected to the corresponding electric field through $\hat{\mathbf{B}}(\mathbf{r}, \omega) = -\frac{i}{\omega} \nabla \times \hat{\mathbf{E}}(\mathbf{r}, \omega)$.

The perturbative energy transfer rate Γ for incoherent Förster-type resonance energy transfer can be expressed in terms of the transition matrix element M_{fi} using Fermi's golden [35] as $\Gamma = \frac{2\pi}{\hbar^2} \rho |M_{fi}|^2$, where ρ is the density of final states associated with energy. The electric or magnetic transitions in the molecules depend linearly on the electric and magnetic fields. M_{fi} describes perturbation, leading to the interaction between a chiral molecule and a second chiral species via two virtual photon exchanges. It is evaluated from the second-order perturbation theory formula

$$M_{fi} = \sum_j \frac{\langle f | H_{\text{int}} | j \rangle \langle j | H_{\text{int}} | i \rangle}{E_i - E_j} \Big|_{E_i=E_f} \tag{4}$$

with the initial state $|i\rangle = |e_d, g_a, \{0\}\rangle$ and final state $|f\rangle = |g_d, e_a, \{0\}\rangle$. And E is the energy of the respective state. Each matrix element corresponds to an event in which one of the

molecules undergoes a transition accompanied by the emission or absorption of a virtual photon. Taking into account a complete set of intermediate states in the calculation, all possible intermediate states $|j\rangle$ can be written as follows:

$$|j_1\rangle = |g_d, g_a, 1_\lambda(\omega, \mathbf{r})\rangle, \quad |j_2\rangle = |e_d, e_a, 1_\lambda(\omega, \mathbf{r})\rangle$$

corresponding to a single excitation in the electric field ($\lambda = e$) or magnetic field ($\lambda = m$) with both molecules in the ground state, and a single field excitation with both molecules in the excited state, where $|1_\lambda(\omega, \mathbf{r})\rangle = \hat{f}_\lambda^\dagger(\mathbf{r}, \omega)|\{0\}\rangle$ is the single-quantum Fock state of collective, polaritonlike bosonic excitations of electric and magnetic types. According to interaction Hamiltonian, the probability amplitude can therefore be divided into four terms: $M_{fi} = M_{ee} + M_{em} + M_{me} + M_{mm}$. The desired transition matrix elements are as follows:

$$\begin{aligned} M_{ee} &= -\mu_0\omega^2 \mathbf{d}_a \cdot \mathbf{G}(\mathbf{r}_A, \mathbf{r}_D, \omega) \cdot \mathbf{d}_d \\ M_{em} &= i\mu_0\omega \mathbf{d}_a \cdot \mathbf{G}(\mathbf{r}_A, \mathbf{r}_D, \omega) \times \overleftarrow{\nabla}_D \cdot \mathbf{m}_d \\ M_{me} &= i\mu_0\omega \mathbf{m}_a \cdot \overrightarrow{\nabla}_A \times \mathbf{G}(\mathbf{r}_A, \mathbf{r}_D, \omega) \cdot \mathbf{d}_d \\ M_{mm} &= \mu_0 \mathbf{m}_a \cdot \overrightarrow{\nabla}_A \times \mathbf{G}(\mathbf{r}_A, \mathbf{r}_D, \omega) \times \overleftarrow{\nabla}_D \cdot \mathbf{m}_d \end{aligned} \quad (5)$$

where $\mathbf{d}_d$ is the downward dipolar transition in the donor and $\mathbf{d}_a$ is the upward one in the acceptor, with $\mathbf{d}_d = \langle g_d | \hat{\mathbf{d}} | e_d \rangle$ and $\mathbf{d}_a = \langle e_a | \hat{\mathbf{d}} | g_a \rangle$. $\mathbf{m}_d$ and $\mathbf{m}_a$ having analogous definitions. The four terms can easily be verified, as they lead to the same results known from free-space QED [36]. The general Green's tensor determines the value of the transmission matrix. However, the electromagnetic response of a complex photonic system is usually not known analytically, so we can adopt semianalytical models when guided by the full numerical solutions.

The electric field generated from the donor and its relative dyadic Green's function are from classical electrodynamics. Consider that a single donor molecule is located at $\mathbf{r}_D$ in inhomogeneous dispersive and absorbing media. It can be assumed that $\mathbf{P}_e(\mathbf{r}, t) = \mathbf{p}_e(t)\delta(\mathbf{r} - \mathbf{r}_D)$ after making the point-dipole approximation, while the distance between the donor and the acceptor is much larger than the molecular size. The background medium is assumed as a linear time-invariant medium $\mathbf{D}_{\text{med}}(\mathbf{r}, \omega) = \epsilon_r(\mathbf{r}, \omega)\epsilon_0\hat{\mathbf{E}}(\mathbf{r}, \omega)$. Moreover, the total displacement field $\mathbf{D}(\mathbf{r}, t) = \mathbf{D}_{\text{med}}(\mathbf{r}, t) + \mathbf{P}_{\text{ex}}(\mathbf{r}, t)$ and the relative permeability μ is assumed to be 1, then Maxwell's equations are cast into an inhomogeneous partial differential equation by performing the temporal Fourier transform [37]

$$\left(\frac{\epsilon_r(\mathbf{r}, \omega)\omega^2}{c^2} \hat{\mathbf{E}}(\mathbf{r}, \omega) - \nabla \times \{ \nabla \times \hat{\mathbf{E}}(\mathbf{r}, \omega) \} \right) = -\mu_0\omega^2 \mathbf{p}_e(\omega)\delta(\mathbf{r} - \mathbf{r}_D) \quad (6)$$

where $\mathbf{p}_e(\omega)$ is the temporal Fourier transform of $\mathbf{p}_e(t)$. The solution of Equation (6) can be expressed in terms of the dyadic Green's function $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ and consists of two terms: $\hat{\mathbf{E}}(\mathbf{r}, \omega) = \hat{\mathbf{E}}^{(0)}(\mathbf{r}, \omega) + \mathbf{G}(\mathbf{r}, \mathbf{r}_D, \omega)(-\mu_0\omega^2)\mathbf{p}_e(\omega)$. The first term is the homogeneous solution describing the internal electric field in the media and we assume it to be 0; the second term corresponds to a response caused by the external polarization field (for example, the plasmons). Then, we obtain the response electric field at the position of acceptor molecule $\mathbf{r}_A$

$$\frac{\hat{\mathbf{E}}^{\text{ED}}(\mathbf{r}_A, \omega)}{\mathbf{p}_e(\omega)} = -\mu_0\omega^2 \mathbf{G}(\mathbf{r}_A, \mathbf{r}_D, \omega) \cdot \mathbf{e}_{ed} \quad (7)$$

where $\mathbf{p}_e(\omega) = \mathbf{p}_e(\omega)\mathbf{e}_{ed}$ describes the electric dipole source of the donor, and $\mathbf{e}_{ed}$ is the unit vector of the electric transition dipole. We define that the magnetic response of the donor's electric dipole, and the electric and magnetic response of the donor's magnetic dipole at the position of the acceptor are $\hat{\mathbf{B}}^{\text{ED}}(\mathbf{r}_A, \omega)$, $\hat{\mathbf{E}}^{\text{MD}}(\mathbf{r}_A, \omega)$ and $\hat{\mathbf{B}}^{\text{MD}}(\mathbf{r}_A, \omega)$, respectively. The

magnetic dipole source is described as $\mathbf{p}_m(\omega) = p_m(\omega)\mathbf{e}_{md}$. Thus, Equation (5) can be expressed as the corresponding electric-magnetic response result as follows:

$$\begin{aligned} M_{ee} &= |\mathbf{d}_a||\mathbf{d}_d| \frac{\mathbf{e}_{ea} \cdot \hat{\mathbf{E}}^{\text{ED}}(\mathbf{r}_A, \omega)}{p_e(\omega)} \\ M_{em} &= |\mathbf{d}_a||\mathbf{m}_d| \frac{\mathbf{e}_{ea} \cdot \hat{\mathbf{E}}^{\text{MD}}(\mathbf{r}_A, \omega)}{p_m(\omega)} \\ M_{me} &= |\mathbf{m}_a||\mathbf{d}_d| \frac{\mathbf{e}_{ma} \cdot \hat{\mathbf{B}}^{\text{ED}}(\mathbf{r}_A, \omega)}{p_e(\omega)} \\ M_{mm} &= |\mathbf{m}_a||\mathbf{m}_d| \frac{\mathbf{e}_{ma} \cdot \hat{\mathbf{B}}^{\text{MD}}(\mathbf{r}_A, \omega)}{p_m(\omega)} \end{aligned} \quad (8)$$

Here, all electric and magnetic fields and corresponding $p_e(\omega)$ and $p_m(\omega)$ can be obtained from computational electromagnetic software based on the FDTD method. Thus, the numerically exact solutions of the Green function are obtained as well. This provides a convenient scheme for studying resonance energy transfer with arbitrarily complex structures. It should be noticed that the form of the transition matrix elements is only applicable in the weak coupling regime.

3. Results and Discussion

First, we investigate the RET between a donor and acceptor pair with a defined transition dipole orientation in the homogeneous medium. In our simulations, the contribution of electric dipole moments of the donor is evaluated by an oscillating electric point dipole source polarized in the z-direction at the origin (0, 0, 0), while the contribution of magnetic dipole moments is evaluated by a magnetic dipole at the same place. The acceptor is set with equal magnitude and polarization as the donor located on the y-axis (0, R, 0). The separation distance between the donor and acceptor is R. All dipole sources used are achiral, and FDTD simulations are only used to evaluate achiral electric-magnetic responses in our method. Combining Equation (5) with the homogeneous Green function [38] ($k = \sqrt{\epsilon_r}\omega/c$), the analytical transfer matrix elements corresponding to purely electric, mixed electric-magnetic, and purely magnetic couplings are written as follows:

$$\begin{aligned} M_{ee}^{zz}(r_A, r_D) &= \frac{d_a d_d e^{ikR}}{4\pi\epsilon_0\epsilon_r} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \\ M_{em}^{zx}(r_A, r_D) &= \frac{d_a m_d e^{ikR}}{4\pi c\epsilon_0\sqrt{\epsilon_r}} \left(\frac{k^2}{R} + \frac{ik}{R^2} \right) \\ M_{me}^{xz}(r_A, r_D) &= \frac{m_a d_d e^{ikR}}{4\pi c\epsilon_0\sqrt{\epsilon_r}} \left(\frac{k^2}{R} + \frac{ik}{R^2} \right) \\ M_{mm}^{xx}(r_A, r_D) &= \frac{m_a m_d e^{ikR}}{4\pi c^2\epsilon_0} \left(\frac{k^2}{R} + \frac{ik}{R^2} - \frac{1}{R^3} \right) \end{aligned} \quad (9)$$

To confirm the robustness of our method in different media and under different source wavelengths, several media and wavelengths are chosen to test the method. The transition dipole moments are chosen to be $d_d = d_a = m_d/c = m_a/c = 1\text{D}$ for simplicity.

The real parts and imaginary parts of electric dipole-electric dipole coupling terms M_{ee}^{zz} as the function of the separation distance R between the donor and acceptor are shown in Figure 2a,b. Setting the source to be $\lambda = 405\text{ nm}$ and defining the host environment to be vacuum, water, and dimethyl sulfoxide (DMSO) with refractive indexes $n_r = 1, 1.33$ and 1.48 ($\epsilon_r = n_r^2$), the real parts of the numerical results from FDTD (solid lines) are found to match the corresponding analytical results (dash lines) and so are the imaginary parts. The distance-dependent oscillations of the real and imaginary parts of M_{ee}^{zz} can be observed varying with different relative permittivities of media. Larger permittivity

leads to a larger wave vector k , which determines the power of the exponential term e^{ikR} , resulting in a smaller spatial oscillation period of M_{ee}^{zz} . The amplitudes of M_{ee}^{zz} with typical laser wavelengths in the visible light band ($\lambda = 405$ nm, 532 nm, 633 nm, and 785 nm) are shown in Figure 2c. $|M_{ee}^{zz}|$ decreases obviously with the increase in λ . It follows R^{-3} in the near zone and R^{-1} in the far zone. Note that the dipole and monitors must be placed at the correct locations within the Yee cell when numerically calculating the transfer matrix elements by FDTD because the x, y, and z components of the electric and magnetic fields are obtained at different locations within the Yee cell. The real part of M_{ee}^{zz} analytically diverges, while FDTD gives a volume-averaged result over the mesh which is finite as $R \rightarrow 0$. There will be an error between the FDTD results and analytical results for small separations, and reducing the mesh size can decrease the relative error. Thus, we only show a portion of the distance-dependent real parts and amplitudes of M_{ee}^{zz} .

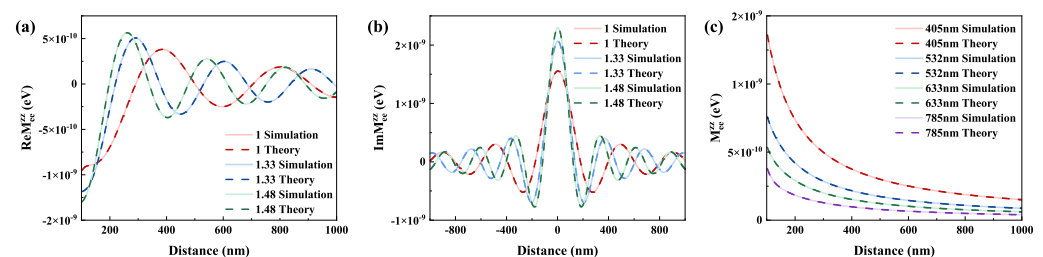


Figure 2. Comparison electric dipole-electric dipole coupling term M_{ee}^{zz} between FDTD results (solid lines) and analytic solution (dash lines) for the dielectric medium cases with different refractive index n_r and wavelengths λ . (a) The real and (b) imaginary parts of M_{ee}^{zz} with $\lambda = 405$ nm, $n_r = 1$ (red), 1.33 (blue) and 1.48 (green). (c) $|M_{ee}^{zz}|$ with $\epsilon_r = 1$, $\lambda = 405$ nm (red), 532 nm (blue), 633 nm (green), and 785 nm (purple).

After the validation of M_{ee} , we move on to the other coupling terms. With $n_r = 1.33$ and $\lambda = 615$ nm, the calculation results are shown in Figure 3. It is obvious that the real parts, imaginary parts, and transition amplitudes from the two methods match each other, respectively. When uniformly setting magnitudes of the electric and magnetic transition dipole moments of the donor and acceptor, the electric dipole-magnetic dipole coupling (M_{ee}) and magnetic dipole-electric dipole coupling (M_{me}) contribute equally to the total energy transfer rate, and the amplitudes of M_{ee} , M_{em} , M_{me} and M_{mm} are comparable. The magnetic transition dipole moment m/c is usually smaller than the electric transition dipole moment d by 2–3 orders for chiral molecules smaller than a few nm [39], and it is why electric-electric dipole coupling is dominant in the RET process and other interactions related to magnetic dipole transitions are neglected. RET rates calculated by the FDTD method are reliable in the separate distance of $R > 20$ nm for our mesh set to 1 nm. We will apply it to more complex environments and discuss it later.

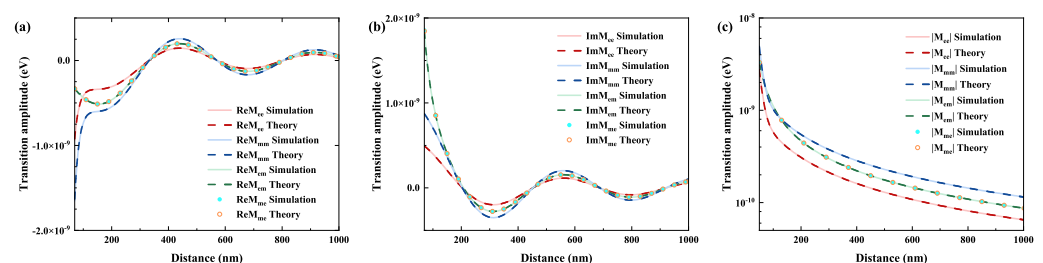


Figure 3. Comparison transfer matrix elements between FDTD results (solid lines) and analytic solution (dash lines) for the dielectric medium cases with $n_r = 1.33$ and $\lambda = 615$ nm. (a) The real parts, (b) the imaginary parts, and (c) the amplitudes of M_{ee} (red), M_{em} (blue), M_{me} (dot), and M_{mm} (green).

We chose a single cuboid Au (core)-Ag (shell) nanorod (Au@Ag NR) which supported the achiral plasmon mode to investigate the plasmon-coupled RET process between a chiral donor-acceptor pair. The geometry setup of Au@Ag NR and the relative positions of NR and the dipoles are shown in Figure 4a. The NR consists of a gold rod core (50 nm long, 16 nm wide, and radius 8 nm of the semi-spherical caps) encased in a silver shell (3 nm thick on the sides and 1 nm thick on the ends). The dielectric constants of gold and silver are adopted from Johnson and Christy [40]. The NR shows a peak of $\lambda = 615$ nm in the normalized scattering spectra, and this wavelength of the scattering maximum is used for every later RET calculation involving Au@Ag NR. The donor and the acceptor, together with the NR, are taken to be in the water ($n_r = 1.33$). All donor and acceptor molecules are approximated by point dipoles. Six sets of the distance of the donor from the surfaces of the NR are chosen for this study, with the distance d marked from d_1 to d_6 ($d = 5, 15, 25, 50, 75$, and 100 nm). Under the condition of whether NR is present or not, the electric and magnetic fields generated from the donor dipole source are calculated for each d .

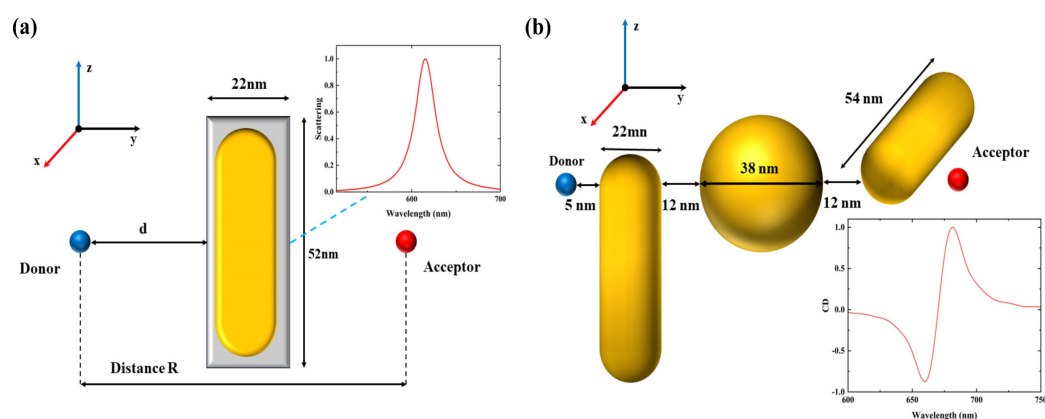


Figure 4. Schematic diagram of donor (blue) and acceptor (red) near achiral/chiral plasmonic nanostructures. (a) Geometry setup of achiral Au (core)-Ag (shell) nanorod (Au@Ag NR) and its normalized scattering spectrum. (b) Geometry setup of chiral nanorod-nanosphere-nanorod (NR-NS-NR) and its normalized CD spectrum.

Polarizing the donor electric dipole in the z -direction and the donor magnetic dipole in the x -direction, while the acceptor has equal polarization, the results of the transition amplitudes are shown in Figure 5a. For a donor at the origin and the acceptor located on the y -axis, the largest enhancement theoretically occurs when the polarization of the donor is parallel to that of the acceptor and the longitudinal modes of localized surface plasmon resonance (LSPR). It is obvious that transition amplitudes have local minima at certain distances near the NR and local maxima on the surface of the NR. The former can be attributed to interference between the dipole electric and magnetic field from the donor and the induced multipole electric and magnetic field from the NR [39], while the latter can be attributed to the localized enhancement effect of the surface plasmon. The response electromagnetic fields of transition electric and magnetic dipoles can be expressed as a superposition of three directional components. We continue to calculate the components in the two remaining directions, and the rotational averaging results, which show the interaction among molecules with arbitrary transition dipole orientations, are shown in Figure 5b. The positions of local maxima and local minima do not change, but the dips in the transition amplitudes are compensated for and smooth by taking the directional average. As shown in Figure 5c, enhancement factors (EFs) are calculated by taking the ratio of transition amplitudes from the case with and without the NR. Immobilizing the distance between the acceptor and surface of the NR to 5 nm, the EF of $|M_{em}|$ (green), $|M_{me}|$ (yellow) show a smooth decay, while that of $|M_{ee}|$ (red) shows a local maximum at a certain distance. In Figure 5d, the distance between the donor and acceptor is immobilized to $R = 127$ nm. The EF of $|M_{ee}|$ is the least sensitive to the position of NR due to the

nonmagnetoelectric environment. When the NR is in an intermediate position between the donor-acceptor pair, the EF of $|M_{ee}|$, $|M_{em}|$, and $|M_{me}|$ can drop below 1. It is reasonable to use our method to find the optimal location of molecules and plasmonic nanostructure to best enhance or suppress the RET process.

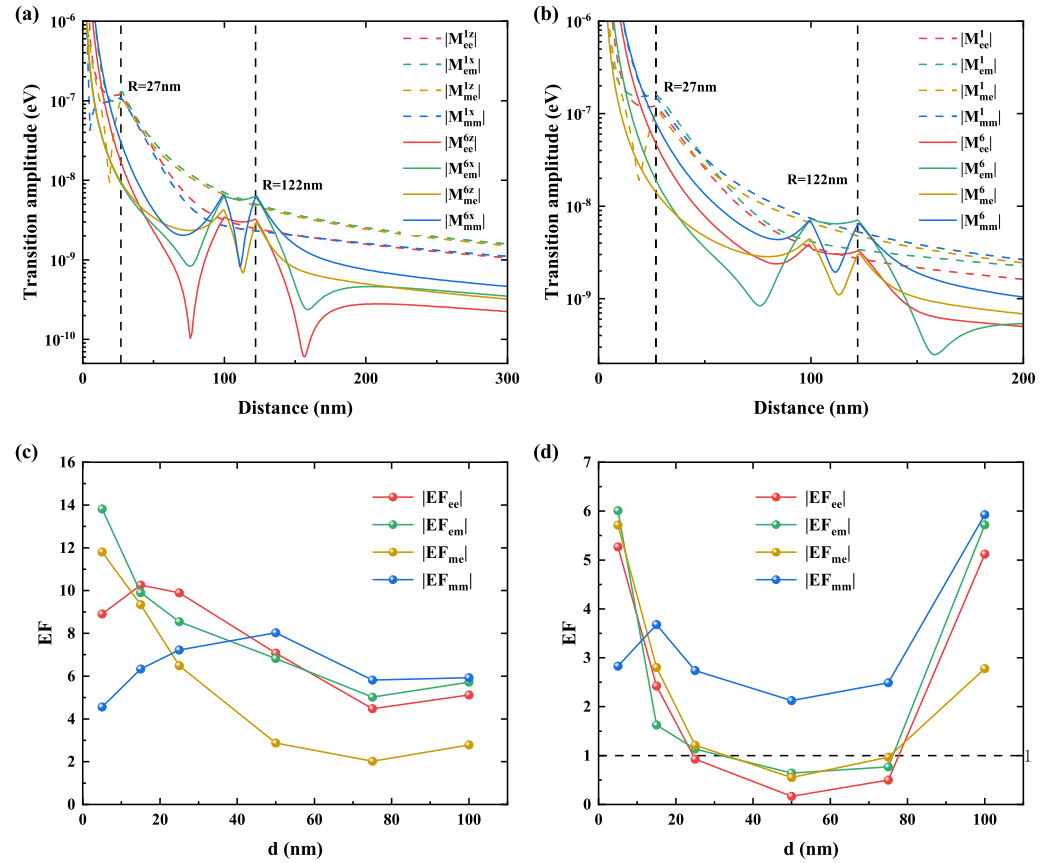


Figure 5. Transition amplitudes with the donor distance $d_1 = 5\text{ nm}$ (dash lines) and $d_6 = 100\text{ nm}$ (solid lines) while the polarization directions of the dipoles are (a) determined and (b) arbitrary. The enhancement factor (EF) of the transition amplitudes is calculated by taking the ratio of $|M_{ee}|$ (red), $|M_{em}|$ (green), $|M_{me}|$ (yellow), and $|M_{mm}|$ (blue) from the simulations with NR and without NR. (c) The acceptor is placed 5 nm from the surfaces of the NR. (d) The acceptor is placed at $R = 127\text{ nm}$.

We turn our attention to the discriminatory RET process. In this work, we follow the definition of discrimination and partition scheme of different contributions to the chiral RET rate in Ref. [29]. Discriminatory and nondiscriminatory rate contributions are written as follows:

$$\begin{aligned}\Gamma_{\text{disc}} &= \frac{2\pi\rho_f}{\hbar} |M_{em} + M_{me}|^2 \\ \Gamma_{\text{nd}} &= \frac{2\pi\rho_f}{\hbar} |M_{ee} + M_{mm}|^2\end{aligned}\quad (10)$$

where ρ_f is the density of the final states. The degree of discrimination is defined as $S = \Gamma_{\text{disc}}/\Gamma_{\text{nd}} = |M_{em} + M_{me}|^2/|M_{ee} + M_{mm}|^2$.

Our nanosystems are embedded in the water, and the local field effects of the medium on the donor and the acceptor transition dipoles lead to an enhancement factor with $c_e = 3\epsilon_r/(1 + 2\epsilon_r)$ [29]. Adding the local field correction (LFC) factor c_e^2 , c_e , and c_e to the relative terms M_{ee} , M_{em} , and M_{me} , we obtain the final expression for degree of discrimination S

$$S = \frac{\Gamma_{\text{disc}}}{\Gamma_{\text{nd}}} = \frac{|M_{em} + M_{me}|^2}{|c_e M_{ee} + c_e^{-1} M_{mm}|^2}\quad (11)$$

In Figure 6a, the degree of discrimination S in the water and near the achiral NR as a function of separation distance R between the donor and the acceptor is plotted. The typical chiral molecule 3-methylcyclopentanone (3MCP) has a comparable electric transition dipole moment and electric transition dipole moment, which leads to a relatively large rotatory strength. Thus, it is chosen as an example of this study. The wavelengths used are $\lambda = 297$ nm for the 3MCP in the water with/without LFC (black solid line/black dash line), and $\lambda = 615$ nm for the cases with/without Au@Ag NR (red/blue solid line). The electric and transition dipole magnetic transition dipoles are $|d| = 2.44 \times 10^{-31}$ cm and $|m|/c = 3.31 \times 10^{-32}$ cm [41]. We obtain similar results to Ref. [29], including the effect of LFC in media and the evaluated S in retarded limits ($S_{r \rightarrow \infty} = 0.08$ with LFC and $\lambda = 297$ nm). But the evaluated S values in nonretarded limits ($S_{r \rightarrow 0}$) are all close to zero due to the endogenous calculation errors for small separations (10 nm). It is one of the problems that need to be optimized and addressed in our future work. We find that plasmon can further enhance the degree of discrimination when the system has been already in a dielectric environment. By placing the donor near ($d = 5$ nm) the NR, the degree of discrimination between two chiral 3MCP molecules can already enhance by roughly 27.5% (from 0.08 to 0.102) in the retarded limits. Though $\lambda = 615$ nm is the maximum scattering of NR but not the transition wavelength for 3MCP, the results are still relevant for the chiral RET process with general chiral molecules.

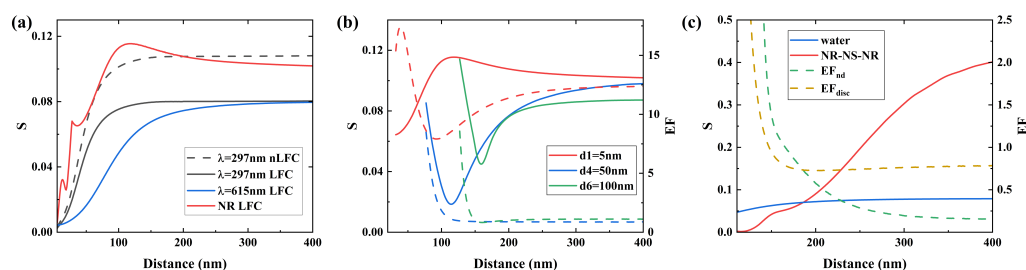


Figure 6. (a) Degree of discrimination S for 3MCP as a function of separation distance R in waters without the local field correction (LFC) (black dashed line) and with LFC (black solid lines). For the case with Au@Ag NR (red solid lines) and without NR (blue solid lines). (b) Degree of discrimination S (solid lines and left y-axis) with $d = 5$ nm (red), 50 nm (blue), and 100 nm (green). The total EF (dash lines and right y-axis) is calculated by taking the ratio of total amplitudes $|M|$ from the cases with NR and without NR. (c) Degree of discrimination S (solid lines and left y-axis) with (red) and without (blue) NR-NS-NR. EF (dash lines and right y-axis) of discriminatory rate (yellow) and nondiscriminatory rate (green).

Figure 6b shows the separation-dependent degree of discrimination S with three different distances as $d = 5$ nm (red solid line), 50 nm (blue solid line), and 100 nm (green solid line) from the donor to the surface of NR. The enhancement effect of plasmon on the degree of discrimination decreases as the donor gets further away from the NR, and the degree appears as a local minimum at the same position as the local minimum in Figure 5a,b. But in the far-zone limit of large distances, the degree of discrimination is still enhanced under the plasmonic effect, and the enhancement effect will eventually disappear with the increasing distance from the donor to the plasmonic nanostructure. Only the donor near the NR ($d = 5$ nm, red dash line) and the total EF (dash lines), which is calculated by taking the ratio of total amplitudes $|M|$ from the cases with NR and without NR, can be enhanced no matter the near-zone limit of small separations or in the retarded or far-zone limit of large distances. It means that utilizing the plasmonic effect to enhance the donor is the better choice rather than the acceptor in the weak coupling regime.

At last, the system of 3MCP coupled with assembled chiral plasmonic chains is investigated to discuss the effect of the chiral plasmonic nanostructure on the chiral RET process. We adopt an experimental configuration for the nanorod-nanosphere-nanorod (NR-NS-NR) from Ref. [42]. As shown in Figure 4b, NR-NS-NR consists of two gold NRs (each 54 nm long and 22 nm wide) and a gold NS (38 nm diameter). The surface-to-surface

distance of two perpendicular rods is 62 nm, and the space between each rod and sphere is 12 nm. To obtain the circular dichroism (CD) spectrum of the structure, we use the FDTD method to obtain absorption and scattering corresponding to left-handed circular polarization (LCP) and right-handed circular polarization (RCP). The circularly polarized sources are generated by two plane waves with perpendicular polarizations and a phase difference of 90 degrees. Then, the CD is given by the difference between extinction (the sum of absorption and scattering) for LCP and RCP. The normalized CD spectrum, which agrees with the experimental measurement, shows a maximum dip at 659 nm and a peak at 682 nm. The wavelength of maximum CD ($\lambda = 682$ nm) is used to set the donor dipole source. The donor is placed at $d = 5$ nm from the surface of the rod. The degree of discrimination S with (red solid line) and without (blue solid line) are shown in Figure 6c. It is obvious that with NR-NS-NR, the degree of discrimination is enhanced to more than 0.4 in the retarded limit, about five-fold compared to that in water and about four-fold compared with achiral Au@Ag NR. The EF of the discriminatory rate (yellow dash line) rapidly decays and tends to change slowly in the retarded limit, while the EF of the nondiscriminatory rate (green dash line) maintains a larger decay rate in the retarded limit and domain in the nonretarded limit. Though the donor is near the NR-NS-NR, the overall RET rate is not enhanced by the plasmonic effect, which may be due to the full 102 nm length structure with a large intrinsic metal loss and the complex interactions of multipolar electromagnetic fields around the particle. The large enhancement of the degree of discrimination can be attributed to the contribution of the chiral plasmonic nanostructure to the electric-magnetic coupling. With our results, we can obtain the conclusion that achiral/chiral plasmon can enhance the RET rate and the degree of discrimination compared to having no plasmon, while chiral plasmon has more potential in the chiral RET control.

4. Conclusions

We develop a quantum theory based on macroscopic quantum electrodynamics to research RET between a chiral donor and acceptor. Combining the FDTD method, all physical quantities can be obtained to calculate the transition matrix elements, which makes us investigate the RET process of chiral molecules near complex achiral and chiral plasmonic structures. The response electromagnetic fields of transition electric and magnetic dipoles are expressed as a superposition of three directional components, which makes it possible to adopt rotational averaging methods to research the interaction among molecules with arbitrary transition dipole orientations. In order to demonstrate our theory approach, the RET process between chiral molecules 3MCP near the plasmonic achiral/chiral nanostructures is investigated. The results show that plasmon will enhance both nondiscriminatory and discriminatory parts of the chiral RET rate. The ratio of discriminatory to nondiscriminatory rates in the system consisting of 3MCP and chiral plasmonic structure is five-fold compared to the system consisting of a 3MCP and achiral plasmonic structure. We find that the chiral plasmon plays an important role in the chiral RET process and can obviously modulate the electric-magnetic coupling. Our findings are important to understand the achiral/chiral electric-magnetic interaction, and design chiral light-harvesting and sensing devices.

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