Modification of electron-hole excitations for a quantum well embedded in an asymmetric dielectric structure

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Starting from the conventional quantum electrodynamical formulation in Coulomb gauge we show that the Coulomb interaction of an electronhole in a quantum well (QW) embedded in an asymmetric dielectric structure with different dielectric constants in barrier regions yields a nondiagonal term of electron-hole pair excitation and the electron-hole vacuum state cannot be longer than the ground state of the system. A Bogoliubov transformation with a representation of new quasiparticles is provided to eliminate the nondiagonal term. The linear response function describing the modified exciton-polariton effect of the system is obtained. Analytical expressions are found for the effective mass of quasiparticles, the modified band gap, and the exciton coupling potential for a quasi-electron-hole pair. The exciton-polariton coupling turns out to contain an additional term originating from intraband processes. It is shown that the effect is significant for QW's with small thicknesses and for semiconductors with narrow band gaps.

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I. INTRODUCTION

In recent years the theory of excitonpolaritons in semiconductor microstructures has attracted considerable interest for many optical device applications.[1] In the early works of exciton polariton in quantum wells (QW's) the background dielectric constant has been taken to be the same for QW and barrier regions. Keldysh[2] is the first who showed that the different dielectric constants in the QW and barrier regions cause an image charge effect which significantly modifies the coupling potential of the electron-hole pair. Later on, this effect was studied in detail by M. Kumagai *et al.*[3] and Tran Thoai[4] for the general case of different dielectric constants in each barrier region. Recently, the effect of dielectric confinement has been shown experimentally in semiconductor nanostructures.[5] This effect of near-surface quantum wells has also been studied for the cases of magnetoexcitons.[6–8] The starting Hamiltonian used in these works to describe the dielectric confinement effect represents only the exciton model with an electron-hole Coulomb coupling potential modified by the inhomogeneous background dielectric constant via the Poisson equation.

In the present paper a more general approach is provided to study the full electromagnetic interaction of a material system consisting of a QW embedded in a space-dependent dielectric. Following the conventional electrodynamical formulation in Coulomb gauge[9] we consider the dielectric as a part of the material system and introduce its own current and charge density $\mathbf{j}_b(\mathbf{r})$ and $\varrho_b(\mathbf{r})$, respectively. It is suggested that a suitable constitutive relation should be held for them to yield the corresponding dielectric constant $\epsilon(\mathbf{r})$. We confine ourselves to the case of a medium with different dielectric constants for left and right barrier regions. The electron field operator for a two-band semiconductor QW is formulated using the envelope function approach in the $\mathbf{k}\cdot\mathbf{p}$ approximation.[10,11] We show that for an asymmetric dielectric structure, the Coulomb interaction in QW's yields a nondiagonal term of electron-hole pair excitation and the electron-hole vacuum state cannot be longer than the ground state of the system. A Bogoliubov transformation with a representation of new quasiparticles is provided to eliminate the nondiagonal term. Using the effective-mass approximation for quasiparticles we obtain the linear response function of the system describing the modified exciton-polariton effect of the system. It is shown that the effect is significant for QW's of small thicknesses and for semiconductors with narrow band gaps.

II. HAMILTONIAN MODEL

We start with the Hamiltonian describing the electromagnetic interaction for a material system consisting of a two-band semiconductor QW embedded in a medium with space-dependent dielectric constant

$$H = \int \Psi^{+}(\mathbf{r})\left[-\frac{\hbar^2\Delta}{2m_0} + U_l(\mathbf{r}) + U_c(\mathbf{r})\right]\Psi(\mathbf{r})d\mathbf{r} - \frac{1}{c}\int \mathbf{A}(\mathbf{r})\cdot\mathbf{J}(\mathbf{r})d\mathbf{r} + H_c + \sum_{\mathbf{q},\lambda}\hbar\omega_q c^{+}_{\mathbf{q}\lambda}c_{\mathbf{q}\lambda}. \qquad (1)$$

The Coulomb interaction H_c concerning QW electron field operator $\Psi(\mathbf{r})$ may be presented in Coulomb gauge as follows:

$$H_c = \frac{1}{2}e^2\int\int \Psi^{+}(\mathbf{r})\frac{\Psi^{+}(\mathbf{r}')\Psi(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\Psi(\mathbf{r})d\mathbf{r}\,d\mathbf{r}' + e\int\int \Psi^{+}(\mathbf{r})\frac{\varrho_b(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\Psi(\mathbf{r})d\mathbf{r}\,d\mathbf{r}'. \qquad (2)$$

The vector potential can be expanded in terms of the photon creation (destruction) operators $c^{+}_{\mathbf{q}\lambda}(c_{\mathbf{q}\lambda})$:

$$\mathbf{A}(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{q}}\mathbf{A}(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}},$$

$$\mathbf{A}(\mathbf{q}) = \sum_\lambda c\sqrt{\frac{2\pi\hbar V}{\omega_q}}\mathbf{e}_{\mathbf{q}\lambda}(c_{\mathbf{q}\lambda} + c^+_{-\mathbf{q}\lambda}).$$

We would like to emphasize that here we have the bare photons whose definition is independent of the presence of the material system including the dielectric. The periodic potential of the perfect bulk semiconductor is denoted as $U_l(\mathbf{r})$, while $U_c(\mathbf{r})$ is the additional confining potential due to the heterostructures.[10,11] The electron-hole field operator of the two-band quantum well semiconductor $\Psi(\mathbf{r})$ is represented as

$$\Psi(\mathbf{r}) = \sum_{\mathbf{K}}[a_{\mathbf{K}}\psi_{c\mathbf{K}}(\mathbf{r}) + b^+_{-\mathbf{K}}\psi_{v\mathbf{K}}(\mathbf{r})], \tag{3}$$

where $a_{\mathbf{K}}(a^+_{\mathbf{K}}), b_{\mathbf{K}}(b^+_{\mathbf{K}})$ are the electron and hole destruction (creation) operators, respectively. Here, and in the following the capital letters $\mathbf{R},\mathbf{K},\ldots$, are used to indicate two-dimensional (2D) vectors parallel to the plane of layers (x,y): $\mathbf{R}$ is the 2D part of $\mathbf{r}$, $\mathbf{K}$ is the respective part of $\mathbf{k}$. Within the framework of the envelope function description and the $\mathbf{k}\cdot\mathbf{p}$ approximation the wave functions $\psi_{s\mathbf{K}}(\mathbf{r}),(s=c,v)$ may be written as[10,11]

$$\psi_{c\mathbf{K}}(\mathbf{r}) = F_{c\mathbf{K}}(\mathbf{r})u_c(0,\mathbf{r}) - i\hbar\frac{\nabla F_{c\mathbf{K}}(\mathbf{r})\cdot\mathbf{v}}{E_c(0)-E_v(0)}u_v(0,\mathbf{r}),$$

$$\psi_{v\mathbf{K}}(\mathbf{r}) = F_{v\mathbf{K}}(\mathbf{r})u_v(0,\mathbf{r}) + i\hbar\frac{\nabla F_{v\mathbf{K}}(\mathbf{r})\cdot\mathbf{v}^*}{E_c(0)-E_v(0)}u_c(0,\mathbf{r}),$$

$$F_{s\mathbf{K}}(\mathbf{r}) = \zeta_s(z)\frac{e^{i\mathbf{K}\cdot\mathbf{R}}}{\sqrt{S}}. \tag{4}$$

Here $E_s(k)$ and $u_s(\mathbf{k},\mathbf{r})$ are the band energies and the periodic parts of the Bloch functions for the perfect bulk semiconductor, respectively, while the vector $\mathbf{v}$ is the interband matrix element of the velocity. The envelope functions $\zeta_s(z),(s=c,v)$ are eigenfunctions of

$$\left[-\frac{\hbar^2}{2m_s}\frac{\partial^2}{\partial z^2} + U_c^s(z)\right]\zeta_s(z) = \varepsilon_s\zeta_s(z). \tag{5}$$

We will confine ourselves to type-I heterostructures,[11] i.e., the $\zeta_s(z)$'s are localized solutions for both conduction and valence bands. For the conduction band, $s=c, m_c \equiv m_e > 0, \varepsilon_c$ is the lowest level of Eq. (5). For the valence band, $s=v, m_v \equiv -m_h < 0, \varepsilon_v$ is the highest level of Eq. (5). The bare electron and hole energies are defined as

$$E_e(\mathbf{K}) \equiv \varepsilon_c + E_c(\mathbf{K}), \quad E_h(-\mathbf{K}) \equiv -\varepsilon_v - E_v(\mathbf{K}).$$

The current density $\mathbf{J}$ is taken to be

$$\mathbf{J}(\mathbf{r}) = \mathbf{j}(\mathbf{r}) + \mathbf{j}_b(\mathbf{r}),$$

where

$$\mathbf{j}(\mathbf{r}) = -\frac{i\hbar e}{2m_o}\{\Psi^+(\mathbf{r})\nabla\Psi(\mathbf{r}) - [\nabla\Psi^+(\mathbf{r})]\Psi(\mathbf{r})\} \tag{6}$$

represents the electron current in two-band semiconductor and $\mathbf{j}_b(\mathbf{r})$ and $\varrho_b(\mathbf{r})$ are the current and charge density, respectively, responsible for the background dielectric constant $\epsilon(z)$. The latter one is a piecewise function as follows:

$$\epsilon(z) = \epsilon_w \quad \text{for } -\frac{1}{2}l < z < \frac{1}{2}l \text{ (QW region)},$$

$$\epsilon(z) = \epsilon_L \quad \text{for } z < -\frac{1}{2}l \text{ (left barrier region)},$$

$$\epsilon(z) = \epsilon_R \quad \text{for } z > \frac{1}{2}l \text{ (right barrier region)} \tag{7}$$

III. EQUATIONS OF MOTION

We can derive now the Heisenberg equations of motion for any operator $\hat{X}$ representing a linear combination of the electron- and hole destruction and creation operators. To this aim we have to write out the commutator

$$[H_c,\hat{X}] = e\int\{\Psi^+(\mathbf{r})\Phi(\mathbf{r})[\Psi(\mathbf{r}),\hat{X}]^+ - [\Psi^+(\mathbf{r}),\hat{X}]^+\Phi(\mathbf{r})\Psi(\mathbf{r})\}d\mathbf{r}. \tag{8}$$

Here $[A,B]^+ \equiv AB+BA$ and $\Phi(\mathbf{r})$ is defined as

$$\Phi(\mathbf{r}) = \int\frac{e\Psi^+(\mathbf{r}')\Psi(\mathbf{r}') + \varrho_b(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'. \tag{9}$$

The charge density operator $\varrho_b(\mathbf{r})$ responsible for the background dielectric constant is connected with the electric field operator by the following constitutive relation:

$$\varrho_b(\mathbf{r}) = -\operatorname{div}\mathbf{P}(\mathbf{r}), \quad \mathbf{j}_b(\mathbf{r}) = \frac{\partial}{\partial t}\mathbf{P}(\mathbf{r}),$$

$$\mathbf{P}(\mathbf{r}) = \frac{\epsilon(z)-1}{4\pi}\mathbf{E}(\mathbf{r}), \quad \mathbf{E}(\mathbf{r}) = -\nabla\Phi(\mathbf{r}) + \mathbf{E}^t(\mathbf{r}). \tag{10}$$

Here the transverse electric field operator $\mathbf{E}^t(\mathbf{r})$ is defined as

$$\mathbf{E}^t(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{q}}\mathbf{E}^t(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}},$$

$$\mathbf{E}^t(\mathbf{q}) = i\sum_\lambda\sqrt{2\pi\hbar\omega_q}\mathbf{e}_{\mathbf{q}\lambda}(c_{\mathbf{q}\lambda} - c^+_{-\mathbf{q}\lambda}). \tag{11}$$

Substituting Eq. (10) into Eq. (9) and performing the integration we can find $\Phi(\mathbf{r})$ presented as follows:

$$\Phi(\mathbf{r}) = e\int G(\mathbf{r}|\mathbf{r}')\Psi^+(\mathbf{r}')\Psi(\mathbf{r}')d\mathbf{r}' + F(\mathbf{r}),$$

$$G(\mathbf{r}|\mathbf{r}') = \frac{1}{S}\sum_{\mathbf{Q}}G(z,z'|Q)e^{i\mathbf{Q}\cdot\mathbf{R}-\mathbf{R}'},$$

$$F(\mathbf{r}) = \frac{1}{S}\sum_{\mathbf{Q}} F(z|\mathbf{Q}) e^{i\mathbf{Q}\cdot\mathbf{R}}. \tag{12}$$

The expressions of $G(z,z'|Q)$ and $F(z|\mathbf{Q})$ for the QW region are found to be

$$G(z,z'|Q) = \frac{2\pi}{\epsilon_w Q}\left(e^{-Q|z-z'|} + \frac{e^{-Ql}}{1-\delta_L\delta_R e^{-2Ql}}\left[\delta_L e^{-Q(z+z')} + \delta_R e^{Q(z+z')} + \delta_L\delta_R e^{-Ql}\left(e^{-Q(z-z')} + e^{Q(z-z')}\right)\right]\right)$$

$$\delta_n \equiv \frac{\epsilon_w - \epsilon_n}{\epsilon_w + \epsilon_n} \quad (n = L,R),$$

$$F(z|\mathbf{Q}) = \frac{1}{L}\sum_{q_z} \frac{E_z^t(\mathbf{q})}{1-\delta_L\delta_R e^{-2Ql}} \times\left[\frac{\delta_L}{Q}\left(-e^{-iq_z l/2} + e^{iq_z l/2}\delta_R e^{-Ql}\right)e^{-Q(z+l/2)} + \frac{\delta_R}{Q}\left(-e^{-iq_z l/2}\delta_L e^{-Ql} + e^{iq_z l/2}\right)e^{Q(z-l/2)}\right]. \tag{13}$$

We have the following result for the left barrier region:

$$G(z,z'|Q) = \frac{4\pi}{Q(\epsilon_w+\epsilon_L)(1-\delta_L\delta_R e^{-2Ql})} \times\left[e^{Q(z-z')} + \delta_R e^{-Ql} e^{Q(z+z')}\right],$$

$$F(z|\mathbf{Q}) = \frac{1}{L}\sum_{q_z}\frac{E_z^t(\mathbf{q})e^{Q(z+l/2)}}{1-\delta_L\delta_R e^{-2Ql}}\left[-e^{-iq_z l/2}\delta_L(1+\delta_R e^{-2Ql}) + e^{iq_z l/2}\delta_R(1+\delta_L)e^{-Ql}\right] \tag{14}$$

and for the right barrier region

$$G(z,z'|Q) = \frac{4\pi}{Q(\epsilon_w+\epsilon_R)(1-\delta_L\delta_R e^{-2Ql})}\left[e^{-Q(z-z')} + \delta_L e^{-Ql} e^{-Q(z+z')}\right],$$

$$F(z|\mathbf{Q}) = \frac{1}{L}\sum_{q_z}\frac{E_z^t(\mathbf{q})e^{-Q(z-l/2)}}{1-\delta_L\delta_R e^{-2Ql}}\left[-e^{-iq_z l/2}\delta_L(1+\delta_R)e^{-Ql} + e^{iq_z l/2}\delta_R(1+\delta_L e^{-2Ql})\right]. \tag{15}$$

Equation (9) is the Poisson's equation written in integral form. The expressions (12)–(15) represent its solution for the special case of charge density $\varrho_b(\mathbf{r})$ defined by Eq. (10). The potential $\Phi(\mathbf{r})$ turns out to be connected with the z component of the transverse field. Formally, it is due to the fact that the constitutive relation (10) is connected with the total electric field. It is not a surprising fact because the longitudinal and transverse fields are strictly separated only for isotropic and homogeneous media. For the case of anisotropic and inhomogeneous medium they may not be independent components. However, this interdependence gives no contribution to the linear constitutive relation which is the purpose of this work. With Eqs. (12) and (13) substituted into Eq. (8) we can find the result as follows:

$$[H_c, a_{\mathbf{K}}] = \hat{N}_a(3) + \varepsilon b^+_{-\mathbf{K}}, \quad [H_c, b_{-\mathbf{K}}] = \hat{N}_b(3) - \varepsilon a^+_{\mathbf{K}} - \Delta_h b_{-\mathbf{K}}, \tag{16}$$

$$\hat{N}_a(3) = -\frac{e}{S}\sum_{\mathbf{Q}}\int\left[\Phi(\mathbf{r})a_{\mathbf{K}-\mathbf{Q}}\zeta_c^2(z) - b^+_{-\mathbf{K}+\mathbf{Q}}\Phi(\mathbf{r})\mathbf{d}\frac{\partial}{\partial\mathbf{r}}\zeta_c(z)\zeta_v(z)\right]e^{-i\mathbf{Q}\cdot\mathbf{R}}d\mathbf{r},$$

$$\hat{N}_b(3) = \frac{e}{S}\sum_{\mathbf{Q}}\int\left[\Phi(\mathbf{r})b_{-\mathbf{K}-\mathbf{Q}}\zeta_v^2(z) - a^+_{\mathbf{K}+\mathbf{Q}}\Phi(\mathbf{r})\mathbf{d}\frac{\partial}{\partial\mathbf{r}}\zeta_c(z)\zeta_v(z)\right]e^{-i\mathbf{Q}\cdot\mathbf{R}}d\mathbf{r},$$

$$\varepsilon = \frac{e^2 d_z}{S}\sum_{\mathbf{Q}}\int\int\zeta_v^2(z)\left[\frac{\partial}{\partial z'}G(z,z'|Q)\right]\zeta_c(z')\zeta_v(z')dzdz',$$

$$\Delta_h = \frac{e^2}{S}\sum_{\mathbf{Q}}\int\int\zeta_v^2(z)G(z,z'|Q)\zeta_v^2(z')dzdz',$$

$$\mathbf{d} \equiv \frac{i\hbar\mathbf{v}}{E_c(0) - E_v(0)}. \tag{17}$$

Here Δ_h represents the Coulomb correction to the hole energy. $\hat{N}_a(3)$ and $\hat{N}_b(3)$ are composed of the terms containing normal ordered products of electron-hole creation or annihilation operators with the potential operator $\Phi(\mathbf{r})$.

It is not difficult to see that ε disappears for a symmetric structure when $\epsilon_L = \epsilon_R$ and the functions $\zeta_c(z)\zeta_v(z), \zeta_v^2(z)$ are even of z. In the present paper we consider the case $\epsilon_L \neq \epsilon_R$. For simplicity the envelope function $\zeta(z)$ is taken to be the same for both conduction and valence bands and the function $\zeta^2(z)$ is suggested to be even. The expression of ε may be represented as follows:

$$\varepsilon = \frac{e^2 d_z}{\epsilon_w l^2}(\delta_R - \delta_L)I(\mu), \quad \mu \equiv \delta_L\delta_R,$$

$$I(\mu) = \int dz\int dz'\int_0^\infty d\eta\,\zeta^2(z)\zeta^2(z')\frac{\eta e^{-\eta}\cosh\left(\eta\frac{z+z'}{l}\right)}{1-\mu e^{-2\eta}}. \tag{18}$$

IV. QUASIPARTICLE REPRESENTATION AND MODIFIED EXCITON CONSTITUTIVE RELATION

With ε different from zero the equations of motion (16) show that the electron-hole vacuum state is not a stationary one and cannot be the ground state of the system. One can apply a Bogoliubov canonical transformation similar to the one used in Refs. 12 and 13 to eliminate the nondiagonal term related with ε. The new creation (α^+,β^+) and annihilation (α,β) operators for quasiparticles are introduced by relations

$$\alpha_{\mathbf{K}} = a_{\mathbf{K}} \cos \vartheta_{\mathbf{K}} + b^+_{-\mathbf{K}} \sin \vartheta_{\mathbf{K}},$$

$$\beta^+_{-\mathbf{K}} = - a_{\mathbf{K}} \sin \vartheta_{\mathbf{K}} + b^+_{-\mathbf{K}} \cos \vartheta_{\mathbf{K}}. \tag{19}$$

The Bogoliubov transformation (19) may be interpreted as using new basic set of one particle functions being linear combinations of the electron-hole eigenfunctions (4) to provide the quantization of electron field operator (3). The parameter $\vartheta_{\mathbf{K}}$ is a free one. These new oneparticle functions now cease to be eigenfunctions of the electron-hole free part of Hamiltonian [the first term in Eq. (1)]. In the new representation this part of the Hamiltonian (1) will be nondiagonal. Its nondiagonal term affects the Heisenberg equation of motion to provide a term similar to the one containing ε of Eq. (16). The choice of parameter $\vartheta_{\mathbf{K}}$ now is made from the requirement that the total nondiagonal term arising in Heisenberg equations should be cancelled. The coefficients of transformation are found to be

$$\tan 2\vartheta_{\mathbf{K}} = - \frac{2\varepsilon}{E_e(\mathbf{K}) + E_h(-\mathbf{K})}. \tag{20}$$

The quasiparticle energies are determined as follows:

$$E_\alpha(\mathbf{K}) = \frac{1}{2}\left(E_e(\mathbf{K}) - E_h(-\mathbf{K}) + \frac{[E_e(\mathbf{K}) + E_h(-\mathbf{K})]^2}{\sqrt{[E_e(\mathbf{K}) + E_h(-\mathbf{K})]^2 + 4\varepsilon^2}}\right),$$

$$E_\beta(\mathbf{K}) = \Delta_h + \frac{1}{2}\left(-E_e(\mathbf{K}) + E_h(-\mathbf{K}) + \frac{[E_e(\mathbf{K}) + E_h(-\mathbf{K})]^2 + 8\varepsilon^2}{\sqrt{[E_e(\mathbf{K}) + E_h(-\mathbf{K})]^2 + 4\varepsilon^2}}\right). \tag{21}$$

Using the effective mass approximation we can represent the quasiparticles energies as

$$E_\alpha(\mathbf{K}) = E_\alpha(0) + \frac{\hbar^2 K^2}{2m_\alpha},$$

$$\frac{1}{m_\alpha} = \frac{1}{m_e} + \frac{1}{2m'}\left[\frac{E_0}{\tilde{E}_0}\left(1 + \frac{4\varepsilon^2}{\tilde{E}_0^2}\right) - 1\right],$$

$$E_\beta(\mathbf{K}) = E_\beta(0) + \frac{\hbar^2 K^2}{2m_\beta},$$

$$\frac{1}{m_\beta} = \frac{1}{m_h} + \frac{1}{2m'}\left[\frac{E_0}{\tilde{E}_0}\left(1 - \frac{4\varepsilon^2}{\tilde{E}_0^2}\right) - 1\right]. \tag{22}$$

Here the following notation has been introduced:

$$E_0 \equiv E_e(0) + E_h(0), \quad \tilde{E}_0 \equiv \sqrt{E_0^2 + 4\varepsilon^2}, \quad \frac{1}{m'} \equiv \frac{1}{m_e} + \frac{1}{m_h}. \tag{23}$$

Our aim is to establish the constitutive relation connecting the current density $\langle \mathbf{j}(\mathbf{r},t)\rangle$ with the total electric field $\langle \mathbf{E}(\mathbf{r}',t')\rangle$. The expression (6) of the current density contains different products of quasiparticle operators such as $\alpha^+\alpha, \beta^+\beta$ and $\beta\alpha, \alpha^+\beta^+$. However, it is shown in Ref. 14 that only the polarization functions of quasiparticle pair excitation $\langle\beta\alpha\rangle$ and $\langle\alpha^+\beta^+\rangle$ are responsible for the linear process. Using the formalism suggested in our previous paper[15] we derive the Heisenberg equation of motion for these functions that yields the following linear constitutive relation:

$$\langle j_i(\mathbf{r},t)\rangle = \int d\mathbf{r}' \int dt' \sigma_{ij}(\mathbf{r},t|\mathbf{r}',t')\langle E_j(\mathbf{r}',t')\rangle,$$

$$\sigma_{ij}(\mathbf{r},t|\mathbf{r}',t') = \zeta^2(z)\zeta^2(z') \times \frac{1}{S}\sum_{\mathbf{Q}} \int \frac{d\omega}{2\pi}[\sigma_{ij}(\mathbf{Q},\omega)e^{i\mathbf{Q}\cdot\mathbf{R}-\mathbf{R}'}e^{-i\omega(t-t')} + cj],$$

$$\sigma_{ij}(\mathbf{Q},\omega) = i\frac{2\hbar e^2}{\tilde{E}_0}\sum_\nu \frac{U_i(\nu,\mathbf{R})U_j^*(\nu\mathbf{R})_{\mathbf{R}=0}}{\hbar\omega - E_g - \frac{\hbar^2 Q^2}{2M} - E_\nu},$$

$$U_i(\nu,\mathbf{R}) = v_i\varphi_\nu(\mathbf{R}) - i\frac{\hbar\varepsilon}{m'\tilde{E}_0}\frac{\partial \varphi_\nu(\mathbf{R})}{\partial R_i}, \quad E_g \equiv \Delta_h + \sqrt{E_0^2 + \varepsilon^2}. \tag{24}$$

The eigenfunctions $\varphi_\nu(\mathbf{R})$ and eigenenergies E_ν of the 2D exciton states are solutions of the equation

$$\left[-\frac{\hbar^2}{2m}\Delta_{\mathbf{R}} + V(\mathbf{R})\right]\varphi_\nu(\mathbf{R}) = E_\nu\varphi_\nu(\mathbf{R}),$$

$$V(\mathbf{R}) \equiv \left(1 - \frac{4\varepsilon}{\tilde{E}_0}\mathbf{d}\frac{\partial}{\partial\mathbf{R}}\right)V_0(R),$$

$$V_0(R) = -\frac{e^2}{S}\sum_{\mathbf{Q}} \int\int \zeta^2(z)G(z,z'|Q)\zeta^2(z')e^{i\mathbf{Q}\cdot\mathbf{R}}dzdz'. \tag{25}$$

Here $V_0(R)$ represents the Coulomb coupling potential modified by the dielectric heterostructure which is identical with the one defined by Tran Thoai.[4] m and M are the reduced and total masses of the quasiparticle pair, respectively. In the case of symmetric structure $(\varepsilon=0)$ Eq. (25) reduces to the one used in the Ref. 4 to determine exciton energies. The classical Maxwell equation may also be derived from the Hamiltonian (1) and together with the established constitutive relation form a closed set of equations describing the exciton-polaritons of the system.[15]

V. DISCUSSION

In the present paper we have used the band electron functions (4) including two-band mixture within $\mathbf{k}\cdot\mathbf{p}$ approximation.[10,11] It gives rise to a coupling of the polarization with the longitudinal induced field that yields a constitutive relation connecting with the total electric field.[15] Moreover, it also provides the asymmetric effect considered here.

Equation (16) gives the same result as if we had included from the start in the Coulomb Hamiltonian H_c a term h_c of the form

$$h_c = \sum_{\mathbf{K}} [-\varepsilon(b_{-\mathbf{K}}a_{\mathbf{K}} + a^{+}_{\mathbf{K}}b^{+}_{-\mathbf{K}}) + \Delta_h b^{+}_{\mathbf{K}}b_{\mathbf{K}}]. \qquad (26)$$

The nondiagonal term related with ε describe an electron-hole pair excitation. This effect may be interpreted as the coupling of the interband dipole ed_z with the electric field resulting from the dielectric inhomogeneity.[16]

The parameter ε is of the order $e^2 d_z/l^2$. With $l \sim 4$ nm, $d_z \sim 2$ nm we have $\varepsilon \sim 10^{-1}$ eV. The characteristic parameter $\xi \equiv \varepsilon/E_0$ defines the degree of modification for exciton state. The modification is significant when the band gap E_0 is comparable with ε. Several semiconductors may satisfy this condition (see, e.g., Ref. 17).

The band gap E_g is redefined from Eq. (24) taking into account the well-known Coulomb shift as well as the quasi-particle modification. The modification of the exciton state is twofold [see Eq. (25)]: it changes the reduce mass m and the coupling potential $V(\mathbf{R})$ determining the bound state ν. The reduce mass turns out to increase as follows:

$$\frac{1}{m} \equiv \frac{1}{m_\alpha} + \frac{1}{m_\beta}, \quad m = m' \frac{\sqrt{E_0^2 + \varepsilon^2}}{E_0}. \qquad (27)$$

An additional term appears in the coupling potential $V(\mathbf{R})$ to broke its symmetry: the coupling 2D potential is no longer isotropic. It now depends on the direction of 2D vector $\mathbf{d}_\perp \equiv (ed_x, ed_y)$ representing the projection of the interband dipole on the layer plane. For the case of small parameter ξ this additional term may be seen as a perturbation. Applying the standard procedure of perturbation theory one can find the negative shift δE for the lowest exciton energy level E_ν. A crude estimation gives

$$\frac{\delta E}{E_\nu} \sim \left(4\xi \frac{d_\perp}{a_0}\right)^2,$$

where a_0 is the Bohr radius. The symmetry breaking of the perturbation may also lead to the removal of degeneracy for some exciton levels.

The exciton-polariton coupling presented in the constitutive relation (24) is modified as follows: the term $v_i v_j^* |\varphi_\nu(\mathbf{R})|^2|_{\mathbf{R}=0}$ is replaced by $U_i(\nu,\mathbf{R})U_j^*(\nu,\mathbf{R})|_{\mathbf{R}=0}$. The additional terms appear as a result of the Bogoliubov transformation (19) involving the intraband terms to define the current density (6).

The study provided in our paper, for simplicity, has been confined to the case when the electron-hole envelope functions are strictly localized in the QW region. It implies a model of infinite barriers. We notice here that the asymmetric effect $(\varepsilon \neq 0)$ may also be provided in the case when the potential of confinement $U_c^s(z)$ in Eq. (5) is not a symmetric function of z.

The asymmetric effect considered in the present paper is provided entirely from the Coulomb interaction written in Coulomb gauge (2) and is not connected with the photon field. Formally, if the photon field is eliminated from Eqs. (1) and (10), the effect will still stand as it follows directly from our result. The constitutive relation (24) then connects the current density with the induced longitudinal field only.

In the present paper the conventional quantum electrodynamical formulation in Coulomb gauge is suggested to describe the electromagnetic interaction of the material system consisting of QW embedded in a space-dependent dielectric. For such a system, an alternative formalism has been developed by Vogel and Welsch[18] treating the electromagnetic field and the background dielectric together as a subsystem. In this formalism the scalar potential $\Phi'(\mathbf{r})$ and the vector potential $\mathbf{A}'(\mathbf{r})$ are introduced with the special gauge

$$\operatorname{div}[\epsilon(z)\mathbf{A}'(\mathbf{r})] = 0 \qquad (28)$$

The scalar potential $\Phi'(\mathbf{r})$ is defined by the relation

$$\Phi'(\mathbf{r}) = \int G(\mathbf{r}|\mathbf{r}'\Psi^{+}(\mathbf{r})\Psi(\mathbf{r})d\mathbf{r}, \qquad (29)$$

where the Green's function $G(\mathbf{r}|\mathbf{r}')$ satisfies

$$\operatorname{div}[\epsilon(z)\nabla G(\mathbf{r}|\mathbf{r}')] = -4\pi\delta(\mathbf{r}-\mathbf{r}') \qquad (30)$$

with the well known electrostatics-boundary conditions.

The Hamiltonian part describing the electromagnetic interaction has been shown to be of the form

$$H_{\text{int}} = \frac{1}{2}\int \Psi^{+}(\mathbf{r})\Psi^{+}(\mathbf{r}')G(\mathbf{r}|\mathbf{r}')\Psi(\mathbf{r}')\Psi(\mathbf{r})d\mathbf{r}d\mathbf{r}' - \frac{1}{c}\int \mathbf{A}'(\mathbf{r})\cdot\mathbf{j}(\mathbf{r})d\mathbf{r}, \qquad (31)$$

where $\mathbf{j}(\mathbf{r})$ is defined by Eq. (6).

With $\epsilon(z)$ defined by Eq. (7) one can solve Eq. (30) and find exactly the same expressions (13)–(15) for the Green's function. Using this result and the expression (3) for the electron-hole field operator $\Psi(\mathbf{r})$ one can find the relation

$$H_c' \equiv \frac{1}{2}\int \Psi^{+}(\mathbf{r})\Psi^{+}(\mathbf{r}')G(\mathbf{r}|\mathbf{r}')\Psi(\mathbf{r}')\Psi(\mathbf{r})d\mathbf{r}d\mathbf{r}' = \frac{1}{2}\int :\Psi^{+}(\mathbf{r})\Psi^{+}(\mathbf{r}')G(\mathbf{r}|\mathbf{r}')\Psi(\mathbf{r}')\Psi(\mathbf{r}):d\mathbf{r}d\mathbf{r}' + h_c, \qquad (32)$$

where the symbol $:AB,\ldots,:$ denotes the normal ordered product of operators $AB,\ldots$, and h_c is defined by Eq. (26).

H_c' yields the same result as Eq. (16) with $\Phi(\mathbf{r})$ replaced by $\Phi'(\mathbf{r})$. Applying the Bogoliubov transformation (19) one can arrive at the same result of linear constitutive relation (24) as presented in the previous section.

It may be shown that Φ' and $\mathbf{A}'$ with the gauge (28) are obtained as a result of the following Gauge transformation:

$$\Phi' \to \Phi - \frac{1}{c}\frac{\partial}{\partial t}\Lambda, \quad \mathbf{A}' \to \mathbf{A} + \nabla\Lambda, \quad \frac{1}{c}\frac{\partial}{\partial t}\Lambda \equiv F(\mathbf{r}), \tag{33}$$

where Φ and $\mathbf{A}$ are the scalar potential and the vector potential of Coulomb gauge, respectively, defined in the present work and $F(\mathbf{r})$ is determined from Eqs. (13)–(15).

The asymmetric effect is due to the fact that we take into account the terms containing transition dipole matrix element $e\mathbf{d}$ in the Coulomb interaction while they are neglected in many other works. We think that it is not consistent to keep this matrix element only in the coupling with photon field, while to omit it in the Coulomb interaction. Some aspect of this question has been discussed in our previous paper.[15] We notice here that the exciton model used in Refs. 3 and 4 considers only bound states of an electron-hole pair due to the coupling potential which is modified by the inhomogeneous background dielectric constant via the Poisson equation. This approach does not take into account all aspect of the Coulomb interaction in the system and the linear constitutive relation (24) cannot be established from this theory.

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