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Blue-shift and broadening of optical spectra from response nonlocality in quantum wells

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ABSTRACT

We show the nonlocal linear optical response of electrons to light field can induce the radiative shift and broadening of optical spectra in square quantum wells (SQW's), and they can be controlled by an applied static electric field. In addition, it is also analyzed how the nonlocal effects will depend on the potential of quantum wells. This work may result in a substantial impact on the growth of the nonlocal materials.

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Nonlocal optical responses in nanostructures have recently attracted extensive interest [1–11]. The so-called nonlocal optical response arises when the polarization at a spatial point is induced by the applied optical fields not only at the same point, but also at other positions within the extent of the relevant electronic (or excitonic) wave-function. The nonlocality can lead to the blue-shift and broadening of optical spectra [1–7]. It is more evident in nanostructures. One may ask if the nonlocal effects can be controlled by a certain method and how the nonlocal effects will depend on the potential of the well. In this paper we presented a systematic study to state these questions.

We assume a p-polarized, monochromatic plane-wave electromagnetic field is incident on a square quantum well (SQW) with an applied static electric field along the z direction. One of the surfaces of the SQW is located at xy plane with z=0, and the other at z=L. Due to the system with the translational invariance along the xy direction, one can assume the electric field $\mathbf{E}(\mathbf{r},\omega)$ and the polarizability $\mathbf{P}(\mathbf{r},\omega)$ have the forms: $\mathbf{E}(\mathbf{r},\omega) = \tilde{\mathbf{E}}(z;\omega) \exp(i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel})$ and $\mathbf{P}(\mathbf{r},\omega) = \tilde{\mathbf{P}}(z;\omega) \exp(i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel})$, where $\mathbf{k}_{\parallel} = (k_x, k_y, 0)$ stands for the wave-vector in the xy plane of the incident electromagnetic wave. From Maxwell's equations, one can derive the integral-differential equation for the electric field $\mathbf{E}(\mathbf{r},\omega)$ in the case

of linear optical response, viz.,

$$\stackrel{\leftrightarrow}{\Lambda}(\omega) \cdot \tilde{\mathbf{E}}(z;\omega) = -\frac{\omega^2}{c^2} \int_0^L \mathrm{d}z' \stackrel{\leftrightarrow}{\chi}(z,z';\omega) \cdot \tilde{\mathbf{E}}(z';\omega), \tag{1}$$

where the tensorial operator $\overset{\leftrightarrow}{A}(\omega)=\overset{\leftrightarrow}{I}[\omega^2/c^2\epsilon^B-k_\parallel^2+\partial^2/\partial z^2]$ —

 $(i\mathbf{k}_{\parallel}+\mathbf{e}_z\partial/\partial z)(i\mathbf{k}_{\parallel}+\mathbf{e}_z\partial/\partial z).$ $\stackrel{\leftrightarrow}{\chi}(z,z';\omega)$ is the nonlocal linear susceptibility tensor. By the density-matrix method as Ref. [4], its nonzero elements for the two-level QW model can be obtained in the low-temperature $(T\to 0)$ and long-wavelength $(\mathbf{k}_{\parallel}\to 0)$ limits as follows:

$$\chi_{XX}(z,z';\omega) = \frac{-e^2}{2\pi\epsilon_0 \,\hbar^2 \,\omega^2} \frac{(\varepsilon_F - \varepsilon_1)^2}{\hbar\omega + \varepsilon_1 - \varepsilon_2 + i\hbar\Gamma_0} \Psi(z)\Psi(z'), \quad (2)$$

$$\chi_{zz}(z,z';\omega) = \frac{e^2}{4\pi\epsilon_0 m^* \omega^2} \frac{(\varepsilon_1 - \varepsilon_F)}{\hbar\omega + \varepsilon_1 - \varepsilon_2 + i\hbar\Gamma_0} \zeta(z)\zeta(z'), \quad (3)$$

with $\Psi(z) = \varphi_1(z)\varphi_2(z)$ and $\zeta(z) = \varphi_1(z)d\varphi_2(z)/dz - \varphi_2(z)d\varphi_1(z)/dz$, where $\varphi_n(z)$ and $\varepsilon_n(n=1,2)$ are the envelope wave functions and the transverse energies of the nth subband in the two-level QW, respectively. For the SQW with an applied static electric field F, $\varphi_n(z) = C_{1n}Ai(\eta_n(z)) + C_{2n}Bi(\eta_n(z))$, with $\eta_n(z) = -(2m^*/(e\hbar F)^2)^{1/3}(\varepsilon_n - |e|Fz)$, where C_{1n} and C_{2n} are the normalization constants. ε_n are determined by the boundary condition $\varphi_n(0) = \varphi_n(L) = 0$. e is an electron charge. m^* is the effective mass of an electron in conduction bands. ϵ^B is the relative

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dielectric constant for the assumed local, isotropic background medium. ϵ_0 is the vacuum permittivity. Γ_0 is the non-radiative relaxation rate. $\varepsilon_F = \varepsilon_1 + L\pi \, \hbar^2 \, N_d/m^*$ is the Fermi energy of the SQW system, where N_d is the donor concentration, and L is the width of the SQW [4].

By the Green's function method, one can solve the components of the electric field from Eq. (1) as follows [4,6]

$$\tilde{E}_{x}(z;\omega) = \tilde{E}_{0x}(z;\omega) + \alpha N_{x} H_{xx}(z;\omega) + \beta N_{z} H_{xz}(z;\omega), \tag{4}$$

$$\tilde{E}_{z}(z;\omega) = \tilde{E}_{0z}(z;\omega) + \alpha N_{x} H_{zx}(z;\omega) + \beta N_{z} H_{zz}(z;\omega), \tag{5}$$

with

$$N_{x} = \int_{0}^{L} \Psi(z) \tilde{E}_{x}(z; \omega) dz, \tag{6}$$

$$N_z = \int_0^L \zeta(z)\tilde{E}_z(z;\omega)\mathrm{d}z,\tag{7}$$

$$H_{XX}(z;\omega) = \int_0^L G_{XX}(z,z';\omega)\Psi(z')dz', \tag{8}$$

$$H_{XZ}(z;\omega) = \int_0^L G_{XZ}(z,z';\omega)\zeta(z')dz', \qquad (9)$$

$$H_{\rm ZX}(z;\omega) = \int_0^L G_{\rm ZX}(z,z';\omega)\Psi(z')dz', \tag{10}$$

$$H_{zz}(z;\omega) = \int_0^L G_{zz}(z,z';\omega)\zeta(z')dz', \qquad (11)$$

$$\alpha = \frac{\mu_0 e^2}{2\pi \hbar^2} \frac{(\varepsilon_F - \varepsilon_1)^2}{\hbar \omega + \varepsilon_1 - \varepsilon_2 + i\hbar \Gamma_0},\tag{12}$$

$$\beta = \frac{\mu_0 e^2}{4\pi m^*} \frac{\varepsilon_F - \varepsilon_1}{\hbar \omega + \varepsilon_1 - \varepsilon_2 + i\hbar \Gamma_0},\tag{13}$$

$$\overset{\leftrightarrow}{G}(z,z';\omega) = \frac{k_{\parallel}}{2i\epsilon^{B}} \left(\frac{c}{\omega}\right)^{2} e^{ik_{\perp}|z-z'|} \begin{pmatrix} k_{\perp}/k_{\parallel} & \operatorname{sgn}(z'-z) \\ \operatorname{sgn}(z'-z) & k_{\parallel}/k_{\perp} \end{pmatrix} + \frac{1}{\epsilon^{B}} \left(\frac{c}{\omega}\right)^{2} \delta(z'-z) \mathbf{e}_{z} \mathbf{e}_{z}, \tag{14}$$

where $\tilde{\mathbf{E}}_0(z;\omega) = \mathbf{E}_0 \exp(\mathrm{i}k_\perp z)$ is the incident field, $k_\perp = (\epsilon^B \omega^2/c^2 - k_\parallel^2)^{1/2}$ is the wave-vector along z direction. G_{xx} , G_{xz} , G_{zx} and G_{zz} are four elements of the tensorial Green's function G_{zz} denotes the unit vector in the z-direction.

Multiplying both sides of Eqs. (4) and (5) by $\Psi(z)$ and $\zeta(z)$, respectively, and integrating the two equations over z across the QW, one can obtain

$$N_{x} = \frac{\tilde{S}_{x}(1 - \beta M_{zz}) + \beta \tilde{S}_{z} M_{xz}}{(1 - \alpha M_{xx})(1 - \beta M_{zz}) - \alpha \beta M_{xz} M_{zx}},$$
(15)

$$N_{z} = \frac{\tilde{S}_{z}(1 - \alpha M_{xx}) + \alpha \tilde{S}_{x} M_{zx}}{(1 - \alpha M_{xx})(1 - \beta M_{zz}) - \alpha \beta M_{xz} M_{zx}},$$
(16)

where $M_{xx} = \int_0^L H_{xx}(z;\omega)\Psi(z)dz$, $M_{xz} = -M_{zx} = \int_0^L H_{xz}(z;\omega)\Psi(z)dz$, $M_{zz} = \int_0^L H_{zz}(z;\omega)\zeta(z)dz$, $\tilde{S}_x = \int_0^L \tilde{E}_{0x}(z;\omega)\Psi(z)dz$, $\tilde{S}_z = \int_0^L \tilde{E}_{0z}(z;\omega)\zeta(z)dz$. For the following analyses, we write Eqs. (15) and (16) into the following matrix form by the rotating wave approximation [1,6]

$$\overset{\leftrightarrow}{\mathbf{D}} \cdot \tilde{\mathbf{N}} = \tilde{\mathbf{S}},\tag{17}$$

where $\tilde{\mathbf{D}}$ is a 2 × 2 matrix with elements $D_{xx} = \hbar\omega + \varepsilon_1 - \varepsilon_2 + i\hbar\Gamma_0 + hM_{xx}, D_{zz} = \hbar\omega + \varepsilon_1 - \varepsilon_2 + i\hbar\Gamma_0 + tM_{zz}, D_{xz} = D_{zx} = 0$. Vector $\tilde{\mathbf{S}} = (\tilde{S}_x, \tilde{S}_z)$ and $\tilde{\mathbf{N}} = g(N_x, N_z)$. Here $g = 1/(\hbar\omega + \varepsilon_1 - \varepsilon_2 + i\hbar\Gamma_0)$, $h = -\mu_0 e^2 (\varepsilon_F - \varepsilon_1)^2/(2\pi \hbar^2)$ and $t = \mu_0 e^2 (\varepsilon_1 - \varepsilon_F)/(4\pi m^*)$.

Letting the observation point $z \to \pm \infty$ in Eqs. (4) and (5), one can obtain the field distribution outside the SQW taking the asymptotic forms as follows [4,6]

$$\tilde{\mathbf{E}}(z) = \mathbf{E}_0 e^{\mathrm{i}k_{\perp}z} + \left(1, 0, \frac{k_{\parallel}}{k_{\perp}}\right) \tilde{\mathbf{E}}_{\mathrm{x}}^R e^{-\mathrm{i}k_{\perp}z}, \quad z \to -\infty, \tag{18}$$

$$\tilde{\mathbf{E}}(z) = \mathbf{E}_0 e^{\mathrm{i}k_{\perp}z} + \left(1, 0, -\frac{k_{\parallel}}{k_{\perp}}\right) \tilde{\mathbf{E}}_x^T e^{\mathrm{i}k_{\perp}z}, \quad z \to \infty, \tag{19}$$

where

$$\tilde{\mathbf{E}}_{x}^{R} = \frac{c^{2}}{2i\omega^{2}\epsilon^{B}} \int_{0}^{L} [\alpha N_{x}k_{\perp}\Psi(z) + \beta N_{z}k_{\parallel}\zeta(z)]e^{ik_{\perp}z}dz, \tag{20}$$

$$\tilde{\mathbf{E}}_{x}^{T} = \frac{c^{2}}{2i\omega^{2}\epsilon^{B}} \int_{0}^{L} [\alpha N_{x}k_{\perp}\Psi(z) - \beta N_{z}k_{\parallel}\zeta(z)]e^{-ik_{\perp}z}dz. \tag{21}$$

Thus the reflectivity (R_p) , transmittance (T_p) of the SQW system are given, respectively, via

$$R_p = \left| \frac{\tilde{E}_{\chi}^R}{E_0 \cos \theta} \right|^2, \qquad T_p = \left| 1 + \frac{\tilde{E}_{\chi}^T}{E_0 \cos \theta} \right|^2, \tag{22}$$

where θ is the incident angle. Thus the optical absorption coefficient (absorbance) is defined as

$$A_p = 1 - R_p - T_p. (23)$$

With these coefficients obtained, we now investigate the radiative shift and broadening of the optical absorption spectra due to the nonlocality of optical responses in the AlGaAs/GaAs SQW with an applied static electric field. We adopt $m^*=0.067m_0$ (m_0 is the mass of a free electron), $\epsilon^B=13.1$, $N_d=10^{17}/{\rm cm}^3$, $\theta=70^0$ and $\hbar\Gamma_0=4.7$ meV [6]. From Eq. (17), one can see that the resonance structures of the optical spectra are determined by

the complex roots of the $det|\mathbf{D}| = 0$. From the expressions of D_{xx} and D_{zz} , we can see the radiative shift and broadening of the optical spectra are attributed to the contributions from the real and imaginary parts of the two terms hM_{xx} and tM_{zz} , respectively. It is evident from the expressions of M_{xx} and M_{zz} that the physical origin of the radiative shift and broadening of the optical spectra lies in the coupling between the dynamical variables (i.e., the microscopic current-densities accompanying the change of the electronic wave functions) via the electromagnetic propagator $G(z, z'; \omega)$. Due to the absence of the quantum-size effects in the x-direction, the contributions from the term hM_{xx} to the radiative shift and broadening of the resonance peak are very small and can be omitted. For seeing the variation of the radiative shift and broadening of the resonance peak with the applied static field, they are plotted as a function of the applied static field F in Figs. 1 and 2, respectively. The solid, dashed and dotted lines are for three different QW widths: (a) 20 nm, (b) 18 nm, (c) 15 nm, respectively. From Figs. 1 and 2, we can see that the nonlocality of optical responses can lead to the radiative shift (i.e., blueshift relative to the energy interval ε_{21}) and broadening of the line spectrum. With an increase of the applied static field F, the blue-shift will decrease monotonously. While broadening of the line spectrum will decrease evidently at first, and then increase slightly. From Figs. 1 and 2, we can also see that the blue-shift and broadening of the line spectrum are more evident in largesize QW's. In the case of the local optical responses, however, there aren't the radiative shift and broadening of the resonance peak in the optical spectra [12].

In Fig. 3, we plot the radiative shift $(\Delta\hbar\omega)$ versus the width of the QW for three different applied electric fields F: (a) 1.0×10^7 V/m, (b) 2.0×10^7 V/m, (c) 3.0×10^7 V/m. The solid lines indicate that the results are calculated by the optical absorption

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