

## Many-body effects on intersubband transitions in semiconductor quantum-well structures

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Many-body effects on intersubband excitations in modulation-doped semiconductor quantum-well structures are calculated by solving numerically the Bethe-Salpeter equation for the corresponding polarization function. Depolarization and excitonic effects cause large changes in the transition energies. In particular, the excitonic binding tends to cancel the corrections due to exchange self-energies. Very good agreement between theoretical results and light scattering experiments is shown. The same formulation also applies to the interpretation of infrared optical absorption spectra.

During the past decade there has been great interest in quasi-two-dimensional electron systems in semiconductor heterojunction structures. Intersubband excitations, such as those active in infrared optical absorption and inelastic light scattering,<sup>1,2</sup> provide direct information on the behavior of the electron gas. Recent work has shown the significance of electron-electron interactions on intersubband transition energies.<sup>3-5</sup> These intersubband transitions are the basis of a novel class of infrared detectors.<sup>6</sup> Theory for linear and nonlinear intersubband optical absorption has also been discussed<sup>7</sup> without considering many-body effects.

The energy-level structure and transition energies of the electron gas in semiconductor quantum wells are often obtained in the local-density approximation (LDA).<sup>8,9</sup> In this paper we present a different approach to the calculation of energy levels and intersubband excitations. We use polarization response functions similar to the exciton Green's functions used in quantum-well interband optical absorption<sup>10</sup> and solve the corresponding Bethe-Salpeter equation numerically. In this approach many-body interactions are considered in the variational Hartree-Fock approximation, i.e., including nonlocal exchange. The calculated lowest-energy levels and intersubband excita-

tions take into account exchange self-energies, depolarization effects, and excitonic correlations (vertex corrections). We show that there is very good agreement between our calculations and inelastic light scattering measurements. There is also agreement with previous LDA calculations.<sup>8</sup> One interesting feature of our results is the prediction that in intersubband excitations the excitonic binding nearly cancels out the contribution of exchange self-energies.

We consider a quasi-two-dimensional electron system in the presence of an infrared optical field  $E(t)$  along the growth direction ( $z$ ), described by the Hamiltonian

$$H = \sum_{n,k,s} E_{nk}^H c_{nks}^\dagger c_{nks} + H_{e-e} - \sum_{n,m,k,s} \mu_{nm} E(t) c_{nks}^\dagger c_{mks}, \quad (1)$$

where  $n, m$  are the subband indices,  $k$  is the wave vector in the  $x$ - $y$  plane, and  $s$  refers to spin.  $\mu_{nm}$  is the intersubband dipole matrix element and  $E_{nk}^H$  is the  $n$ th subband Hartree energy which can be obtained self-consistently by solving the Poisson and Schrödinger equations simultaneously including the effects of the donors.  $H_{e-e}$  is the remaining electron-electron interaction,

$$H_{e-e} = \frac{1}{2} \sum_{n,n',m,m',k,k',q,s,s'} \sum' V_{nm,n'm'}(q) c_{nk+qs}^\dagger c_{n'k'-qs}^\dagger c_{m'k's} c_{mks}, \quad (2)$$

where

$$V_{nm,n'm'}(q) = V_q \delta_{nm} \delta_{n'm'} + V_q \int dz \int dz' f_n(z) f_m(z) (e^{-q|z-z'|} - 1) f_{n'}(z') f_{m'}(z'). \quad (3)$$

$V_q = 2\pi e^2/\epsilon q$  in cgs units and  $f_n(z)$  is the self-consistent  $n$ th subband Hartree wave function.

Define the density matrix as

$$\rho_{\alpha\beta s}(k) = \langle c_{\beta ks}^\dagger c_{\alpha ks} \rangle \quad (4)$$

and let  $E(t) = E_\omega \exp(-i\omega t) + \text{c.c.}$  For a two-band model and within the *rotating-wave* and variational Hartree-Fock approximations, we obtain the equation of motion for  $\rho_{21s}(k)$ , assuming only the lowest subband is occupied,

$$(E_{2k}^{\text{HF}} - E_{1k}^{\text{HF}} - \hbar\omega - i\gamma) \rho_{21s}(k) + \sum_{k'} V_{12,21}(0) [\rho_{21s}(k') + \rho_{21s}(k')] - \sum_{k'} V_{11,22}(k-k') \rho_{21s}(k') = n_{1k} \mu_{21} E_\omega e^{-i\omega t}, \quad (5)$$

where

$$E_{2k}^{\text{HF}} = E_{2k}^{\text{H}} - \sum_{k'} V_{12,21}(k-k') n_{1k'}, \quad (6)$$

$$E_{1k}^{\text{HF}} = E_{1k}^{\text{H}} - \sum_{k'} V_{11,11}(k-k') n_{1k'} \quad (7)$$

are the first and second subband Hartree-Fock energies, and  $\gamma$  is a phenomenological broadening.  $n_{1k} = \rho_{11\uparrow}(k) = \rho_{11\downarrow}(k) = \Theta(k_F - k)$ , where  $\Theta$  is the unit step function,  $k_F = (2\pi N_s)^{1/2}$  the Fermi wave vector, and  $N_s$  the quasi-two-dimensional carrier density. In the above equations, all Coulomb matrix elements are calculated using the static dielectric constant  $\epsilon_0$ , except for the depolarization field  $V_{12,21}(0)$  which is calculated without the singular first term on the right-hand side of Eq. (3) and using

$$\epsilon(\omega) = \epsilon_\infty \frac{\omega^2 - \omega_{\text{LO}}^2}{\omega^2 - \omega_{\text{TO}}^2}, \quad (8)$$

where  $\hbar\omega_{\text{LO}} = 36.7$  meV,  $\hbar\omega_{\text{TO}} = 33.6$  meV,  $\epsilon_\infty = 11.1$ , and  $\epsilon_0 = 13.2$ .

Equation (5) is the Bethe-Salpeter equation for the intersubband polarization function and corresponds to the summation of bubble and ladder diagrams. It describes intersubband transitions modified by exchange self-energies, depolarization effects, and excitonic correlations, all treated on equal footing. Since the source term on the right-hand side of Eq. (5) does not depend on spin and the intersubband charge polarizability is proportional to  $\rho_{21\uparrow} + \rho_{21\downarrow}$ , we obtain  $\rho_{21\uparrow} = \rho_{21\downarrow} \equiv \rho_{21}$ . Define a function  $P(k)$  by

$$\rho_{21}(k) = n_{1k} \mu_{21} E_\omega e^{-i\omega t} P(k). \quad (9)$$

Then the intersubband charge polarizability can be expressed as<sup>11</sup>

$$\begin{aligned} X^c(\omega) &= 2 \sum_k \mu_{12} \rho_{21}(k) / E_\omega e^{-i\omega t} \\ &= 2 |\mu_{21}|^2 \sum_{k \leq k_F} P(k), \end{aligned} \quad (10)$$

where  $P(k)$  satisfies

$$\begin{aligned} (E_{2k}^{\text{HF}} - E_{1k}^{\text{HF}} - \hbar\omega - i\gamma)P(k) \\ + \sum_{k' \leq k_F} [2V_{12,21}(0) - V_{11,22}(k-k')]P(k') = 1. \end{aligned} \quad (11)$$

This equation can be solved using a modified quadrature method similar to that employed in Ref. 10 to describe quantum-well interband absorption. Finally, the charge-density response  $I_{\text{CD}}$  seen in light scattering and infrared optical absorption is

$$I_{\text{CD}} \propto \text{Im} X^c(\omega) / \epsilon(\omega). \quad (12)$$

The intersubband spin polarizability  $X^s(\omega)$  and spin-density response  $I_{\text{SD}}$  can be determined in a similar fashion by considering  $\rho_{21\uparrow} - \rho_{21\downarrow}$  and a corresponding source term. We find

$$\begin{aligned} I_{\text{SD}} &\propto \text{Im} X^s(\omega) \\ &\propto \text{Im} \sum_{k \leq k_F} L(k), \end{aligned} \quad (13)$$

where  $L(k)$  satisfies

$$\begin{aligned} [E_{2k}^{\text{HF}} - E_{1k}^{\text{HF}} - \hbar\omega - i\gamma]L(k) \\ - \sum_{k' \leq k_F} V_{11,22}(k-k')L(k') = 1. \end{aligned} \quad (14)$$

Comparing the equation for  $L(k)$  with that for  $P(k)$ , we find that

$$P(k) = \frac{L(k)}{1 + 2V_{12,21}(0) \sum_{k \leq k_F} L(k)}. \quad (15)$$

Therefore, we only need to solve the integral equation (14) for  $L(k)$  and obtain  $P(k)$  from (15), instead of solving two integral equations. We note that the above equations have also been considered in Ref. 12, where an approximate analytical solution has been given.

For numerical illustration, we consider a 250-Å  $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$  quantum well with one-side delta doping and a spacer layer of 250 Å. We assume  $x = 0.3$  and  $\Delta E_c / \Delta E_v = 67/33$ , where  $\Delta E_c$  ( $\Delta E_v$ ) is the conduction-band (valence-band) band-gap offset energy. The Hartree energies for the first two subbands are compared with the Hartree-Fock energies (6) and (7) near the subband edge  $k = 0$  as functions of the two-dimensional carrier density  $N_s$ , in Fig. 1(a). The corresponding separations between the first two subbands at the band edge ( $k = 0$ ) and at the Fermi edge ( $k = k_F$ ) are shown in Fig.

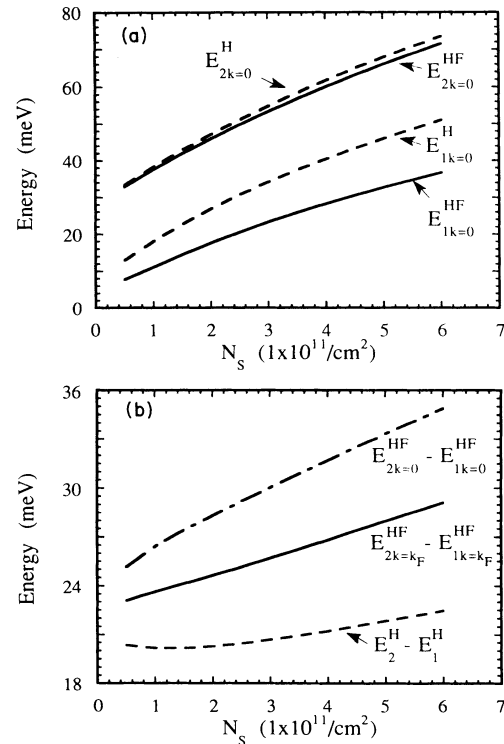


FIG. 1. (a) Energies of the first two subbands at the band edge ( $k = 0$ ) as functions of the two-dimensional carrier density  $N_s$ . The solid lines are the Hartree-Fock energies and the dashed lines are the Hartree energies. (b) The corresponding intersubband energy spacings at the Fermi edge ( $k = k_F$ ) and at  $k = 0$ . (a) and (b) are calculated for a 250-Å  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}/\text{GaAs}$  quantum well with one-side delta doping and a spacer layer of 250 Å.

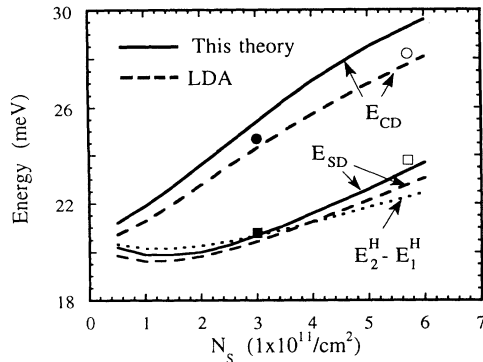


FIG. 2. Theoretical peak positions using this theory (solid lines) and the LDA (dashed lines) in charge-density ( $E_{CD}$ ) and spin-density ( $E_{SD}$ ) excitation spectra. The parameters are the same as in Fig. 1. The circles (CD) and squares (SD) are experimental data. The intersubband Hartree energy is shown as the dotted line.

1 (b). The big increase in energy separation due to the exchange interaction is in qualitative agreement with the calculation of Bandara *et al.*,<sup>3</sup> except that they use wave functions for an undoped infinite well, while we use wave functions from a self-consistent solution which takes into account the finite well depth and space-charge effects. The increase in subband energy separation can be understood from the fact that exchange is most effective for the occupied  $n=1$  states [Fig. 1 (a)].

In Fig. 2 we show the theoretical results (solid lines) for the peak positions in charge-density and spin-density excitation spectra as functions of the two-dimensional carrier density  $N_s$ . The parameters are the same as those in Figs. 1 (a) and 1 (b). The intersubband Hartree energy is shown as the dotted line for illustration. We also calculated the peak energies using the LDA method (dashed lines),<sup>4,5</sup> with the potential form of Gunnarsson and Lundqvist.<sup>13</sup> The circles and squares are experimental data. The solid symbols were measured on a sample with a well width of 250 Å, a barrier aluminum mole fraction of  $x=0.3$ , and a 500-Å spacer layer. (The precise value of the width of the spacer layer is unimportant for all samples considered in this paper.) The open symbols were measured on another sample with the same well width,  $x=0.33$ , and a 250-Å spacer layer. The agreement between our theory, the LDA, and experiment is very good. One qualitative difference (the most important one) between our theory and LDA is that LDA does not distinguish between occupied and unoccupied states, so that the renormalization of the second subband is overestimated, leading to a smaller subband spacing. Note that our theory does not require any parametrization. The small difference in the aluminum mole fraction ( $x=0.33$  compared with  $x=0.3$ ) again makes little difference in the theoretical calculations. In general, the separation between the energies  $E_{CD}$  and  $E_{SD}$  increases with carrier concentration. The curves of  $E_{SD}$  and intersubband Hartree energy indicate that the excitonic binding tends to cancel the corrections due to exchange self-energies.

In Figs. 3(a)–3(c), we show calculated light scattering

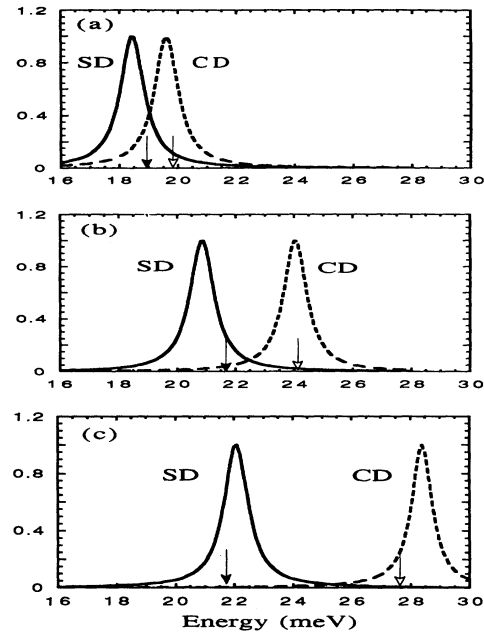


FIG. 3. Light scattering spectra for charge-density (CD) and spin-density (SD) excitations in three different samples. The measured peak positions are shown as vertical arrows. The barrier Al mole fraction ( $x$ ), carrier density ( $N_s$ ), and well width ( $L_w$ ) for the three samples are as follows: (a)  $x=0.15$ ,  $N_s=0.55 \times 10^{11}/\text{cm}^2$ ,  $L_w=250$  Å; (b)  $x=0.24$ ,  $N_s=1.8 \times 10^{11}/\text{cm}^2$ ,  $L_w=240$  Å; (c)  $x=0.18$ ,  $N_s=4.2 \times 10^{11}/\text{cm}^2$ ,  $L_w=204$  Å.

intensities for charge- and spin-density excitations in another three samples. Sample (a) has a barrier aluminum mole fraction  $x=0.15$ , a carrier concentration  $N_s=0.55 \times 10^{11}/\text{cm}^2$ , and a well width  $L_w=250$  Å. All these numbers were determined experimentally. Sample (b) has  $x$  determined to be between 0.21 and 0.24,  $N_s$  measured to be  $1.8 \times 10^{11}/\text{cm}^2$ , and the designed well width is 250 Å. We take  $x=0.24$ ,  $L_w=240$  Å, and the same carrier concentration to match the data. Sample (c) has two-sided symmetric doping instead and the data are from Ref. 14. The parameters are  $x=0.18$ ,  $L_w=204$  Å, and  $N_s=4.2 \times 10^{11}/\text{cm}^2$ . The same parameters are used in the calculation. The experimental data for the peak energies are shown as vertical arrows. In general, the agreement between our theory and experiment is very good. The experimental data of sample (c) have also been compared favorably with LDA theory by Katayama and Ando.<sup>8</sup>

In conclusion, we have presented a variational theory within the Hartree-Fock approximation to study many-body effects on intersubband transitions. Our theory agrees very well with the measured charge- and spin-density excitations in inelastic light scattering experiments. It also agrees with the results of LDA. For intersubband optical absorption, exchange self-energy effects have been shown previously to be important.<sup>3</sup> However, depolarization and excitonic effects were not considered in that work. We include all these effects in this paper; hence, the charge polarizability presented here should also be useful to interpret infrared optical absorption data.

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