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Band-mixing effect on the emission spectrum of modulation-doped semiconductor quantum wells

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Emission spectra of n-type modulation-doped semiconductor quantum wells are calculated in a multivalley effective-mass model which includes the valence-band mixing. The many-body effect is taken into account by consideration of the single electron-hole pair excitations and the shakeup process. The resulting emission spectra for both parallel and perpendicular polarizations are found in reasonable agreement with the reported experimental data.

Recent experimental studies of the optical emission from electrons confined in GaAs-$Al_xGa_{1-x}As$ quantum wells have shown that the photoluminescence has an unexpectedly strong component with polarization normal to the planes.[1] Effective-mass theories that include excitonic interactions and conventional symmetry-breaking mechanisms do not account for this phenomenon. Although it was conjectured[1] that the many-body effect analogous to the shakeup in x-ray emission[2,3] may explain such a phenomenon, no calculations have been reported to verify the assumption.

In this Rapid Communication, we report a calculation of the emission spectra of modulation-doped GaAs-$Al_xGa_{1-x}As$ quantum wells, including the effect of valence-band mixing and the shakeup process. The results are found in reasonable agreement with the experimental data. The unexpectedly strong normal component in the emission spectrum is attributed to the combined effect of valence-band mixing at nonzero wave vectors[4–6] and the shakeup process. In the shakeup process, the electron-hole recombination is accompanied with all possible excitations of the final many-electron system.[2,3]

The initial state of the system of interest consists of a two-dimensional electron gas of N particles and a single hole, all confined in a semiconductor quantum well. The Hamiltonian for this many-body system is given by[6]

$$H=\sum_{\mathbf{k}}\epsilon_c(\mathbf{k})a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+\frac{1}{2}\sum_{\mathbf{k}\mathbf{k}'\mathbf{q}}v_{ee}(\mathbf{q})a_{\mathbf{k}+\mathbf{q}}^{\dagger}a_{\mathbf{k}'-\mathbf{q}}^{\dagger}a_{\mathbf{k}'}a_{\mathbf{k}}+\sum_{\mathbf{p}}\epsilon_v(\mathbf{p})b_{\mathbf{p}}^{\dagger}b_{\mathbf{p}}-\sum_{\mathbf{kpq}}v_{eh}(\mathbf{q})a_{\mathbf{k}+\mathbf{q}}^{\dagger}b_{\mathbf{p}+\mathbf{q}}^{\dagger}b_{\mathbf{p}}a_{\mathbf{k}}\ , \tag{1}$$

where $\epsilon_c(\mathbf{k})$ and $\epsilon_v(\mathbf{p})$ are the first conduction-subband and valence-subband dispersion relations, respectively, for the quantum well. Here we use $\mathbf{k}$, $\mathbf{p}$, and $\mathbf{q}$ to denote two-dimensional wave vectors for the electron, hole, and wave-vector transfer, respectively. $a_{\mathbf{k}}^{\dagger}$ ($a_{\mathbf{k}}$) and $b_{\mathbf{p}}^{\dagger}$ ($b_{\mathbf{p}}$) create (annihilate) an electron and hole, respectively. $v_{ee}(\mathbf{q})$ and $v_{eh}(\mathbf{q})$ denote the electron-electron and electron-hole Coulomb interactions, respectively. The function $\epsilon_c(k)$ is given in the effective-mass approximation by a simple parabolic expression. The function $\epsilon_v(\mathbf{p})$ is obtained by a multiband effective-mass method as described in Ref. 5. The electron-electron interaction is given by[5]

$$v_{ee}(\mathbf{q})=\int dz_1\int dz_2|f_c(z_1)|^2|f_c(z_2)|^2\times\frac{e^2}{\epsilon_0 q}\exp(-q|z_1-z_2|)\ , \tag{2}$$

where ϵ_0 is the static dielectric constant and $f_c(z)$ is the lowest-energy solution to the effective-mass equation for an electron in a one-dimensional potential $V_e(z)=V_s(z)+V_d(z)$. Here, $V_s(z)$ is the square-well potential for the electron and V_d is a smooth-varying potential, describing the band-bending due to doping. The forms for $V_s(z)$ and $V_d(z)$ are described in Ref. 5. The electron-hole interaction is similarly given by

$$v_{eh}(\mathbf{q})=\int dz_e\int dz_h|f_c(z_e)|^2|g_v(z_h)|^2\times\frac{e^2}{\epsilon_0 q}\exp(-q|z_e-z_h|)\ , \tag{3}$$

where $g_v(z)$ is lowest-energy solution to the multiband effective-mass equation for a spin-$\frac{3}{2}$ hole in a one-dimensional potential $V_h(z)=\bar{V}_s(z)-V_d(z)$. Here, $\bar{V}_s(z)$ is the square-well potential for the hole (see Ref. 5).

We donate the N-electron ground state in the absence of the hole as Φ_N, and consider the fundamental excitation of Φ_N due to the presence of the hole. At low temperature we assume that the N electrons fill a Fermi sphere up to k_F, the Fermi wave vector. We define $\Phi_{\mathbf{kq}}\equiv a_{\mathbf{k}+\mathbf{q}}^{\dagger}a_{\mathbf{k}}b_{\mathbf{q}}^{\dagger}\Phi_N$ as the single electron-hole pair excitation, in which an electron in the lowest conduction subband with wave vector $\mathbf{k}$ ($|\mathbf{k}|<k_F$) is excited outside the Fermi surface ($|\mathbf{k}+\mathbf{q}|>k_F$) (it is not possible to excite the electron into a different subband within our approximation) and a hole with wave vector $\mathbf{q}$ is created. Note that all such excitations have zero total momentum and can be coupled together via the electron-hole interaction. We express the N-electron plus one-hole state Ψ_i as a linear combination of the single electron-hole pair excitations, viz.,

$$\Psi_i=\sum_{\mathbf{k},\mathbf{q}}F_i(\mathbf{k},\mathbf{q})\Phi_{\mathbf{kq}}\ , \tag{4}$$

where $F_i(\mathbf{k},\mathbf{q})$ shall be called the envelope function. Here we have ignored the possible many-particle excitations in the state Ψ_i. The envelope function $F_i(\mathbf{k},\mathbf{q})$ can be found by solving the Schrödinger equation $(H-E_i)\Psi_i=0$, which can be cast into the equation

$$[\Delta_{\mathbf{kq}}+\epsilon_v(\mathbf{q})-E_i]F_i(\mathbf{k},\mathbf{q})-\sum_{\mathbf{q}'}v_{eh}(\mathbf{q}')F_i(\mathbf{k},\mathbf{q}+\mathbf{q}')+\sum_{\mathbf{q}'}v_{eh}(\mathbf{q}')F_i(\mathbf{k}-\mathbf{q}',\mathbf{q}+\mathbf{q}')=0\ , \tag{5}$$

with the constraint $F_i(\mathbf{k},\mathbf{q})=0$ if $\tilde{\epsilon}_c(\mathbf{k})>\epsilon_F$ or $\tilde{\epsilon}_c(\mathbf{k}$

$+\mathbf{q}) < \epsilon_F$ (ϵ_F is the Fermi energy). In (5) $\Delta_{\mathbf{kq}} \equiv \tilde{\epsilon}_c(\mathbf{k}+\mathbf{q}) - \tilde{\epsilon}_c(\mathbf{k})$, where $\tilde{\epsilon}_c(\mathbf{k})$ is the "renormalized" conduction-band dispersion relation. In the Hartree-Fock approximation, it is given by[6]

$$\tilde{\epsilon}_c(\mathbf{k}) = \epsilon_c(\mathbf{k}) - \frac{1}{2}\sum_{\mathbf{k}'} v_{ee}(\mathbf{k}-\mathbf{k}')\theta(\epsilon_F - \epsilon_n(\mathbf{k}'))$$

and in the parabolic approximation, we found

$$\tilde{\epsilon}_c(\mathbf{k}) \simeq \frac{\hbar^2 k^2}{2m_e^*} + \frac{e^2 k_F}{\epsilon_0}\left[\left(\frac{k}{2k_F}\right)^2 - 1\right] .$$

The parabolic approximation is fairly good for $k < 2k_F$. For $k > 2k_F$, we found that the band renormalization effect is very small and we neglected it completely in our calculation. The second term in (5) describes the Coulomb scattering process in which an electron is scattered from $(\mathbf{k}+\mathbf{q})$ to $(\mathbf{k}+\mathbf{q}+\mathbf{q}')$ and a hole is scattered from $\mathbf{q}$ to $(\mathbf{q}+\mathbf{q}')$. The third term in (5) describes the Coulomb scattering process in which a hole left in the Fermi sea is scattered from $\mathbf{k}$ to $(\mathbf{k}-\mathbf{q}')$ and a hole in the valence band is scattered from $\mathbf{q}$ to $(\mathbf{q}+\mathbf{q}')$. Note that in the limit $N=1$ ($\epsilon_F=0$), the third term in (5) vanishes and (5) reduces to the effective-mass equation for the exciton problem.

To simplify the problem, we shall ignore the third term in (5) completely. This term makes a very small contribution, because the envelope function for the ground state is sharply peaked at $|\mathbf{k}| = k_F$. Equation (5) is now decoupled for each $\mathbf{k}$, and the initial states can be donoted as $\Psi_i(\mathbf{k})$. To find the solution $F_i(\mathbf{k},\mathbf{q})$, we expand it in terms of many Gaussian functions of the form $\exp(-\alpha\Delta_{\mathbf{kq}})$, viz.,

$$F_i(\mathbf{k},\mathbf{q}) = \sum_{\alpha} C_{\mathbf{k}}(\alpha)\exp(-\alpha\Delta_{\mathbf{kq}}) \quad . \tag{6}$$

The expansion (6) is desirable for several reasons: (1) it gives an accurate exciton binding energy and wave function in the limit $N=1$, (2) it is the most general form for the ground state if the hole kinetic energy vanishes; thus it is a very good approximation, as long as the hole effective mass is much larger than the electron effective mass, and (3) all matrix elements for these Gaussian functions are fairly easy to calculate. The expansion coefficients in (6) are nearly independent of $\mathbf{k}$. Thus for any $\mathbf{k}$ within the Fermi sphere, the envelope function takes the form $F^{(i)}(\Delta_{\mathbf{kq}})$ with the ground-state energy

$$E_0^{(i)}(\mathbf{k}) \simeq E_0^{(i)}(k_F) + [\tilde{\epsilon}_c(k_F) - \tilde{\epsilon}_c(\mathbf{k})] ,$$

where $E_0^{(i)}(k_F)$ is the lowest-energy solution to (5) with $|\mathbf{k}| = k_F$.

The energy $E_0^{(i)}(k_F)$ and the expansion coefficient $C_{\mathbf{k}}(\alpha)$ can be obtained numerically by solving a generalized eigenvalue problem in the nonorthogonal Gaussian basis. To simplify the calculation, a parabolic approximation has been used for the first valence subband. The hole effective mass is chosen to be $0.35m_0$ such that the kinetic-energy contribution is approximately the same as that obtained by using the exact expression. We find that the system has bound states for k_F less than 0.019 Å^{-1}. The binding energy of the system reduces to that of the exciton as $k_F \to 0$. We also note that the shape of the envelope function $F^{(i)}(\Delta_{\mathbf{kq}})$ does not change appreciably with the binding energy, even when the system becomes unbound. This is reasonable, because the reduction in binding energy arises from the more stringent constraint of the Fermi surface for increasing k_F, while the strength of the potential which determines the polarization remains the same.

Knowing the polarization in the initial state due to the electron-hole interaction, we are now ready to examine the emission spectrum of the system in the shakeup process. The possible final states of the system after the recombination of one electron-hole pair can be described by

$$\Psi_f(\mathbf{k}'\mathbf{q}) \equiv a^{\dagger}_{\mathbf{k}'+\mathbf{q}} a_{\mathbf{k}'} a_{\mathbf{q}} \Phi_N , \tag{7}$$

in which an electron at $\mathbf{q}$ is annihilated (due to the recombination with a hole at $\mathbf{q}$ in Ψ_i) and another electron at $\mathbf{k}'$ is simultaneously scattered off the Fermi sphere to a state with wave vector $(\mathbf{k}'+\mathbf{q})$ ($|\mathbf{k}'+\mathbf{q}| > k_F$). The emission spectrum at low temperatures is then given by

$$I_\epsilon(h\omega) \sim \sum_{\mathbf{k}} \exp[-E^{(i)}(\mathbf{k})/k_B T] \sum_{\mathbf{k}'\mathbf{q}} |\langle \Psi_i(\mathbf{k})|\hat{\epsilon}\cdot\mathbf{p}|\Psi_f(\mathbf{k}',\mathbf{q})\rangle|^2 \delta(\hbar\omega + E^{(f)}(\mathbf{k}',\mathbf{q}) - E^{(i)}(\mathbf{k})) , \tag{8}$$

where $E^{(f)}(\mathbf{k}',\mathbf{q}) \equiv \Delta_{\mathbf{k}'\mathbf{q}} - \tilde{\epsilon}_c(\mathbf{q})$ denotes the final-state energy and $\hat{\epsilon}$ denotes the polarization of the luminescence. A Boltzmann factor has been introduced because the system was initially kept at constant temperature T. Substituting (4) and (7) into (8) yields

$$I_\epsilon(\hbar\omega) \sim \sum_{\mathbf{q}} |\hat{\epsilon}\cdot\mathbf{P}(\mathbf{q})|^2 \sum_{\mathbf{k}} F^{(i)}(\Delta_{\mathbf{kq}}) \exp[-E^{(i)}(\mathbf{k})/k_B T] \delta(\hbar\omega + E^{(f)}(\mathbf{k},\mathbf{q}) - E^{(i)}(\mathbf{k})) , \tag{9}$$

where $\mathbf{P}(\mathbf{q})$ is the momentum matrix element between the conduction subband and the valence subband at $\mathbf{q}$. Note that $\mathbf{P}(\mathbf{q})$ varies strongly with $\mathbf{q}$ due to the valence-band mixing.[4,5] The squared momentum matrix elements for the transitions from the first heavy-hole (HH1) and first light-hole (LH1) subband to the first conduction subband (CB1) for two polarizations $\hat{\epsilon} \| \hat{\mathbf{x}}$ ($\hat{\mathbf{y}}$) and $\hat{\epsilon} \| \hat{\mathbf{z}}$ for a 221-Å GaAs-$Al_{0.23}Ga_{0.77}As$ quantum well are shown in Fig. 1.

In order to demonstrate the importance of the many-body effect, we have also calculated the emission spectra without considering the shakeup process. In this case the initial state is given by $\Psi_{\mathbf{q}}^{(i)} = b_{\mathbf{q}}^{\dagger}\Phi_N$ (the N-electron ground state plus a hole at $\mathbf{q}$) and the final state is given by $\Psi_{\mathbf{q}}^{(f)} = a_{\mathbf{q}}\Phi_N$ (a missing electron at q in the N-electron ground state). At a low temperature T, the emission spectrum can be derived according to the Fermi golden rule. We obtain

$$I_\epsilon(\hbar\omega) \sim \sum_{\mathbf{q}} \exp[-\epsilon_v(\mathbf{q})/k_B T] |\hat{\epsilon}\cdot\mathbf{P}(\mathbf{q})|^2 \delta(\hbar\omega - \epsilon_v(\mathbf{q}) - \tilde{\epsilon}_c(\mathbf{q}))\theta(\epsilon_F - \tilde{\epsilon}_c(\mathbf{q})) , \tag{10}$$

where we have ignored the small temperature fluctuation in the N-electron complex.

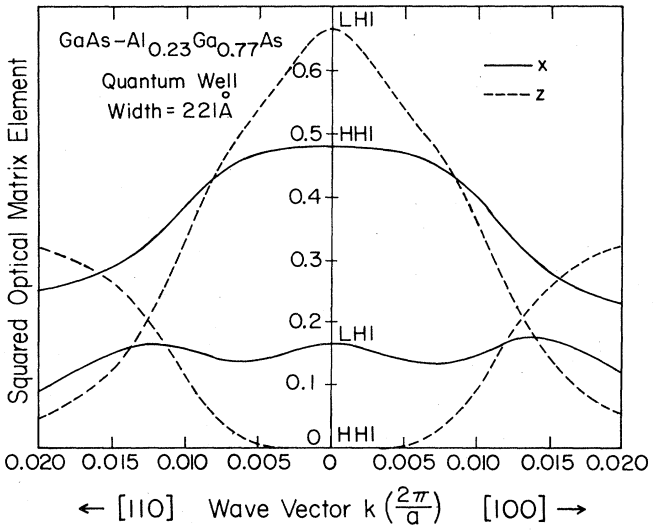

FIG. 1. Squared momentum matrix elements for the transitions from the first heavy-hole (HH1) and first light-hole (LH1) subband to the first conduction subband (CB1) for two polarizations $\hat{\epsilon} \| \hat{x}(\hat{y})$ (solid curves) and $\hat{\epsilon} \| \hat{z}$ (dashed curves) for a 221-Å GaAs-$Al_{0.23}Ga_{0.77}As$ quantum well.

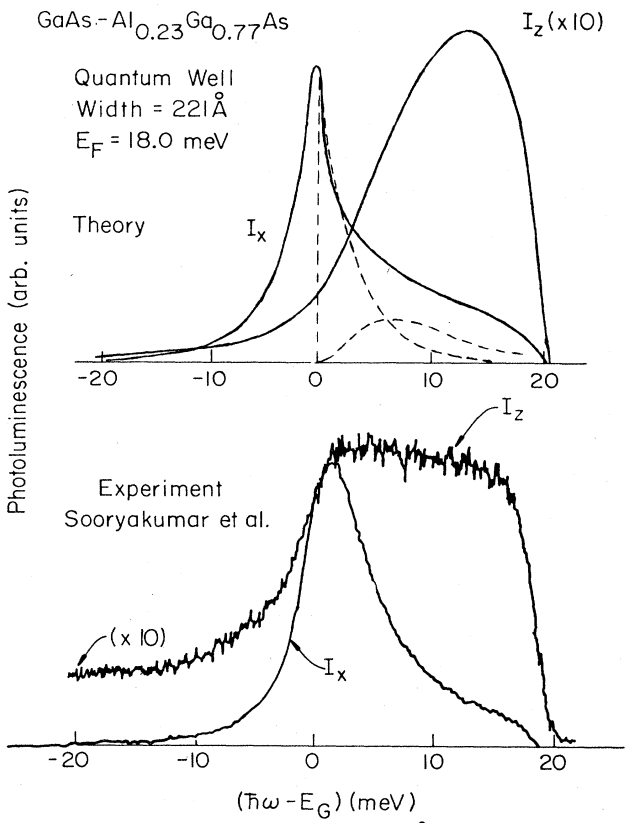

FIG. 2. The emission spectrum of a 221-Å GaAs-$Al_{0.23}Ga_{0.77}As$ modulation-doped quantum well. The Fermi level is about 18.0 meV. I_x and I_z denote the x and z polarization components, respectively. E_G is defined as $\epsilon_c(\mathbf{k}) + \epsilon_v(\mathbf{k})$ at $\mathbf{k} = 0$. Solid curve is with many-body effect; dashed curve is without many-body effect. The experimental data shown below are taken from Ref. 1.

The emission spectra obtained from (9) (solid curves) and from (10) (dashed curves) for the same quantum well with $\epsilon_F = 18.0$ meV at $T = 5$ K are shown in Fig. 2. The experimental data of Sooryakumar *et al.*, are also shown in the lower panel for comparison. Note that at $\epsilon_F = 18.0$ meV, the second conduction subband is also partially filled. The emission intensity from this subband, however, is negligible, since the HH1-CB2 transition matrix element is weak. The z-component emission spectra are multiplied by 10 in Fig. 2. As seen in Fig. 2, the results obtained from (10) which ignored the many-body effect are in strong disagreement with the data. The x-component emission spectrum obtained from (9) which includes the many-body effect is found in very good agreement with the experimental spectrum of Sooryakumar *et al.*[1] The z-component emission spectrum obtained from (9) is also in reasonable agreement with the data considering the approximations used.

The apparent large "mixing" for the z component in the initial state at $\hbar\omega \simeq E_G$, which is unexplainable by (10), can be accounted for by the combination of valence-band mixing at $q \neq 0$ and the shakeup process. The large z-component emission intensity at $\hbar\omega \simeq E_G$ comes from recombinations at $\mathbf{q} \neq 0$ where $|\hat{\mathbf{z}} \cdot \mathbf{P}(\mathbf{q})|^2$ is appreciable (see Fig. 1), accompanying an excitation in the $(N-1)$-electron final state. The sharp cutoff at $\hbar\omega - E_G \simeq E_F$ for the z component is also in qualitative agreement with the data.

The remaining discrepancy between the theory and experiment may be attributed to the omission of many-electron excitations of the initial state and other approximations used in our treatment.

In conclusion, we have studied the emission spectra of modulation-doped semiconductor quantum wells based on a multiband effective-mass model, including the valence-band mixing. The shakeup process is taken into account within the single electron-hole pair excitation approximation. The theoretical results compare very favorably with the experimental data. In particular, the anomalously large z-component emission intensity at $\hbar\omega \simeq E_g$ is at least qualitatively accounted for. We find that the polarization of the initial many-body system due to electron-hole Coulomb interaction, in which a hole at the top of the heavy hole subband is scattered into other states in the same subband, plays a key role in determining the emission spectra.

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