

Localized exciton states with giant oscillator strength in quantum well in vicinity of metallic nanoparticle

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(Received 4 January 2010; revised manuscript received 7 April 2010; published 8 June 2010)

Effects of the resonant interaction between an exciton in a semiconductor quantum well and localized plasmon excitations in a spherical metal nanoparticle are studied. The calculations show that (1) if the exciton energy level is lower than the energy of the plasmon, the exciton-plasmon interaction may result in the localization of the exciton as a whole in the vicinity of the nanoparticle; (2) a giant (by several orders of magnitude) increase in the oscillator strength of the localized exciton transition takes place near the resonance between the exciton and the plasmon levels. The energy of the lowest excited level of the system was calculated by the variational method, the dependence of the localized exciton energy on the distance between the particle, and the quantum well as well as on the particle radius was obtained.

DOI: [10.1103/PhysRevB.81.235308](https://doi.org/10.1103/PhysRevB.81.235308)

PACS number(s): 73.20.Mf, 71.35.Cc, 73.21.Fg, 78.67.Bf

I. INTRODUCTION

The properties of metal nanoparticles have been studied intensively in different branches of science: physics, chemistry, biology, and medicine.¹ Modern nanofabrication technologies have made possible obtaining metallic nanoparticles with controllable shape and size as well as clusters and arrays of nanoparticles.^{2,3} The unique optical properties of localized surface plasmons⁴ are the subject of considerable interest due to perspectives of applications including the surface-enhanced Raman scattering,^{5,6} biosensors,⁷ the enhancement of electronic transition probability,^{8,9} and optical data storage devices.¹⁰ In the recent decades, hybrid structures consisting of metallic nanoparticles and semiconductor quantum dots^{11–13} or single molecules,^{14,15} semiconductor quantum dots in metal¹⁶ have attracted much attention. It was shown that the emission from a single-quantum dot may be influenced essentially by a small metal nanoparticle in the vicinity.^{11,17} In the present paper, the effect of the exciton-plasmon coupling on the spectra, the character of the motion, and the lifetime of an exciton in a semiconductor quantum well are investigated. The exciton energy spectrum is an essentially continuous exciton band. Therefore, in contrast to the previous studies in which the mixing of the plasmon states with a single level or several discrete levels was considered, we have studied how the presence of a metal nanoparticle may change the continuous spectrum and the propagation of an exciton along the quantum well. Effects of the interaction of quantum-well excitons with a single molecule were studied in Ref. 18. The interaction, considered in our paper, of an exciton with a metallic nanoparticle is much stronger than the interaction with a molecule. As will be shown, this can lead to the significant modification of the spectra, the change in the parameters of optical transitions, and to the localization of the exciton as a whole in the vicinity of the metal particle.

II. MODEL OF THE SYSTEM

We consider a semiconductor crystal with an imbedded spherical metallic nanoparticle and a quantum well (Fig. 1).

The center-to-center distance between the nanoparticle and the quantum well is denoted as d , the radius of the nanoparticle and the well width are denoted as R and L , respectively, z is the direction of the crystal growth. Let us suppose that the energy of the exciton level in the quantum well is close to the localized plasmon level of the nanoparticle. It will be shown that the coupling of these states results in the appearance of distinct features in the exciton spectra. We will assume that the quantum well is deep so the wave functions of the electron and the hole forming the exciton are localized in the quantum-well layer.

The Hamiltonian of the system can be written as

$$\hat{H} = \hat{H}_{el} + \hat{H}_p + \hat{H}_{int}, \quad (1)$$

where \hat{H}_{el} is the Hamiltonian of the electronic subsystem of the quantum well, $\hat{H}_p = \sum_{l,m} \hbar \omega_l \hat{A}_{lm}^+ \hat{A}_{lm}$ is the Hamiltonian of the plasmon subsystem, \hat{A}_{lm}^+ and \hat{A}_{lm} are the creation and the annihilation operators of the plasmon of the frequency ω_l , \hat{H}_{int} is the Hamiltonian of the exciton-plasmon interaction (see Appendix),

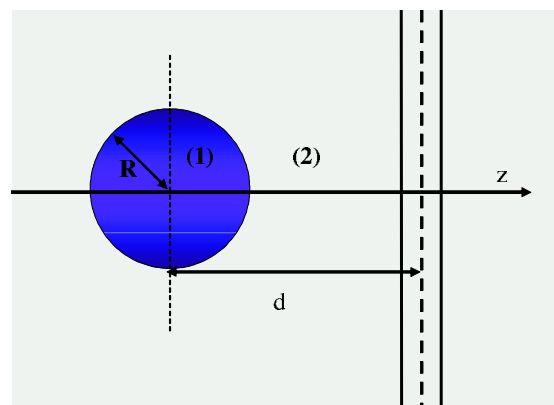


FIG. 1. (Color online) A schematic illustration of the heterostructure containing the quantum well and the metallic nanoparticle.

$$\hat{H}_{\text{int}} = e \sum_{lm} \frac{1}{2} \left(\frac{\hbar}{B_{lm}\omega_l} \right)^{1/2} \sum_i [\varphi_{lm}^{(2)*}(\mathbf{r}_i) \hat{A}_{lm}^+ + \varphi_{lm}^{(2)}(\mathbf{r}_i) \hat{A}_{lm}]. \quad (2)$$

In Eq. (2) \mathbf{r}_i is the coordinate of the i th electron in the quantum well, $\varphi_{lm}^{(1)}(\mathbf{r})$ and $\varphi_{lm}^{(2)}(\mathbf{r})$ are the harmonics of the solution of the Laplace equation for the conductive sphere.¹⁹ We will find the solution of the Schrödinger equation with the Hamiltonian of Eq. (1) in the form of the superposition of two functions,

$$\Psi = \sum_f a_f \Psi_f^{\text{exc}}(\dots \mathbf{r}_i \dots) |0\rangle + \sum_{l,m} b_{lm} \Psi_0(\dots \mathbf{r}_i \dots) \hat{A}_{lm}^+ |0\rangle. \quad (3)$$

The first term in Eq. (3) is the wave function of the system describing the state with the quantum well in the excited state $\Psi_f^{\text{exc}}(\dots \mathbf{r}_i \dots)$ and the metal nanoparticle in the lowest state $|0\rangle$. The second term corresponds to the state with the nanoparticle in the excited plasmon state with the angular orbital momentum l and its projection along a specified axis m and the quantum well in the lowest state $\Psi_0(\dots \mathbf{r}_i \dots)$. The interaction can effect both the translation movement of the exciton and the internal state of the exciton.

It is convenient to present the wave function of the excited state of the crystal as the superposition of the excited states in the Wannier representation,

$$\Psi_f^{\text{exc}}(\dots \mathbf{r}_i \dots) = \sum_{\mathbf{n}, \mathbf{n}', \alpha, \alpha'} F_{\alpha, \alpha'}^f(\mathbf{n}, \mathbf{n}') \Psi_{\mathbf{n}, \mathbf{n}', \alpha, \alpha'}(\dots \mathbf{r}_i \dots), \quad (4)$$

where $\Psi_{\mathbf{n}, \mathbf{n}', \alpha, \alpha'}(\dots \mathbf{r}_i \dots)$ is the antisymmetrized multielectron wave function of the state in which one electron is taken from the valence-band Wannier state α' at the site \mathbf{n}' and placed at the site \mathbf{n} in the conduction-band Wannier state α . The unknown coefficients $F_{\alpha, \alpha'}^f$ determine the overlap function of the exciton and can be found from the Schrodinger equation with the Hamiltonian of Eq. (1).

The interaction H_{int} results in the mixing between the exciton and the plasmon states, which is proportional to the matrix elements of the potential $\varphi_{lm}(\mathbf{r}_j)$ on the Wannier functions $\int \Psi_{\mathbf{n}, \mathbf{n}', \alpha, \alpha'}^*(\dots \mathbf{r}_i \dots) \varphi_{lm}(\mathbf{r}_j) \Psi_0(\dots \mathbf{r}_i \dots) \Pi d\mathbf{r}_i$. Due to the strong localization of the Wannier functions, the matrix element will be nonvanishing only for $\mathbf{n} = \mathbf{n}'$. Besides, the potential $\varphi_{lm}(\mathbf{r}_j)$ varies insignificantly within a unit cell so it can be represented as the power series in $\mathbf{r} - \mathbf{n}'$. As the result, the mixing is proportional to the matrix element of the coordinate $\mathbf{r} - \mathbf{n}'$ between the Wannier functions. Below we will study the exciton-plasmon interaction in the dipole approximation $l=1$, $m=0, \pm 1$. It is known, that in semiconductors, the valence band arises from the p state whereas the conduction band arises from the s state. We will suppose that the spin-orbit splitting is strong. The heavy- and light-hole states in the quantum well are splitted. We will consider the effect of the plasmon excitations on the heavy-hole subsystem. Also we will neglect the mixing of the heavy- and light-hole subbands. It is known²⁰ that in the semiconductor with the zinc-blende structure, the following wave functions can be

chosen to diagonalize the Hamiltonian for the heavy-hole subband:

$$\psi_{3/2, +3/2}^h = -\frac{1}{\sqrt{2}}(|X\rangle + i|Y\rangle)\chi_{\uparrow},$$

$$\psi_{3/2, -3/2}^h = \frac{1}{\sqrt{2}}(|X\rangle - i|Y\rangle)\chi_{\downarrow}$$

and for the conduction band

$$\psi_{1/2, +1/2}^e = |S\rangle\chi_{\uparrow},$$

$$\psi_{1/2, -1/2}^e = |S\rangle\chi_{\downarrow},$$

where χ_{\uparrow} and χ_{\downarrow} are the spin functions, $|X\rangle$ and $|Y\rangle$ are the p -like Bloch functions of the valence-band states, and $|S\rangle$ is the s -like function of the conduction band. The matrix elements of the momentum operator for the interband transitions are equal to

$$\begin{aligned} \langle \psi_{1/2, \pm 1/2}^e | p_x | \psi_{3/2, \pm 3/2}^h \rangle &= \mp i \langle \psi_{1/2, \pm 1/2}^e | p_y | \psi_{3/2, \pm 3/2}^h \rangle \\ &= \mp \frac{1}{\sqrt{2}} \langle S | p_x | X \rangle, \end{aligned}$$

$$\langle \psi_{1/2, \pm 1/2}^e | p_z | \psi_{3/2, \pm 3/2}^h \rangle = 0.$$

Therefore, in this case the interaction will mix only the plasmon state $m=1$ or $m=-1$ with one of the exciton transitions for which the electron- and the hole-spin projections are equal to $(\frac{1}{2}, \frac{3}{2})$ or $(-\frac{1}{2}, -\frac{3}{2})$. So, the problem is reduced to the study of the mixing of a single-plasmon oscillation with a certain exciton state in the quantum well. In this case, Eq. (3) may be presented in the following form:

$$\psi_m = a[\Psi_m^{\text{exc}}(\dots \mathbf{r}_i \dots) |0\rangle + b\Psi_0(\dots \mathbf{r}_i \dots) \hat{A}_m^+ |0\rangle], \quad (5)$$

where b is the unknown parameter, which describes the share of the plasmon state in the state of the system, $a = (\sqrt{1 + |b|^2})^{-1}$, the index m is equal to $+1$ or -1 for the plasmon states and $(\frac{1}{2}, \frac{3}{2})$ or $(-\frac{1}{2}, -\frac{3}{2})$ for the exciton states, respectively.

It will be shown below that the exciton-plasmon interaction results in the exciton localization as a whole in the vicinity of the nanoparticle. We will find the lowest-energy state of the system by the variational method. Let us evaluate the average energy of the system,

$$\begin{aligned} \bar{H} &= \langle \psi_m | \hat{H} | \psi_m \rangle \\ &= \frac{1}{1 + |b|^2} \{ \bar{H}_{\text{QW}} + |b|^2 (\hbar\omega_1 + E_0) + bV_m^* + b^*V_m \}, \quad (6) \end{aligned}$$

where $\bar{H}_{\text{QW}} = \langle \Psi_m^{\text{exc}} | H_{\text{el}} | \Psi_m^{\text{exc}} \rangle$,

$$V_m = \langle \Psi_0 | \frac{e}{2} \left(\frac{\hbar}{B_{1m}\omega_1} \right)^{1/2} \sum_i \varphi_m^{(2)*}(\mathbf{r}_i) | \Psi_m^{\text{exc}} \rangle.$$

$E_0 = \langle \Psi_0 | \hat{H}_{\text{el}} | \Psi_0 \rangle$ is the energy of the lowest state of the quantum well, $\omega_1 = \omega_p / \sqrt{\epsilon^{(1)} + 2\epsilon_{\infty}^{(2)}}$, ω_p is the plasma frequency, $\epsilon^{(1)}$ and $\epsilon_{\infty}^{(2)}$ are the frequency independent part of the dielec-

tric constant of the nanoparticle and the high-frequency dielectric constant of the quantum well, respectively.

The matrix element \bar{H}_{QW} has to be calculated for the multielectron wave functions. The method, described in Ref. 21, allows us to write down the matrix element \bar{H}_{QW} using the envelope exciton wave function $F(\mathbf{n}, \mathbf{n}')$,

$$\bar{H}_{\text{QW}} = E_0 + \sum_{\mathbf{n}, \mathbf{n}'} F^*(\mathbf{n}, \mathbf{n}') H_{\text{exc}} F(\mathbf{n}, \mathbf{n}'), \quad (7)$$

where H_{exc} is the Hamiltonian of the Wannier exciton in the semiconductor crystal in the effective-mass approximation. The coefficient b in Eq. (5) is the variational parameter of the problem. Other variational parameters, which determine the envelope exciton wave function $F(\mathbf{n}, \mathbf{n}')$, will be introduced later. By minimizing the energy functional Eq. (6) with respect to b , one can obtain

$$b = \frac{V_m}{\bar{H} - \hbar\omega_1 - E_0}, \quad (8)$$

$$\begin{aligned} \bar{E} &= \sum_{\mathbf{n}, \mathbf{n}'} F^*(\mathbf{n}, \mathbf{n}') H_{\text{exc}} F(\mathbf{n}, \mathbf{n}') - \frac{\hbar}{4B_{1m}\omega_1} \\ &\times \frac{1}{\hbar\omega_1 - \bar{E}} \left| \sum_{\mathbf{n}} (\nabla\varphi_{1m}^{(2)}(\mathbf{n}) \cdot e\mathbf{r}_m) F(\mathbf{n}, \mathbf{n}) \right|^2, \quad (9) \end{aligned}$$

where $\bar{E} = \bar{H} - E_0$ is the energy of the excitation of the system and $e\mathbf{r}_m^*$ is the interband matrix element of the dipole operator of the chosen exciton state m . The first term in Eq. (9) determines the exciton energy in the quantum well in absence of the resonance interaction with the metallic nanoparticle. The second term is the energy of the electric dipole in the nanoparticle electric field and describes the exciton-plasmon dipole interaction. It arises from the resonance interaction between the plasmon level and the electron levels in the quantum well. To interpret this term, let us write it in the following form:

$$- \sum_{\mathbf{n}, \mathbf{n}'} F_m^*(\mathbf{n}', \mathbf{n}') M(\mathbf{n}, \mathbf{n}') F_m(\mathbf{n}, \mathbf{n}), \quad (10)$$

where

$$\begin{aligned} M(\mathbf{n}, \mathbf{n}') &= - \frac{\hbar}{4B_{1m}\omega_1(\hbar\omega_1 - \bar{E})} \\ &\times [\nabla\varphi_{1m}^{(2)}(\mathbf{n}') \cdot e\mathbf{r}_m^*][\nabla\varphi_{1m}^{(2)*}(\mathbf{n}) \cdot e\mathbf{r}_m]. \quad (11) \end{aligned}$$

The interaction given by Eq. (10) describes the motion of the exciton as the whole in the quantum well with the matrix element of the resonance interaction $M(\mathbf{n}, \mathbf{n}')$. The exciton with the center of mass at the cite \mathbf{n} annihilates with creation of the plasmon and then the plasmon annihilates creating an exciton with the center of mass at a different site \mathbf{n}' . Such motion of the exciton reminds the propagation by the resonant Frenkel's mechanisms, however the transition of the electronic excitation occurs not directly from a certain point to another but through the excitation of the plasmon in the nanoparticle. The interaction between the Frenkel exciton lo-

calized in one material with the Wannier-Mott exciton localized in another one was studied in Ref. 22. In the presented paper, the character of the exciton movement changes from the Wannier-type to the Frenkel-type due to the resonance interaction with the nanoparticle in the same material. The resonance interaction leads to the repulsion of the energy levels. If the exciton energy level is lower than that of the plasmon, its level moves down and the exciton is attracted as a whole to the nanoparticle. In the opposite case, the exciton is repelled from the particle. The exciton radius is much larger than the period of the crystal lattice. This allows replacing the summation by the integration in Eq. (9) and introducing the envelope Wannier exciton wave function $F(\mathbf{n}, \mathbf{n}') \rightarrow \frac{1}{\Omega_0} \Psi(\mathbf{r}_e, \mathbf{r}_h)$, Ω_0 is the primitive cell volume. Taking into account also the smoothness of the wave function, the Hamiltonian of the exciton H_{exc} can be written as

$$\begin{aligned} H_{\text{exc}} &= E_g - \frac{\hbar^2}{2m_e} \Delta_e - \frac{\hbar^2}{2m_h} \Delta_h - \frac{e^2}{\varepsilon_0^{(2)} |\mathbf{r}_e - \mathbf{r}_h|} \\ &+ W_{\text{QW}}(z_e, z_h) - U_{\text{img}}(\mathbf{r}_e, \mathbf{r}_h), \quad (12) \end{aligned}$$

$$\begin{aligned} U_{\text{img}}(\mathbf{r}_e, \mathbf{r}_h) &= - \frac{e^2 R}{2r_e \varepsilon_0^{(2)}} \left\{ \frac{1}{\left| \mathbf{r}_e - \frac{R^2}{r_e^2} \mathbf{r}_e \right|} - \frac{1}{\left| \mathbf{r}_h - \frac{R^2}{r_e^2} \mathbf{r}_e \right|} + \frac{1}{r_h} - \frac{1}{r_e} \right\} \\ &+ \frac{e^2 R}{2r_h \varepsilon_0^{(2)}} \left\{ \frac{1}{\left| \mathbf{r}_e - \frac{R^2}{r_h^2} \mathbf{r}_h \right|} - \frac{1}{\left| \mathbf{r}_h - \frac{R^2}{r_h^2} \mathbf{r}_h \right|} + \frac{1}{r_h} - \frac{1}{r_e} \right\}. \quad (13) \end{aligned}$$

Here E_g is the energy gap for the semiconductor of the quantum well, $W_{\text{QW}}(z_e, z_h)$ is the potential of the quantum well, $\varepsilon_0^{(2)}$ is the low-frequency dielectric constant of the semiconductor, and $m_{e(h)}$ is the electron (the hole) effective mass. The image charge potential $U_{\text{img}}(\mathbf{r}_e, \mathbf{r}_h)$ takes into account the low-lying electron excitations of the nanoparticle with the frequencies much less than the plasma frequency.

We will look for the localized exciton states, which appear due to the nanoparticle. There are the following qualitative reasons for the possibility of the localized states. The resonant interaction for the considered disposition of levels leads to the lowering of the exciton energy, which decreases with increasing the distance between the exciton and the nanoparticle. Therefore, the resonant interaction creates a potential well for the exciton in which it can be localized. The variational function of the lowest exciton state, describing the localization of the exciton as a whole due to the interaction with the plasmon, may be chosen in the form, typical for the excitons in deep quantum wells,

$$\Psi(\mathbf{r}_e, \mathbf{r}_h) = f_e(z_e) f_h(z_h) \Psi_{2D}(r_{\parallel}^e, r_{\parallel}^h), \quad (14)$$

$$\Psi_{2D}(r_{\parallel}^e, r_{\parallel}^h) = \frac{2}{\pi\alpha\beta} \exp\left[-\frac{R_{\parallel}}{\beta} - \frac{r_{\parallel}}{\alpha}\right], \quad (15)$$

$$f_{e(h)}(z_{e(h)}) = \sqrt{\frac{2}{L}} \sin\left(\frac{\pi z_{e(h)}}{L}\right), \quad (16)$$

where α and β are the variational parameters corresponding to the radius of the electron-hole coupling within the exciton and the radius of the localization of the exciton center of mass, respectively, $\mathbf{r}_{\parallel}^{e(h)}$ is the coordinate of the electron (the hole) in the plane of the quantum well, $\mathbf{r}_{\parallel} = \mathbf{r}_{\parallel}^e - \mathbf{r}_{\parallel}^h$, $\mathbf{R}_{\parallel} = (m_e \mathbf{r}_{\parallel}^e + m_h \mathbf{r}_{\parallel}^h) / M$ is the coordinate of the center of mass of the exciton in the plane of the quantum well. If the well width is small ($L \ll R, d$), the potential $\phi_{1m}^{(2)}(\mathbf{r})$ varies only slightly with z inside the quantum well so one can obtain

$$E_m^{2D}(\alpha, \beta) = \int \Psi_{2D}(\mathbf{r}_{\parallel}^e, \mathbf{r}_{\parallel}^h) H_{\text{exc}}^{2D} \Psi_{2D}(\mathbf{r}_{\parallel}^e, \mathbf{r}_{\parallel}^h) d\mathbf{r}_{\parallel}^e d\mathbf{r}_{\parallel}^h - \frac{e^2 \hbar}{4m_0^2 \omega_0^2 B_{1m} \omega_1 [\hbar \omega_1 - E_m^{2D}(\alpha, \beta)]} \left| \int \Psi_{2D}(r_{\parallel}, r_{\parallel}) \times [\nabla \phi_{1m}^{(2)}(d, \mathbf{r}_{\parallel}) \cdot \mathbf{p}_m] d\mathbf{r}_{\parallel} \right|^2, \quad (17)$$

$\mathbf{p}_m = im_0 \omega_0 \mathbf{r}_m$ is the interband matrix element of the momentum operator of the exciton state.

III. CALCULATIONS AND DISCUSSION

As mentioned above, we will consider the system for which the plasmon energy is higher than the exciton energy. The effect of the mixing of the plasmon and the exciton levels is strongest in the case of the resonance of these levels. However, we will assume that the distance between the plasmon and the exciton levels $\Delta E = \hbar \omega_1 - E_{\text{exc}}$ is greater than the width of the plasmon level ($\Gamma \sim 0.1$ eV). In this case, the system levels are separated into two types: excitonlike, in which the share of plasmon excitation is small, and plasmonlike, in which the share of the exciton is small. In our calculations, the parameter b do not exceed 0.2. Nevertheless, as will be shown, even in such condition the effect of the nanoparticle is strong. We will look for the lowest level of the Eq. (17) for the variational wave function of Eq. (14). Since, as assumed, the exciton level is situated lower than the plasmon level and they should be more effective in emission spectra, we will talk about the exciton state later, during the discussion of the results.

The calculations were performed for a silver nanoparticle with the plasma frequency $\omega_p = 1.7601 \times 10^{16}$ rad/s, $\varepsilon^{(1)} = 8.926$ (Ref. 23), the parameters of the semiconductor were chosen to be close to the ZnTe quantum well with the (Zn,Be)Te barriers,^{24–26} the high-frequency dielectric constant of ZnTe $\varepsilon_{\infty}^{(2)} = 6.9$, the static dielectric constant $\varepsilon_0^{(2)} = 9.8$, the electron effective mass $m_e = 0.12m_0$, the hole effective mass $m_h = 0.6m_0$,²⁶ m_0 is the mass of the free electron, and p_m was determined from the longitudinal-transverse splitting according to the condition²⁷ $\omega_{LT} = 4e^2 |p_m|^2 / \hbar \omega_0^2 m_0^2 a_B^3 \varepsilon_{\infty}^{(2)}$, where the longitudinal-transverse splitting of the exciton state $\hbar \omega_{LT} = 1.5$ meV (Ref. 28). The plasmon level $l=1$ of the metal nanoparticle inside the semiconductor for the mentioned parameters is equal $\hbar \omega_1 = 2.43$ eV. We studied the effects of the exciton-plasmon

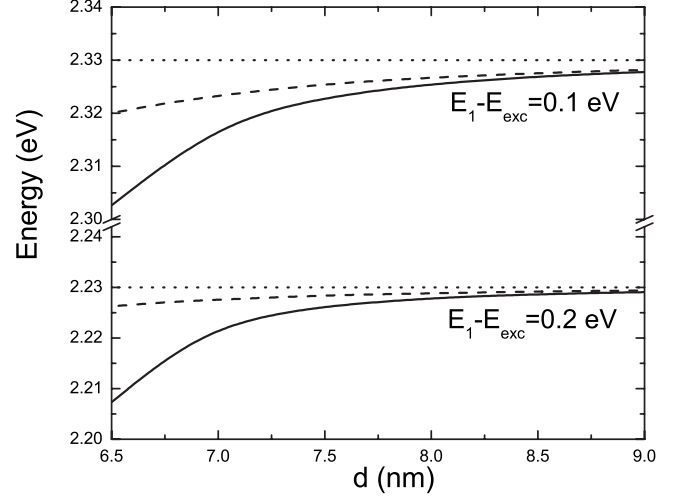


FIG. 2. Energy of the lowest exciton state with (solid line) and without (dashed line) the interaction with the image charges as the function of the distance d between the particle and the quantum well for two values of the resonance detuning $\Delta E = 0.1$ and 0.2 eV. Here $E_1 = \hbar \omega_1$ is the energy of the first excited plasmon level. The radius of the nanosphere $R = 60$ Å. The horizontal dotted lines denote the exciton energy E_{exc} in the absence of the exciton-plasmon interaction.

mixing in dependence on the position of the bottom of the exciton band, which is determined by the composition of the quantum well and the barrier layers.

Application of the variational method gives the following results. The dependence of the energy of the lowest exciton state on distance d between the particle and the quantum well is presented in Fig. 2. One can see that the coupling between the exciton and the nanosphere leads to the considerable decrease in the exciton energy. The localization of the center of mass of the exciton due to the attraction to the particle occurs. The corresponding binding energy and the localization radius for the center of mass of the exciton depend on the distance to the particle d as well as on the radius of the nanosphere (Figs. 3 and 4). The binding energy of the exciton to the nanoparticle is order of several tens of millielectron volt. The calculations show that the interaction of the exciton with the image charges strengthen the attraction between the exciton and the particle for the small particle-well distances d but their effect decreases rapidly with increasing the distance. One can see in Fig. 5 that the exciton-plasmon interaction affects the exciton radius insignificantly, yet it considerably changes the degree of the localization of the exciton. For the small distances, the exciton level position is essentially influenced by the interaction with the image charges. As the distance rises, the exciton energy is determined by the more long-range resonance interaction with the plasmons.

The optical properties of the considered system are characterized by the the oscillator strength of the transition between the ground state $\Psi_0|0\rangle$ and the lowest excited state ψ_m [see Eq. (5)],²⁹

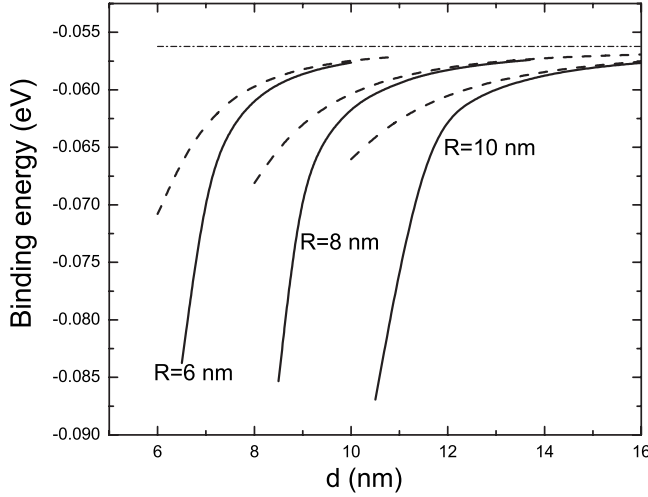


FIG. 3. The binding energy of the exciton for nanoparticles of different radii as the function of the distance between the quantum well and the nanoparticle with (solid line) and without (dashed line) taking into account the interaction with the image charges. The horizontal dashed-dotted line denotes the exciton binding energy in the absence of the exciton-plasmon interaction. The resonance detuning $\Delta E=0.1$ eV.

$$F_{01} = \frac{2m_0\bar{E}}{\hbar^2 e^2} |\mathbf{P}_{01}|^2, \quad (18)$$

where \bar{E} is the energy of the transition, determined from the minimization of the energy of the system given by Eq. (9), \mathbf{P}_{01} is the matrix element of the dipole moment, calculated for the wave functions of the initial and the final states of the system. The operator of the dipole moment consists of two

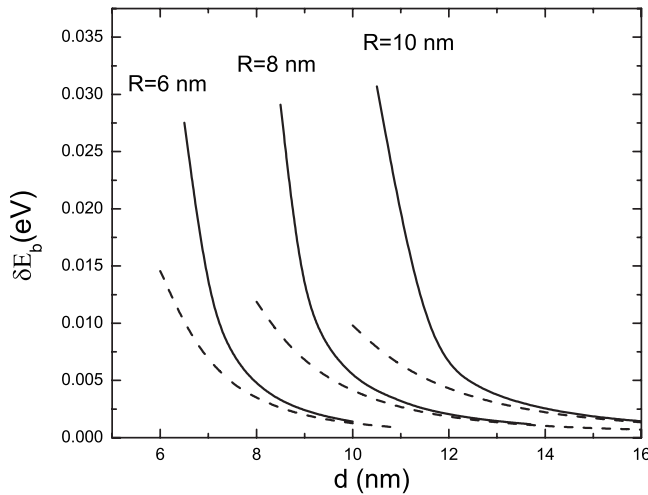


FIG. 4. The energy of the binding of the exciton to the nanoparticle $\delta E_b = |E_b - E_{b0}|$, where E_b is the exciton binding energy and E_{b0} is the exciton binding energy in the quantum well far from the nanoparticle), as the function of the distance between the quantum well and the nanoparticle with (solid line) and without (dashed line) the interaction with the image charges. The resonance detuning $\Delta E=0.1$ eV.

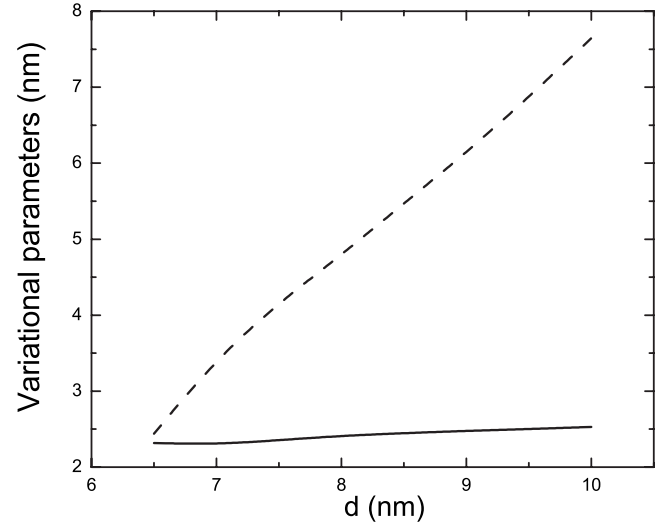


FIG. 5. The variational parameters α (solid) and β (dashed), which determine, respectively, the exciton radius and the radius of the localization of the exciton center of mass for $R=6$ nm as the function of the distance between the quantum well and the nanoparticle. The resonance detuning $\Delta E=0.1$ eV.

parts: the dipole moments of the nanoparticle and that of the semiconductor,

$$\mathbf{P} = \mathbf{P}_{\text{met}} + \mathbf{P}_{\text{sc}} \quad (19)$$

As shown in Appendix, the operator of the dipole moment of the nanoparticle can be written as

$$\begin{aligned} \mathbf{P}_{\text{met}} = & \sum_{lm} \frac{\omega_p^2}{8\pi\omega_l^2} \left(\frac{\hbar}{B_{lm}\omega_l} \right)^{1/2} \\ & \times \left[A_{lm} \int_{r<R} \nabla \varphi_{lm}^{(1)}(\mathbf{r}) d\mathbf{r} + A_{lm}^+ \int_{r<R} \nabla \varphi_{lm}^{(1)*}(\mathbf{r}) d\mathbf{r} \right] \end{aligned} \quad (20)$$

and the operator of the dipole moment of the semiconductor is the sum of the dipole moments of the electrons in the semiconductor crystal,

$$\mathbf{P}_{\text{sc}} = e \sum_i \mathbf{r}_i, \quad (21)$$

\mathbf{r}_i is the coordinate of the i th electron.

To calculate the oscillator strength, one has to calculate the matrix element of the operator of Eq. (19) with the multi-electron wave functions of the ground state $\Psi_0|0\rangle$ and the lowest excited state ψ_m . The matrix element of the operator \mathbf{P}_{sc} is equal to $\mathbf{P}_{\text{sc},01} = e \mathbf{r}_m^* \int \Psi_{2D}^*(\mathbf{r}, \mathbf{r}) d\mathbf{r}$. The obtained results are shown in Fig. 6. Near the nanoparticle, the oscillator strength has a maximum and decreases as the particle-well distance vanishes (the solid line). This happens due to the decrease in the exciton level due to the interaction with the image charges. This maximum is absent for the calculations in which the interaction with the image charges are not taken into account (the results shown with dashed lines). The intensity of the transition decreases with increasing the particle-well distance in a certain range, shown in Fig. 6. For

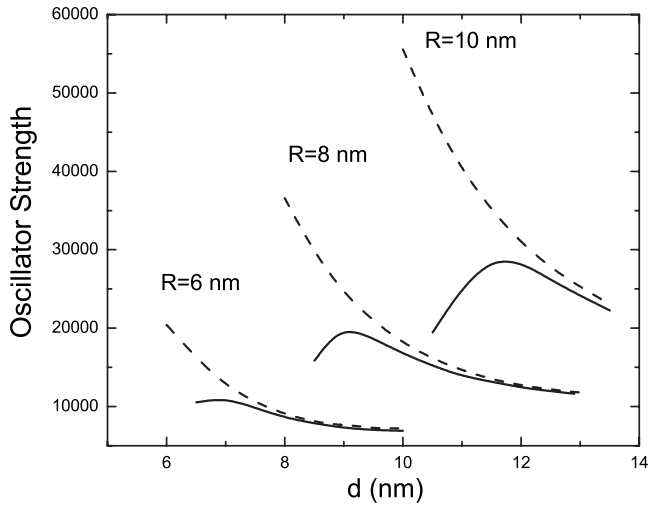


FIG. 6. The exciton oscillator strength as the function of the nanoparticle-well distance with (solid lines) and without (dashed lines) interaction with the image charges. The resonance detuning $\Delta E = 0.1$ eV.

even further increase in the distance, the interaction between the exciton and the particle decreases, and, consequently, the localization radius β grows. The oscillator strength of the optical transition decreases for a further increase in the distance between the nanoparticle and the quantum well. As can be seen in Fig. 6, in this range of distances, the oscillator strength is large due to the mixing of the exciton states in the quantum well with the states of the surface plasmon. The main contribution to the matrix element of the dipole moment of the transition, the square of which is proportional to the oscillator strength, comes from the dipole moment of the transition in the metal nanoparticle. However, the contribution of the dipole moment of the exciton transition to the matrix element rises with increasing the distance from the quantum well, and the role of the quantum-well states in the formation of the oscillator strength increases. For example, for the nanoparticle with the radius $R = 6$ nm and for the detuning $\Delta E = 0.1$ eV, the ratio of the square of the matrix element of the dipole moments of the plasmon transition to that of the exciton transition $\frac{|P_{\text{met},01}|^2}{|P_{\text{sc},01}|^2}$ is equal to 3.6 for the distance value $d = 8$ nm, and is equal to 0.5 for $d = 10$ nm. The oscillator strength begins to increase as the particle-well separation exceeds a certain value (this region is not shown in Fig. 6). This increasing is caused by the expansion of the delocalization area of the exciton as a whole. In the latter case, the coherent summation of the dipole transitions of different regions occurs, and, therefore, the oscillator strength increases, i.e., Rashba effect arises. Rashba effect was observed for the impurity states in molecular crystals^{30,31} and for the biexcitons.³²⁻³⁴ For example, the ratio of the biexciton oscillator strength to the exciton oscillator strength in CuCl is equal to 2.5×10^3 (Ref. 35). The obtained values of the oscillator strength allow evaluation of the radiative lifetime of the exciton by the standard formula $\tau = \frac{3m_0c^3}{2\sqrt{\epsilon_0}\epsilon^2\omega^2F_{01}}$. For example, for the nanoparticle with the radius $R = 6$ nm, the lifetime is about 5×10^{-12} s for d

$= 10$ nm. In the considered case, the radiative bandwidth rises strongly with increasing d and, respectively, the radiative lifetime of the state becomes very small, for the nanoparticle mentioned above the lifetime decreases to 7×10^{-13} s for $d = 17$ nm. At the same time, the exciton binding energy decreases as the distance rises. For this reason in Fig. 6, we present the oscillator strength dependence on the distance for the range where the radiative bandwidth is less than the energy of the binding of the exciton to the nanoparticle. For larger distances, the radiative width of the exciton level becomes comparable with the binding energy of the exciton to the localization place, and, consequently, the approximations, used for the calculations, become incorrect because the radiative lifetime will affect the exciton too.

In the mixed states, both the oscillator strength and the damping are “borrowed” from the plasmon state to the exciton state. This leads to the broadening of the exciton band. One can introduce an imaginary addition to the surface-plasmon energy $\omega_l \rightarrow \omega_l - i\frac{\Gamma}{2}$ in order to evaluate the damping. Both the real and the imaginary parts of the energy $E \rightarrow E - i\frac{\hbar\Gamma}{2}$ (where Γ is the damping of the mixed level) are obtained from Eq. (9). The localization of the exciton level, considered in this paper, is meaningful if its separation δE_b from the exciton band exceeds the linewidth. Calculations show that the ratio of the imaginary and the real part of the energy decreases as the detuning from resonance grows. This is caused by the decrease in the relative contribution of the nanoparticle to the excited state. For the considered values of the resonance detuning, the broadening caused by the mixing is less than the shift of the levels. The damping $\hbar\Gamma_2$ and the binding energy of the exciton as a whole $\delta E_b = |E_b - E_{b0}|$ for the nanoparticle with the radius $R = 10$ nm and the center-to-center distance $d = 11$ nm are equal, respectively, $\hbar\Gamma_2 = 4.9$ meV and $\delta E_b = 6.3$ meV for the detuning $\Delta E = 0.1$ eV, $\hbar\Gamma_2 = 1.3$ meV, and $\delta E_b = 2.9$ meV for $\Delta E = 0.2$ eV, $\hbar\Gamma_2 = 0.4$ meV, and $\delta E_b = 1.7$ meV for $\Delta E = 0.3$ eV. So, in some region of the values of the detuning, the splitting-off of the exciton level from the exciton band is larger than the broadening of the level.

One can obtain similar estimates for the effect of the damping of the surface plasmon on the detuning of the phase relationships between the wave functions of the exciton and the plasmon, the superposition of which determines the wave function of the discrete level, splitted from the exciton band. The localization of the excitation at the moment of time $t = 0$ in one of the subsystems (for example, in the localized exciton state Ψ_m^{exc}) leads to the time oscillations of the wave function of the system describing the transitions of the excitation between the quantum well and the nanoparticle. The frequency of the oscillations is equal to $\omega_d = \frac{\sqrt{\Delta E^2 + |2V_m|^2}}{\hbar}$, where V_m is the matrix element of the transition and ΔE is the detuning. The time of the damping of the oscillations t_D is connected with the linewidth of the surface plasmon and has the order of value $t_D \approx \frac{2}{\Gamma}$. So, if $\omega_D t_D > 1$, the excitation has time to pass from one subsystem to the other several times before damping out. For the numerous example, considered above, one can obtain $\omega_D t_D = 2$ for $\Delta E = 0.1$ eV, $\omega_D t_D = 4$ for $\Delta E = 0.2$ eV, and $\omega_D t_D = 6$ for $\Delta E = 0.3$ eV. Here the splitting-off of the exciton level from the exciton

band is still larger than the level broadening caused by the mixing.

We consider the system where the bottom of the exciton band is situated lower than the surface-plasmon level. For the opposite order of the levels, the localized exciton states do not appear because in that case, the resonant interaction shifts the exciton levels deeply into the band whereas in the studied case, it pushes the levels out. As the result, the Forster energy transfer from the quantum well occurs leading to the damping of exciton excitations.

Let us analyze the case when the metallic nanoparticle is situated outside of the semiconductor. For such system, the interaction of the nanoparticle with the quantum well changes in comparison with the case considered above in the paper. Also, the shift of the frequencies of the surface-plasmon oscillations takes place. Let the spherical nanoparticle with the dielectric constant $\varepsilon^{(1)}(\omega)$ be placed in the medium with the dielectric constant ε at the distance d_1 between the particle and the semiconductor with the dielectric constant $\varepsilon_\infty^{(2)}$. The spectrum and the electric fields of the oscillations arising in the system is determined from the solutions of the Laplace equation for the electric potential inside the nanoparticle, in the semiconductor, and outside of the semiconductor, as well as from the boundary conditions for the Maxwell equations. It is easy to find the solutions, if we will take into account only the harmonics $\frac{(Y_{1,1} \pm Y_{1,-1})}{\sqrt{2}}$, $Y_{1,0}$ in the spherical-harmonic expansion of the solution of the Laplace equation. As the result, one can obtain, that the electric fields generated by the oscillating dipole \mathbf{P} parallel to the quantum-well interface in the structure with the nanoparticle outside the semiconductor and in the uniform medium differ by the multiplier $\lambda_1 = \frac{2\varepsilon}{(\varepsilon + \varepsilon_\infty^{(2)})}$. This means that the matrix element of the moment operator \mathbf{P}_{met} in the Eq. (19) has to be multiplied by λ_1 . The frequency of the surface plasmon for the nanoparticle with the radius R at the distance d_1 from the semiconductor can be written as

$$\varepsilon^{(1)}(\omega) = -\varepsilon \frac{2 + \lambda_2 \left(\frac{R}{2d_1}\right)^3}{1 - \lambda_2 \left(\frac{R}{2d_1}\right)^3}, \quad (22)$$

where $\lambda_2 = \frac{(\varepsilon_\infty^{(2)} - \varepsilon)}{(\varepsilon_\infty^{(2)} + \varepsilon)}$.

Let us choose for the frequency dependence of the dielectric permittivity in the Drude model $\varepsilon^{(1)}(\omega) = \varepsilon^{(1)} - \frac{\omega_p^2}{\omega^2}$ and suppose that the values $\varepsilon^{(1)}$, $\varepsilon_\infty^{(2)}$, and ε do not depend on the frequency. Then one can obtain for the frequency of the surface Plasmon,

$$\omega = \left[\frac{\omega_p}{\varepsilon^{(1)} + \varepsilon \frac{2 + \lambda_2 \left(\frac{R}{2d_1}\right)^3}{1 - \lambda_2 \left(\frac{R}{2d_1}\right)^3}} \right]^{1/2}. \quad (23)$$

For $\varepsilon^{(1)}=1$ and for the nanoparticle in the vacuum ($\varepsilon=1$), one can obtain the well-known equation $\omega = \frac{\omega_p}{\sqrt{3}}$ whereas for

the nanoparticle in the uniform medium ($\lambda_2=0$), one can obtain the equation used in the paper. In the general case, the position of the nanoparticle outside the semiconductor leads to the decrease in the plasmon frequency but this decrease is not substantial. Even for the nanoparticle placed on the surface $R=d_1$, the relative shift is about 10% (relative to the plasmon frequency in the absence of the semiconductor), and the energy of the plasmon is high. So, the system, considered in the paper, with the nanoparticle placed inside the semiconductor is preferable for the study of the resonance effects of the exciton-plasmon interaction because in this case, the plasmon frequency is close to the exciton frequencies of many semiconductors. Besides, due to the condition $\lambda_1 < 1$, the position of the particle outside the crystal leads to the decrease in the electric field created by the oscillations, which results in the decrease in the effects of the resonance interaction by about one order of magnitude.

Summarizing, the localized exciton states would manifest themselves in the optical spectra of the studied hybrid structures as an additional line with extremely short lifetime situated under the exciton band.

IV. CONCLUSIONS

The lowest-energy levels of the excited states of the system consisting of a metallic nanoparticle and a quantum well in a semiconductor are calculated variationally taking into account the exciton-plasmon interaction in the dipole-dipole approximation. The dependence of the energy of the localized excitons on the distance between the nanoparticle and the quantum well and on the nanoparticle radius was obtained. It is shown that the presence of the metallic nanoparticle in the vicinity of the quantum well can qualitatively alter the exciton states and the optical properties of the structure. In particular, (1) if the exciton energy level is lower than the energy of the plasmon, the interaction of the exciton with the surface plasmon can cause localization of the exciton near the nanoparticle; (2) strong resonance interaction between the exciton and the plasmon states changes the character of the propagation of the exciton as a whole from the Wannier-type to the Frenkel-type type; (3) the giant increase in the oscillator strength of the localized exciton transition occurs near the resonance between the exciton and the plasmon levels. Calculations show that in the case of the exciton-plasmon resonance, the oscillator strength for the transition of the localized exciton can exceed by several orders of magnitude greater than the oscillator strength of the free exciton in the quantum well. This effect is caused by (a) the ‘‘borrowing’’ from the dipole plasmon transition intensity by the intensity of the exciton transition and (b) the large radius of the exciton state localized near the nanoparticle and, consequently, the enhancement of the oscillator strength due to the Rashba effect.

ACKNOWLEDGMENTS

This work was supported partly by the Swedish Institute (SI) through the Visby program. The authors thank Bo Lied-

berg, Alexander Onipko, and Igor Goliney for helpful discussion.

APPENDIX: THE QUANTIZATION OF THE STATES OF THE PLASMON OSCILLATIONS OF THE METALLIC NANOPARTICLE IN THE SEMICONDUCTOR MATRIX

The Lagrange function of the nanoparticle in the semiconductor matrix can be written as

$$L = \frac{1}{8\pi} \int_{r < R} d\mathbf{r} \left[\varepsilon^{(1)} (\nabla \varphi)^2 - 8\pi (\nabla \varphi \cdot \mathbf{P}) + \frac{(4\pi)^2}{\omega_p^2} \dot{P}^2 \right] + \frac{1}{8\pi} \int_{r > R} d\mathbf{r} \left[\varepsilon_\infty^{(2)} (\nabla \varphi)^2 - 8\pi (\nabla \varphi \cdot \mathbf{P}) + \frac{4\pi}{\gamma} \dot{P}^2 - \frac{4\pi}{\gamma} \omega_0^2 P^2 \right], \quad (\text{A1})$$

where $\mathbf{P} = e \sum_i \mathbf{r}_i$ (\mathbf{r}_i is the coordinate of the i th electron) is the dipole moment of the unit volume, $\varphi(\mathbf{r})$ is the electrostatic potential, $\gamma = \omega_0 \omega_{\text{LT}} \varepsilon_\infty^{(2)} / 2\pi$, $\varepsilon^{(1)}$ is the frequency independent part of the dielectric constant of the metal nanoparticle, $\varepsilon_\infty^{(2)}$ is the high-frequency dielectric constant of the semiconductor, and ω_{LT} is the longitudinal-transverse splitting of the exciton state. It can be shown that the chosen Lagrange function [Eq. (A1)] leads via application of the the variational principle to the Maxwell's equations, the dynamic equation for polarization as well as to the corresponding boundary conditions for the system. We will find the periodical in time solutions of the obtained equations for the spherical nanoparticle in the form

$$\varphi(\mathbf{r}) = \sum_{lm} a_{lm} \begin{cases} \varphi_{lm}^{(1)}(\mathbf{r}), & r \leq R \\ \varphi_{lm}^{(2)}(\mathbf{r}), & r > R, \end{cases} \quad (\text{A2})$$

$$\mathbf{P}(\mathbf{r}) = \sum_{lm} a_{lm} \begin{cases} \mathbf{P}_{lm}^{(1)}(\mathbf{r}), & r \leq R \\ \mathbf{P}_{lm}^{(2)}(\mathbf{r}), & r > R, \end{cases} \quad (\text{A3})$$

where

$$\mathbf{P}_{lm}^{(1)}(\mathbf{r}) = \frac{\omega_p^2}{4\pi\omega_l^2} \nabla \varphi_{lm}^{(1)}(\mathbf{r}) \quad (\text{A4})$$

and

$$\mathbf{P}_{lm}^{(2)}(\mathbf{r}) = \frac{\gamma}{\omega_l^2 - \omega_{0b}^2} \nabla \varphi_{lm}^{(2)}(\mathbf{r}), \quad (\text{A5})$$

ω_{0b} is the position of the exciton band in the energy spectrum of the semiconductor, ω_p is the plasma frequency. Here and further on the indexes 1 and 2 denote physical values related to the metallic particle and to the semiconductor crystal, respectively, ω_l are the eigenfrequencies of the system. We have used the solutions of the Laplace equation for the metallic sphere in the dielectric matrix $\varphi_{lm}^{(1,2)}(\mathbf{r})$ as the eigenfunctions. As known,⁴ the eigenfrequencies for the spherical particle depend only on the quantum number l ,

$$\omega_l = \sqrt{\frac{l\omega_p^2}{\varepsilon^{(1)}l + \varepsilon_\infty^{(2)}(l+1)}}.$$

Using the Lagrange function of Eq. (A1) and the standard secondary quantization procedure,³⁶ one can obtain the Hamiltonian of the system in the representation of secondary quantization $H = \sum_l \hbar \omega_l A_{lm}^+ A_{lm}$, where A_{lm}^+ and A_{lm} are the creation and the annihilation operators of the plasmon with the frequency ω_l , $\hat{a}_{lm} = \frac{1}{2} \left(\frac{\hbar}{B_{lm}\omega_l} \right)^{1/2} (\hat{A}_{l,m} + (-1)^m \hat{A}_{l,-m}^+)$, where

$$B_{lm} = \frac{\omega_p^2}{8\pi\omega_l^4} \oint_S \varphi_{lm}^{(1)}(\mathbf{r}) \nabla \varphi_{lm}^{(1)*}(\mathbf{r}) d\mathbf{S} - \frac{\gamma}{2(\omega_l^2 - \omega_{0b}^2)^2} \oint_S \varphi_{lm}^{(2)}(\mathbf{r}) \nabla \varphi_{lm}^{(2)*}(\mathbf{r}) d\mathbf{S}. \quad (\text{A6})$$

In this way, we can obtain the operator of the electrostatic potential of the system outside of the metallic nanoparticle,

$$\hat{\varphi} = \sum_{lm} \frac{1}{2} \sqrt{\frac{\hbar}{B_{lm}\omega_l}} [\hat{A}_{lm} \varphi_{lm}^{(2)}(\mathbf{r}) + \hat{A}_{lm}^+ \varphi_{lm}^{(2)*}(\mathbf{r})] \quad (\text{A7})$$

and the operator of the dipole moment of the sphere,

$$\hat{P} = \sum_i \frac{\omega_p^2}{8\pi\omega_l^2} \sqrt{\frac{\hbar}{B_{lm}\omega_l}} \times \left[\hat{A}_{lm} \int \nabla \varphi_{lm}^{(1)}(\mathbf{r}) d\mathbf{r} + \hat{A}_{lm}^+ \int \nabla \varphi_{lm}^{(1)*}(\mathbf{r}) d\mathbf{r} \right]. \quad (\text{A8})$$

The integration is performed over the volume of the metallic sphere in Eq. (A8) and over the surface of the metallic sphere in Eq. (A6).

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