Resonant Light Absorption by Semiconductor Quantum Dots

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The cross section of light absorption by semiconductor quantum dots is calculated in the resonance with Γ6 × Γ7 excitons in cubic crystals Td. The interference of stimulating and induced electric and magnetic fields is taken into account. The cross section is proportional to the exciton nonradiative damping γ.

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I. INTRODUCTION

When the size-quantized semiconductor objects (quantum wells, quantum wires, quantum dots) are irradiated by light, elastic light scattering and absorption intensify resonantly if the light frequency ωl equals to the exciton frequency ω0. The resonant peak width is determined by the exciton broadening Γ, which consists of nonradiative and radiative broadening, i.e., Γ = γ + γr. Therefore, measurements of light scattering and absorption may serve as a convenient method of research of exciton properties in the mentioned above objects. An important role of the radiative damping γr was proved for the first time in1,2,3. Light reflection by some structures, consisting of quantum wells, wires and dots was considered in4. In the present work, a semiclassical method of retarded potentials is applied for calculation of the light absorption cross section of a quantum dot. The method allows to avoid using of boundary conditions for electric and magnetic fields. That is especially important in a case of quantum dots of arbitrary forms.

The method of the retarded potentials is described and applied to the light scattering by a semiconductor quantum dot5. The calculations are performed for the resonance of stimulating light with the exciton Γ6 × Γ7 in cubic crystals Td. An exciton is formed by an electron from the twice degenerated conduction band Γ6 and by a hole from the twice degenerated valence band Γ7, chipped off by the spin-orbital interaction. The same excitons are considered in6. The resonant light absorption is calculated below.

II. INDUCED ELECTRIC AND MAGNETIC FIELDS

Electron and hole wave functions have a structure (we use designations of5)

\[ \Psi_{e1} = i S \uparrow, \quad \Psi_{e2} = i S \downarrow, \]  \tag{1}

\[ \Psi_{h1} = \frac{1}{\sqrt{3}} (X - iY) \uparrow - \frac{1}{\sqrt{3}} Z \downarrow, \]
\[ \Psi_{h2} = \frac{1}{\sqrt{3}} (X + iY) \uparrow + \frac{1}{\sqrt{3}} Z \downarrow. \]  \tag{2}

Combining (1) and (2) in pairs, we obtain a four times degenerated excitonic state, for which interband matrix elements of the quasi-momentum operator are equal

\[ p_{cv1} = \frac{p_{cv}}{\sqrt{3}} (e_x - ie_y), \]
\[ p_{cv2} = \frac{p_{cv}}{\sqrt{3}} (e_x + ie_y), \]
\[ p_{cv3} = \frac{p_{cv}}{\sqrt{3}} e_z, \]
\[ p_{cv4} = -\frac{p_{cv}}{\sqrt{3}} e_z, \]  \tag{3}

where

\[ p_{cv} = i \langle S | \hat{p}_z | X \rangle, \]  \tag{4}

\[ e_x, e_y \text{ and } e_z \text{ are the unit vectors aligned parallel to the crystallographic axes.} \]

An exciton Γ6 × Γ7 is the most simple object, unlike the excitons containing light or heavy holes. We obtain that all the measurable values do not depend on the direction of the crystallographic axes vectors, i.e., in the case of Γ6 × Γ7 excitons, a crystal plays a role of an isotropic medium.

When the light irradiation is monochromatic, the stimulating electric and magnetic fields can be written as

\[ \mathbf{E}_6^\pm (r, t) = E_0 \mathbf{e}_x^\pm e^{i(k_r x - \omega_l t)} + c.c., \]
\[ \mathbf{H}_7^\pm (r, t) = E_0 \nu (e_x \pm e_y^\pm) e^{i(k_r x - i\omega_l t)} + c.c.. \]  \tag{5}

The axis z is aligned parallel to the light wave vector \( \mathbf{k}_l \), whose magnitude equals \( k_l = \omega_l/\nu/c \), \( \nu \) is the light refraction coefficient, identical inside and outside of the quantum dot. We use a circular polarization

\[ e^\pm_\ell = \frac{1}{\sqrt{2}} (e_x \pm ie_y). \]  \tag{6}
Induced electric and magnetic fields on the large distances from the quantum dot, which determine light scattering and absorption, are calculated in \[S.\]

The values

\[P(k) = \int d^3r e^{-ikr} F(r)\]  \hspace{1cm} (7)

are essential in the theory \((F(r)\) is the real exciton wave function at \(r_e = r_h = r, \ r_e(r_h)\) is the electron (hole) radius - vector). The "envelope" wave function \(F(r)\) is calculated in the effective mass approximation.

The electric and magnetic fields are calculated precisely, when \(P(k)\) depends on the magnitude \(k\) only, i. e.,

\[P(k) = P(k).\]  \hspace{1cm} (8)

The condition (8) is carried out, if the function \(F(r)\) is spherically symmetric or if the quantum dot sizes \(R\) are much less than the stimulating light wave length, and

\[P(k) \simeq P(0).\]  \hspace{1cm} (9)

For example, condition (8) is carried out in the case of a spherical quantum dot, limited by an infinitely high rectangular potential barrier. Then the "envelope" function

\[F(r) = F(r) = \frac{1}{2\pi R} \sin^2\left(\frac{\pi r}{R}\right) \Theta(R - r)\]

corresponds to the lowest exciton energy level \(\hbar\omega_0\), and

\[P(k) = \frac{2}{kR} \int_0^\pi dx \sin\frac{kRx}{\pi} \sin\frac{x^2}{x}, \ \ P(0) = 1.\]  \hspace{1cm} (10)

Under exact calculation of the induced fields we mean the account of all the orders on the light - electron interaction (containing the parameter \(e^2/\hbar c\), what corresponds to the account of all the processes of light reabsorption and reradiation. In \[S.\], the results

\[\Delta E_{r \to \infty}(r, t) = -\frac{3}{4} \frac{E_0}{kR} \frac{\gamma_r}{kR} \left[|e_r e_s^+|^2 + (e_r e_s^+) e_s^+ \right] \times \frac{e^{i(kR - \omega t)}}{\omega - \omega_0 + i\Gamma/2} + c.c.,\]  \hspace{1cm} (11)

\[\Delta H_{r \to \infty}(r, t) = \frac{3i\nu}{4} \frac{E_0}{kR} \frac{\gamma_r}{kR} \left[|e_r e_s^+|^2 - (e_r e_s^+) e_s^+ \right] \times \frac{e^{i(kR - \omega t)}}{\omega - \omega_0 + i\Gamma/2} + c.c.,\]  \hspace{1cm} (12)

are obtained, \(\omega_0 = \omega_0 + \Delta \omega_0\) is the exciton energy renormalized by the electron-light interaction, \(e_s^+\) is the circular polarization vector, and

\[\gamma_r = \frac{2\nu e^2}{9\hbar c} \left( \frac{p_{cv}}{m_0 c} \right)^2 \omega_0^2 |P(k)|^2,\]  \hspace{1cm} (13)

\(\hbar \omega_g\) is the band gap.

III. THE POINTING VECTOR. THE LIGHT SCATTERING CROSS SECTION

On the large distances from the quantum dot, the Pointing vector is equal

\[S_{r \to \infty} = S_0 + S_{\text{inter}} + S_{\text{scat}},\]  \hspace{1cm} (14)

where

\[S_0 = \frac{c}{4\pi} E_0 \times H_0 = \left( \frac{\epsilon \nu}{2\pi} \right) E_0^2 e_z,\]  \hspace{1cm} (15)

\[S_{\text{inter}} = \frac{c}{4\pi} (E_0 \times \Delta H + \Delta E \times H_0),\]  \hspace{1cm} (16)

\[S_{\text{scat}} = \frac{c}{4\pi} \Delta E \times \Delta H.\]  \hspace{1cm} (17)

With the help of (11) and (12), we obtain

\[S_{\text{scat}} = \frac{9\pi}{4} S_0 \frac{\gamma_r}{kR} \frac{r}{(k_\ell R)^2} \left( \frac{\omega - \omega_0}{\omega - \omega_0} \right)^2 + \Gamma^2/4,\]  \hspace{1cm} (18)

and the magnitude of the total flux of scattered light equals

\[\Pi_{\text{scat}} = \frac{3\pi}{2} S_0 \frac{\gamma_r}{k_\ell} \left( \frac{\omega - \omega_0}{\omega - \omega_0} + \Gamma^2/4.\right]  \hspace{1cm} (19)

In our calculations, we used the ratios

\[|e_r^+ e_s^-|^2 = |e_r^+ e_s^-|^2 = \frac{1}{4}(1 + \cos \Theta),\]

\[|e_r^+ e_s^-|^2 = |e_r^+ e_s^-|^2 = \frac{1}{4}(1 - \cos \Theta),\]  \hspace{1cm} (20)

where \(\Theta\) is the scattering angle, i. e., the angle between the vectors \(k_\ell\) and \(r\). Then, the integration on angles \(\Theta\) is executed. Dividing \(\Pi_{\text{scat}}\) on the flux density \(S_0\) of stimulating light energy and using the ratio \(k_\ell = 2\pi/\lambda_\ell\), we obtain the total scattering cross section

\[\sigma_{\text{scat}} = \frac{3}{2\pi} \frac{\lambda_\ell^2}{(\omega - \omega_0)^2 + \Gamma^2/4.\]  \hspace{1cm} (21)

It follows from (21) that under condition \(\gamma \ll \gamma_r\) in the resonance the total scattering cross section equals \((3/2\pi)\lambda_\ell^2\), where \(\lambda_\ell\) is the light wave length. Otherwise, at \(\gamma \gg \gamma_r\) the cross section in the resonance decreases in \((\gamma/\gamma_r)^2\) times. The damping \(\gamma_r\) is determined in (13), and under condition \(R \ll \lambda_\ell\), the approximation \(P(k) \simeq P(0)\) is applicable.

IV. A ROLE OF THE EXCITONIC NONRADIATIVE DAMPING IN LIGHT ABSORPTION

Results for cross sections of light scattering by a quantum dot are obtained with the help of the quasi-classical
method and coincide with results of the quantum perturbation theory in the lowest approximation on the electron-light interaction. However, the semiclassical method, consisting of the calculation of electric and magnetic fields, allows to determine also the light absorption section by quantum dots. The light absorption is caused by nonradiative damping $\gamma$ of excitons and in frameworks of our problem is equal 0 at $\gamma = 0$. Such result was obtained for a quantum dot in the case of monochromatic irradiation\textsuperscript{12}. In the case of pulse irradiation at $\gamma = 0$ integral absorption is equal to zero\textsuperscript{9,11,12}. The reason is that the energy dissipation spent by light on the exciton creation is absent at $\gamma = 0$, and the energy comes back at the exciton annihilation. The process of reabsorption and reradiation proceeds infinitely.

At calculation of the light absorption coefficient of a quantum well, it was found out that it is necessary to take into account the interference of stimulating and induced electromagnetic fields (see, for example\textsuperscript{10}).

Let us show that the last statement is true for the quantum dot also.

**V. THE INTERFERENCE CONTRIBUTION INTO THE ENERGY FLUX**

Let us calculate the interference contribution into the Pointing vector. Substituting (5), (11) and (12) in (16), we obtain

\[
S_{\text{Interf}} = S_z + S_\perp,
\]

\[
S_z = -\frac{3}{4} \frac{\gamma r}{k r} S_0 e_z |e_+^z e_-^z|^2 \times \left( \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} + c.c. \right),
\]

\[
S_\perp^\pm = \frac{3}{4} \frac{\gamma r}{k r} S_0 \times \left( e_+^z (e_+^z e_-^z) (e_+^z e_-^z) \frac{e^{i(kr - k r)}}{\omega t - \omega_0 - i/2} + c.c. \right).
\]

where the indexes $+(-)$ correspond to the right (left) polarization of the stimulating light.

Since expressions (22) and (23) correspond to the case $r \to \infty$, it is obvious that only the angles $\Theta \to 0$ can contribute into the constant energy flux because of the factor $e^{i(kr - k r)}$. However, in the RHS of (23) there is the factor $e_+^z e_-^z$, which equals 0 at $\Theta = 0$. Therefore, the constant energy flux on large distances from a quantum dot corresponds only to the vector $S_z$. Let us calculate the energy flux

\[
\Pi_z = \int ds S_z,
\]

passing through a surface, perpendicular to the direction $z$ of stimulating light in time unit on a very large distance $z$ behind the quantum dot. The surface element is equal $ds = \rho d\rho d\varphi$, and $\rho = z \tan \Theta$, $r = z / \cos \Theta$. Having executed integration on the angle $\varphi$, which gives a factor $2\pi$, using (20) and passing on from the variable $\rho$ to the variable $\Theta$, we obtain

\[
\Pi_z = -3 \pi \frac{\gamma r}{8} S_0 \times \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} \int_0^{\pi/2} d\Theta \times \sin \Theta (1 + \cos \Theta)^2 e^{i k z (1 - (1/\cos \Theta))} + c.c..
\]

(25)

Further, let us pass to the variable $t = \cos^{-1} \Theta - 1$ from the variable $\Theta$

\[
\Pi_z = -3 \pi \frac{\gamma r}{8} S_0 \times \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} \times \int_0^\infty dt \left( \frac{2 + t}{1 + t} \right)^2 e^{-i k z t} + c.c..
\]

(26)

At $z \to \infty$, having executed integration on $t$, we obtain

\[
\Pi_z = -3 \pi \frac{\gamma r}{2 k r} S_0 e_z \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} \times \left( \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} + c.c. \right).
\]

(27)

independent on $z$. In (27), the contributions depending to $t$ at $z \to \infty$ are omitted, as well as the contributions oscillating very rapidly on $z$.

Thus, we obtain that the interference contribution to the Pointing vector at $z \to \infty$ results into the constant energy flux directed oppositely to the axis $z$. It means that the energy flux of stimulating light decreases on the value $\Pi_z$.

From expressions (25) and (26), it is obvious that at $z \to \infty$ the basic contribution into the integral on $\Theta$ is brought in by the very small angles, when $e^{i(kr - k r)}$ approaches to the unit.

The "transverse" component $S_\perp$ of the interference contribution to the Pointing vector may be written down as

\[
S_\perp^\pm = -3 \pi \frac{\gamma r}{8 \sqrt{2}} S_0 \frac{e^{i(kr - k r)}}{\omega t - \omega_0 - i/2} \times \left( \frac{e^{i(kr - k r)}}{\omega t - \omega_0 + i/2} + c.c. \right).
\]

(28)

In comparison with similar expression (22) for $S_z$, the RHS of (28) contains an additional factor $\sin \Theta$, which corresponds to the factor $e_z e^+_z = (\pm i / \sqrt{2}) \sin \Theta$ from the RHS of (23). Due to this additional factor, the contribution $S_\perp^\pm$ approaches to zero in the area of very small angles $\Theta$, where $e^{i(kr - k r)}$ approaches to the unit at $z \to \infty$. A direct calculation shows that the flux approaches to zero at $z \to \infty$. Therefore, it may be omitted.

**VI. THE LIGHT ABSORPTION CROSS SECTION**

Since $\Gamma = \gamma r + \gamma$, the flux (27) consists of two parts :

\[
\Pi_z = -e_z \Pi_{scat} - e_z \Pi_{abs},
\]

(29)
the total flux of the scattered energy, and
\[ - \frac{\Pi_{\text{abs}}}{\gamma} \] time unit. Having divided (30) on the density
\[ \Pi_{\text{abs}} = \sigma_0 \frac{3}{2} \frac{\gamma \gamma}{\pi r^2} \left( \omega - \omega_0 \right)^2 + (\Gamma/2)^2, \] of the electron-light interaction, and the absorption section is of the first order. In the resonance, at \( \gamma < \gamma_r \) the ratio of the absorption section to the scattering section equals \( \gamma/\gamma_r < 1 \). Otherwise, at \( \gamma \gg \gamma_r \), the same ratio equals \( \gamma/\gamma_r > 1 \). The largest result for the absorption section turns out at comparable values \( \gamma \) and \( \gamma_r \). Thus, at \( \gamma = \gamma_r \) in the resonance,
\[ \sigma_{\text{abs}}^\text{res} = \frac{3}{\pi} \frac{\lambda^2}{\gamma}. \] (32)

In Fig. 1, the dependencies of \( \sigma_{\text{abs}}^\text{res} \) and \( \sigma_{\text{abs}}^\text{res} \) on the parameter \( \gamma_r/\gamma \) are represented.

Thus, the semiclassical method of the retarded potentials has allowed to calculate induced electric and magnetic fields arising at a light irradiation of a quantum dot if the stimulating light frequency is in the resonance with the exciton frequency. The method has allowed to avoid using of boundary conditions for fields. That has considerably facilitated the solution of the problem. On the large distances from the quantum dot, fields are calculated precisely, i.e., without an expansion in series on the electron-light interaction.

For excitons \( \Gamma_6 \times \Gamma_7 \) in cubic crystals of \( T_d \) class, the expressions for light scattering and absorption sections are obtained. It is shown that the account of the interference contributions into the Pointing vector is necessary for calculation of the light absorption.

VII. CONCLUSION

Thus, the semiconductor method of the retarded potentials has allowed to calculate induced electric and magnetic fields arising at a light irradiation of a quantum dot if the stimulating light frequency is in the resonance with the exciton frequency. The method has allowed to avoid using of boundary conditions for fields. That has considerably facilitated the solution of the problem. On the large distances from the quantum dot, fields are calculated precisely, i.e., without an expansion in series on the electron-light interaction.

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