Surface-plasmon-polariton-assisted dipole–dipole interaction near metal surfaces

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Received January 26, 2011; accepted April 13, 2011;
posted April 21, 2011 (Doc. ID 141243); published May 23, 2011

We investigate the surface plasmon polariton (SPP)-assisted interaction between two dipoles near a metal surface. The radiation energy from a dipole can excite SPPs and transport to another dipole through the channel of the localized SPP modes. This energy transfer can be much more efficient than direct energy transfer via dipole–dipole radiation interaction in free space. A simple analytical model is proposed to describe the underlying physics behind the influence of SPP on the dipole–dipole interaction energy, and it predicts a wide variety of complicated interaction features that agree well with rigorous calculations. © 2011 Optical Society of America

OCIS codes: 240.6680, 240.0240.

In recent years, surface plasmon polaritons (SPPs) have raised extensive interest due to their unique optical properties [1–3]. Previous works showed that quantum dots could couple to nanowires or nanoparticles [1,4,5], and SPPs could propagate along nanoparticles chains [6,7], nanowires [4,8–10], grooves [11,12], wedges [13], and metal films [14–16]. SPP modes of long distance and high mode confinement can exist in some structures [4,6–13], and SPPs can exhibit unidirectional propagation properties along composite nanowires [8], interferometers, and ring resonators [12]. Pustovit and Shahbazyan showed that the existence of metal nanoparticles can influence the interaction among nearby dipoles [17].

Because photons emitted from a quantum dot can be coupled to the propagating SPP modes supported on nanowires and metal films, it is expected that two quantum dots near the metal structures can interact with each other indirectly via SPPs. In this Letter, we study the interaction between two molecules placed above an air–metal interface. By modeling the molecules' two dipoles, we will calculate the dipole–dipole interaction energy under the influence of the metal surface. By comparing with the free-space dipole–dipole interaction energy, the interaction energy enhancement ratio with different frequencies and positions will be discussed. These results may be helpful for understanding the SPP-assisted interaction energy between quantum entities.

The structure we study is depicted in Fig. 1. We consider an interface between a semi-infinite metal Ag and the air background. Two dipoles are placed near the interface in the air side. The vertical distances between the two dipoles and the interface are d_1 and d_2, respectively, and r is the horizontal distance between the two dipoles. We calculate the interaction energy of the two dipoles as a function of dipole distance r both with and without the existence of the semi-infinite metal by the Green's function method [18]. Suppose the field radiating from one dipole $\mathbf{p}_1$, modulating by the metal surface, and exerting on another dipole $\mathbf{p}_2$ is $\mathbf{E}_2$, the total interaction energy between $\mathbf{p}_1$ and $\mathbf{p}_2$ is simply $U = -\mathbf{p}_2 \cdot \mathbf{E}_2$. In comparison, the $\mathbf{p}_1$ and $\mathbf{p}_2$ interaction energy in free space is given by

$$U_0 = \frac{e^{ikr}}{4\pi\varepsilon_0}\left\{\frac{k^2}{r_0}(\mathbf{n}\times\mathbf{p}_1)\cdot(\mathbf{n}\times\mathbf{p}_2) + [3(\mathbf{n}\cdot\mathbf{p}_1)(\mathbf{n}\cdot\mathbf{p}_2) - \mathbf{p}_1\mathbf{p}_2]\left(\frac{1}{r_0^3} - \frac{ik}{r_0^2}\right)\right\}, \quad (1)$$

which is just the standard dipole–dipole interaction. In this equation, $\mathbf{n}$ is the unit vector in the direction of $\mathbf{r}$. In both calculations, we only focus on the magnitude of the interaction energy.

The interaction energy enhancement factor is defined as $F = U/U_0$. Our calculation results are shown in Fig. 2 (solid curves), where d_1 and d_2, are fixed as 10 nm. The dipoles are oriented perpendicularly to the metal surface. The solid black, red, green, and blue curves in Fig. 2 correspond to different dipole radiation wavelengths of 700, 500, 450, and 400 nm, respectively. From these results, we can find that the interaction energy changes depend on the radiation wavelength and the distance between the two dipoles. At some distances, the interaction energy between the two dipoles with the metal interface can be enhanced efficiently.

To understand these results more clearly, we propose a simple analytical model for better and deeper insights. As is well known, the SPP wave vector at the air–metal interface can be written as

$$k_{\mathrm{spp}} = k_0\sqrt{\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+\varepsilon_2}}, \quad (2)$$

where k_0 is the wavenumber of radiation light in vacuum and ε_1 and ε_2 are the permittivity of the air and metal, respectively. As the dipole radiation contains all of the wave vector $\mathbf{k}$ in the near-field region, the SPPs can

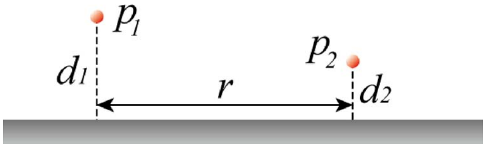

Fig. 1. (Color online) Sketch of our simulations involving two dipoles placed above the surface of semi-infinite Ag.

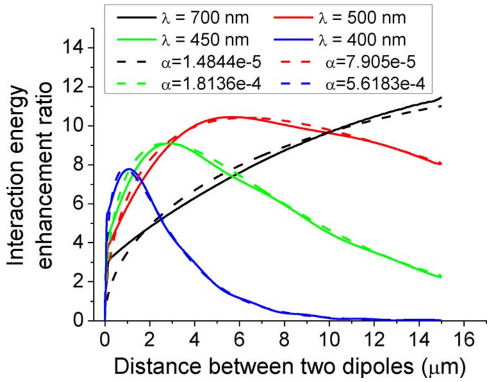

Fig. 2. (Color online) Interaction energy as a function of the distance between two dipoles. The black, red, green, and blue curves correspond to different dipole radiation wavelengths of 700, 500, 450, and 400 nm, respectively. The solid curves are the results of rigorous calculation with the Green's function method, while the dashed curves are calculated with our simple analytical model [Eq. (5)] with an appropriate fitting coefficient of η.

always be stimulated at the air–metal interface due to the phase match with some radiation field. The attenuation of the SPP mode electric field involves contributions from two factors. One factor is the propagation loss due to the metal absorption, which is in the form of exponential attenuation $e^{-\alpha r}$. The other factor comes from the spreading of energy power with respect to the transport distance. For a cylindrical wave in two-dimensional space where the SPP modes are confined, the electric field will decrease in the form of $r^{-1/2}$ as the distance r increases. The magnitude of the electric field at the air–metal interface can thus be expressed as

$$E_{\text{spp}} \sim r^{-1/2} e^{-\alpha r} \eta E_0, \tag{3}$$

where E_0 is the magnitude of incident dipole radiating field; η is a fitting coefficient representing the coupling efficiency between the dipole and the SPP mode; and α is the propagation constant, which is the imaginary part of the SPP wave vector (k_{spp}). The value of α is dependent on the wavelength and the permittivity of the metal. In our cases, the value of α is $1.4844 \times 10^{-5}\ \text{nm}^{-1}$ (for 700 nm), $7.905 \times 10^{-5}\ \text{nm}^{-1}$ (for 500 nm), $1.8136 \times 10^{-4}\ \text{nm}^{-1}$ (for 450 nm), and $5.6183 \times 10^{-4}\ \text{nm}^{-1}$ (for 400 nm), respectively.

On the other hand, the dipole radiation electric field in three-dimensional free space can be expressed as

$$E_{\text{dipole}} \sim r^{-1} E_0. \tag{4}$$

In Eq. (4), r^{-1} is the attenuating factor originating from energy conservation. Because the medium is air, there is no loss. Dividing Eq. (3) by Eq. (4) and considering that the energy of a dipole is $-\mathbf{p} \cdot \mathbf{E}$, we can get the interaction energy enhancement ratio:

$$R = E_{\text{spp}} / E_{\text{dipole}} \sim \eta \sqrt{r} e^{-\alpha r}. \tag{5}$$

By Eq. (5), the values of R with different radiation wavelengths and distances are calculated and shown in Fig. 2. The results from the simple analytical model (dashed curves) agree surprisingly very well with the direct calculation results (solid curves). With Eq. (5), we can explain the results in Fig. 2. In the region within a short distance, the propagation loss is ignorable. As the field attenuation in two-dimensional space is much slower than in three-dimensional space ($r^{-1/2}$ versus r^{-1}), the interaction is enhanced in this region. When the distance is longer, the propagation loss due to the SPP dissipation dominates the field attenuation, so in this region, the interaction between dipoles is degraded.

The results of Fig. 2 indicate that the distance corresponding to the maximum R becomes longer when the radiation wavelength shifts to a longer wavelength. This is because for the metal of Ag, the loss for SPPs (α) is lower at a longer wavelength, and the electric field can propagate farther along the air–metal interface. The results also tell us that for a longer dipole radiation wavelength, the distance range that can enhance the interaction energy between two dipoles becomes wider, and the maximum enhancement factor is relatively higher.

In order to intuitively observe the propagating process of SPPs in the metal surface, we plot in Fig. 3 the electric-field distribution from the source dipole with the radiation wavelength of 500 nm. The source dipole is 100 nm above the metal surface. From this result, we can clearly find that most of the radiation energy is coupled to the SPP mode, and it propagates along the air–metal interface. The same results can also be observed with other radiation wavelengths.

The above results have shown that two dipoles placed near the metal surface will interact with each other via the SPP wave, with the interaction energy enhancement ratio closely related to the stimulated SPP mode and the horizontal distance between the two dipoles. We further take the horizontal distance between the two dipoles r, the distance between the dipole and the metal surface d_1, d_2 as variables and investigate their effects on the interaction energy enhancement ratio. First, we fix $r = 2000$ nm and $\lambda = 500$ nm, and we calculate the interaction energy enhancement ratio as a function of d_1 and d_2, with the results shown in Fig. 4(a). The black curves in Fig. 4(a) are the contour lines with the same energy enhancement ratio at each line. These curves are symmetrical about the $d_1 = d_2$ line. When d_1 and d_2 are short enough (such as d_1, $d_2 < 200$ nm), the contours are approximately straight lines, which indicates that the interaction energy enhancement ratios are the same with the same value of $(d_1 + d_2)$. In addition, for a fixed d_1, we can get the relationship between the interaction energy

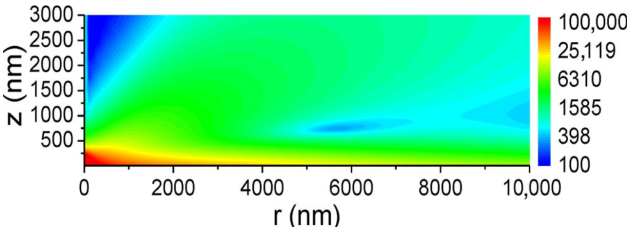

Fig. 3. (Color online) Field distribution of a dipole near the metal interface. The dipole is 100 nm above the metal surface, and the wavelength is 500 nm. The intensity of the electric field is shown in logarithmic scale.

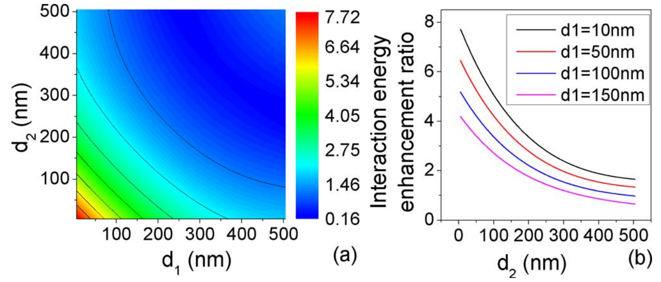

Fig. 4. (Color online) (a) Interaction energy enhancement ratio as a function of d_1 and d_2. Here, we fixed the horizontal distance of $r = 2000\,\mathrm{nm}$, and the dipole radiation wavelength of 500 nm. (b) The enhancement ratios with the change of d_1 (or d_2). The d_2 (or d_1) is fixed at 10, 50, 100, and 150 nm, respectively.

enhancement ratio and d_2, with the results displayed in Fig. 4(b). Here, we select the fixed d_1 at 10, 50, 100, and 150 nm, respectively. The enhancement ratio decreases monotonically with the increasing of d_2. This is because the SPP mode is attenuated exponentially perpendicular to the air–metal surface, and the interaction between the SPP mode and the dipole will be degraded with the increasing of d_2. It should be pointed out that in experiments when the dipole is very close to the surface, the SPP coupling will be quenched.

Next, the interaction energy enhancement ratio as a function of r and d_2 is considered by fixing d_1 and λ. We have considered two conditions of $d_1 = 10\,\mathrm{nm}$, $\lambda = 500\,\mathrm{nm}$ [Fig. 5(a)] and $d_1 = 500\,\mathrm{nm}$, $\lambda = 500\,\mathrm{nm}$ [Fig. 5(b)], respectively. In the first case, due to the strong coupling between the source dipole and the SPP mode, the interaction of the two dipoles is dominated by the SPP process. An enhancement ratio of 10.0 can be reached at some positions. For a fixed horizontal position r, the energy enhancement ratio decreases with larger d_2, while for a fixed vertical position d_2, with the increasing of r, the energy enhancement ratio increases first, and then decreases. At the same time, the contours shows a simple pattern at this condition ($d_1 = 10\,\mathrm{nm}$, $d_2 < 500\,\mathrm{nm}$). For a small d_2 ($d_2 < 300\,\mathrm{nm}$), the contours are almost of the same shape, and for a larger d_2, the contours present more complicated features, but they are still regular. However, when the source dipole is much farther away from the air–metal interface ($d_1 = 500\,\mathrm{nm}$), the interaction energy enhancement ratio becomes low, and the maximum enhancement ratio is only 2.0. The contours in Fig. 5(b) (black curves) appear very complicated. This is because in this condition, the energy of the SPP mode is relatively weak, and parts of the energy will propagate in the free space and interact with each other directly.

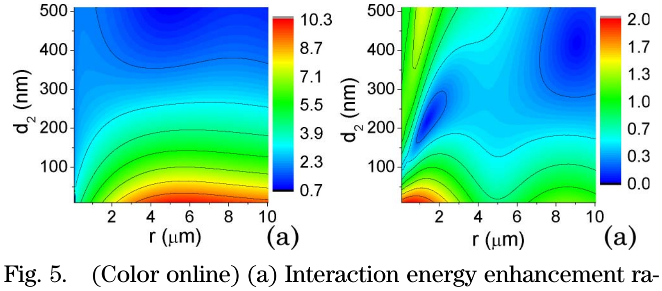

Fig. 5. (Color online) (a) Interaction energy enhancement ratio as a function of r and d_2 (dipole radiation wavelength $\lambda = 500\,\mathrm{nm}$ and $d_1 = 10\,\mathrm{nm}$). (b) $d_1 = 500\,\mathrm{nm}$, and other structure parameters are the same as in (a).

In conclusion, we have shown that SPPs play an important role in modulating the interaction between two dipoles near metal surfaces. The interaction between two dipoles is enhanced at some positions while degraded for other positions. When both dipoles are near the metal surface, the SPP dominates the electromagnetic field. We have proposed a simple analytical model to describe the physical process. The model predicts the maximum interaction position excellently consistent with rigorous calculations. When the distance between the dipoles and the surface is comparable to the wavelength, both the SPP wave and the direct radiation make contributions, and their interference results in very complicated dipole–dipole interaction characteristics. The study is helpful for understanding the mechanism of quantum interaction mediated by SPPs, such as quantum entanglement of qubits mediated by the plasmonic waveguide [19].

This work was supported by the National Natural Science Foundation of China (NSFC) under grants 60736041 and 10874238 and the National Key Basic Research Special Foundation of China under grants 2007CB613205 and 2007CB935703.

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