Influence of nonparabolicity on collective intersubband spin- and charge-density excitation spectra

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The influence of the conduction-band nonparabolicity on the collective spin and charge-density excitation spectra in $GaAs/Al_xGa_{1-x}As$ quantum wells is discussed theoretically in the framework of the local-density approximation. Emphasis is placed on estimating the importance of band population effects at finite temperature. A line-shape calculation shows that the nonparabolicity shifts resonance energies and leads to the additional broadening of the excitation spectra. Qualitatively different behavior of the linewidth (as a function of the electron concentration) is found for the charge- and spin-density excitations.

Intersubband spin-density excitations (SDE's) in quantum-well systems exist through the exchange-correlation Coulomb interaction in a way similar to that by which intersubband charge-density excitations (CDE's) exist through the direct Coulomb interaction. The CDE's are observable by far-infrared absorption and Raman-scattering measurements, while the SDE's have only been observed in Raman-scattering measurements.

The intersubband excitation spectra have usually been described within a self-consistent field theory, where the exchange and correlation Coulomb interactions are included through the local-density approximation (LDA). Recent works have shown that there is very good agreement between this theory and inelastic light measurements.[1,2] LDA calculations also agree with the results obtained in the variational Hartree-Fock approximation.[3]

In most of the theoretical papers devoted to the CDE's and SDE's the case of parabolic subbands is considered. It is well known that the original nonparabolicity of the bulk bands leads to nonparallel dispersions of the upper and lower subbands in the well. The excited-state subband is flatter than the ground-state subband and, consequently, the subband separation depends on $\mathbf{k}_t$ (the two-dimensional in-plane wave vector). Results reported by Brozak *et al.*[4,5] (see also Ref. 6) suggest that inclusion of this dependence is essential to the accurate description of the intersubband excitation spectra in relatively thin quantum wells (QW's). However, the authors of Refs. 4 and 5 restricted the calculations of the excitation energies to $T=0$ K, neglecting, in addition, the homogeneous broadening of the levels. When the temperature is raised, electrons populate higher-lying states on the ground subband and, because the upper subband has a lower curvature, the average intersubband separation decreases (compared to the $T=0$ case). Consequently, we can expect that effects of nonparabolicity should be more pronounced at a finite temperature.[6]

The purpose of this paper is to discuss, within the LDA, the influence of the nonparabolicity on collective CDE and SDE spectra in GaAs quantum-well structures taking into account the line broadening induced by electron scattering and giving a special emphasis to the effects connected with the change of the thermical population of the ground-subband states with increasing T. We show that the contribution of the nonparabolicity to the linewidth depends not only on the sheet electron concentration and temperature but also on the type of the excitation. The theory predicts that, at room temperature, the nonparabolicity-induced contribution to the charge density (CD) linewidth strongly decreases with increasing the electron concentration, while in the case of the single-particles (SP) and spin-density (SD) excitations, the situation is opposite.

The above property of the CDE is very important from a QW device physics point of view because the transition linewidth determines both the selectivity and efficiency of the photodetectors, or, even more so, the intensity of nonlinear optical processes.[7]

Theoretically, the collective excitation spectra are given by the imaginary part of the density-density correlation functions, $\chi_i(\mathbf{q},\omega)$,

$$I_i(\mathbf{q},\omega) \propto -\mathrm{Im}\chi_i(\mathbf{q},\omega) , \tag{1}$$

where the subscript refers to the CD or SD response and $\mathbf{q}$ is the wave-vector exchange. In further calculations we restrict for simplicity to the case when $\mathbf{q}\rightarrow\mathbf{0}$.

Let us neglect for the moment the nonparabolicity of the bulk crystal. Using the time-dependent LDA and taking into account only two subbands, we get the following expression for the response function:[1,4,5]

$$\chi_i(\mathbf{q},\omega)=\chi_0(\mathbf{q},\omega)/[1-\gamma_i\chi_0(\mathbf{q},\omega)] . \tag{2}$$

Here $\chi_0(\mathbf{q},\omega)$ is the intersubband susceptibility in the absence of Coulomb interactions. For $\mathbf{q}\rightarrow\mathbf{0}$ it is given by

$$\chi_0(0,\omega)=-4\sum_{\mathbf{k}_t} F(E_0(\mathbf{k}_t))E_{10}(\mathbf{k}_t)/[E_{10}^2(\mathbf{k}_t)-(\hbar\omega)^2 - i\hbar\omega\Gamma] , \tag{3}$$

where $E_{10}(\mathbf{k}_t)=E_1(\mathbf{k}_t)-E_0(\mathbf{k}_t)$, $E_j(\mathbf{k}_t)$ is the single-particle energy in the jth subband calculated in the LDA, Γ is a phenomenological parameter simulating the collision broadening of the line, and $F(E)$ is the Fermi-Dirac distribution function. The Fermi energy appearing in this function can be obtained from the normalization conduction $2\sum_{\mathbf{k}_t}F(E_0(\mathbf{k}_t))=N_s$, where N_s is the surface density of the electrons. (As in previous papers, we assume that only ground subband $j=0$ is occupied.) The Coulomb interaction [in Eq. (2)] is written in terms of the direct term α_{CD} and exchange-correlation terms β_i

$$\gamma_{CD}=\alpha_{CD}-\beta_{CD} \tag{4}$$

and

$$\gamma_{SD}=-\beta_{SD} \ . \tag{5}$$

In the case of the single quantum well

$$\alpha_{CD}=4\pi e^2\epsilon_\infty^{-1}\int_{-\infty}^{\infty}dz\left[\int_{-\infty}^{z}dz'\varphi_0(z')\varphi_1(z')\right]^2 \tag{6}$$

and

$$\beta_i=-\int_{-\infty}^{\infty}dz\ U_i(z)\varphi_0^2(z)\varphi_1^2(z) \ , \tag{7}$$

with[1]

$$U_{CD}=(-1.706a_B^3r_s^2)[1+0.6213r_s/(11.4+r_s)](R_{GaAs}) \ , \tag{8}$$

$$U_{SD}=(-1.706a_B^3r_s^2)[1+1.36r_s/(1+10r_s)](R_{GaAs}) \ , \tag{9}$$

where $r_s(z)=[4\pi a_B^3 n(z)/3]^{-1/3}$, $n(z)=N_s\varphi_0^2(z)$, $\varphi_j(z)$ is the wave function for the jth subband, a_B is the effective Bohr radius in GaAs, R_{GaAs} is the effective Rydberg for GaAs, and E_{10} is the k_t independent intersubband energy. The first term in Eqs. (8) and (9) is identical and is connected with the exchange interaction. The remaining terms are a consequence of electron correlations.

From Eqs. (1) and (2) we find that when the nonparabolicity is ignored, the collective excitation spectrum has Lorentzian shape with the peak position at

$$E_i=E_{10}(1+\gamma_i 2N_s/E_{10})^{1/2}=E_{10}(1+\tilde{\gamma}_i)^{1/2} \ . \tag{10}$$

The exact calculation of the excitation spectra in the case when the nonparabolicity is not negligible is a more difficult task. However, the aim of our work is not obtaining the full form of the density-density correlation function in the multiband envelope-function approximation and subsequent numerical calculation of the excitation spectra in conjunction with the ground-state properties in a full self-consistent procedure. What we want to do is to discuss the influence of the nonparallel dispersion of the upper and lower subbands on the CD and SD excitation spectra (the peak position and linewidth) at finite T and Γ. Therefore, we make a number of simplifying approximations. However, none of them affects our results in any essential way. In the GaAs/Al_xGa_{1-x}As QW's, where the intersubband resonances have been observed, the deviation from the parabolic model is not very significant. Thus, in the first approximation, we can use Eqs. (1)–(9). However, $E_{10}(\mathbf{k}_t)$ (and Fermi energy) appearing in the expression for $\chi_0(0,\omega)$ should be calculated to include the nonparabolicity. (The effect of the nonparabolicity on the parameters γ_{SD} and γ_{CD} is neglected in our approximation.)

We calculate the in-plane subband dispersion along the line of our[6] implementation of the empirical two-band model proposed in Refs. 8 and 9 ignoring for simplicity the temperature dependence of the band parameters and neglecting band-bending effects. In the narrow QW's (where the nonparabolicity can be substantial) the relative correction to the intersubband energy $E_{10}(0)$ resulting from the band-bending effect is rather small, $<3-4\%$.[10,11] We will also ignore (calculating γ_i) the electron correlation effects, which are negligible compared to the exchange effects provided that the electron concentration is not too small. We assume also that the temperature dependence of the exchange effects can be neglected.[10]

We have performed the numerical calculation of the subband energies for GaAs/$Al_{0.35}Ga_{0.65}$As QW's with the technically interesting thickness 75 Å taking the band parameters the same as in Ref. 6. (We want to note that such types of QW's were studied experimentally in many papers.[11,12]) The dependence of the subband separation on $\mathbf{k}_t$ resulting from our calculations (for details see Ref. 6) is consistent with that obtained with the help of the more sophisticated models.[12] The nonparallel dispersions of the subbands in the QW can be described introducing the effective mass that is not only different for the ground and excited subbands but also varies (though rather weakly) with increasing k_t (see Fig. 1 in Ref. 6). At $T=0$ only the states with $k_t<k_F$ (the wave vector at Fermi energy) are occupied. Thus, when the electron concentration is not too large, the above-mentioned variation can be neglected and the approximate value for the in-plane masses of the subband can be determined from a least-square fit of the energy dispersion from $k_t=0$ to k_F onto a parabola.[4,5] Due to this approximation it is possible to obtain the analytical expression for the energies of the collective excitations [the poles of the electron response functions $\chi_i(0,\omega)$ at $\Gamma=0$] (Refs. 4 and 5)

$$E_i^2=E_{10}^2(0)[\zeta^2\exp(\vartheta_i)-\exp(-\vartheta_i)]/2\sinh(\vartheta_i) \ , \tag{11}$$

where $\zeta=E_{10}(k_F)/E_{10}(0)$ and $\vartheta_i=(\zeta-1)/\tilde{\gamma}_i$.

The variation of E_{SD} and E_{CD} with N_s in 75-Å GaAs/$Al_{0.35}Ga_{0.65}$As QW's is shown in Fig. 1. The resonant energies calculated from Eq. (10) [but with E_{10} replaced by $E_{10}(0)$ resulting from the two-band model] are also plotted for comparison. We see that the dependence of the subband separation on k_t leads to the downward shift of the collective excitation energies with increasing N_s. The difference $\Delta E=E_{CD}-E_{SD}$ increases nearly linearly with increasing N_s and is rather weakly affected

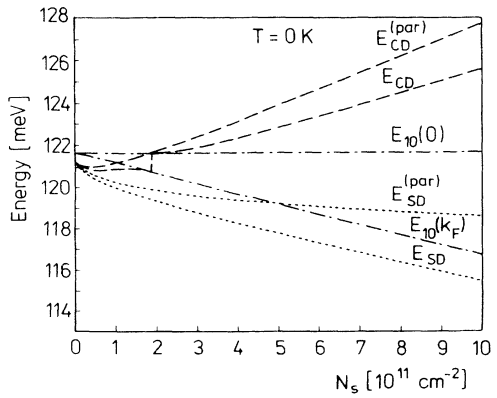

FIG. 1. Dependence of the resonant energies E_{CD} and E_{SD} on the electron density in 75-Å GaAs/$Al_{0.35}Ga_{0.65}$As QW's. For comparison we show also the resonant energies $E_{CD}^{(par)}$ and $E_{SD}^{(par)}$ calculated assuming that $E_{10}(k_t)=E_{10}(0)$.

by the nonparallel dispersion of the subbands. The numerical values of ΔE are in the reasonable agreement with that observed in Raman scattering by Ramsteiner *et al.*[13] (see also Ref. 12).

Some comment should be made on the steplike structure observed in Fig. 1 at $N_s \approx 2\times 10^{11}$ cm^{-2}. Detailed inspection of Eq. (11) leads us to conclude that, in the absence of the collision ($\Gamma=0$), the energy of the collective intersubband excitation E_i is always larger than $E_{10}(0)$ (if $\tilde{\gamma}_i > 0$) or smaller than $E_{10}(k_F)$ (if $\tilde{\gamma}_i < 0$), i.e., the collective intersubband modes are above or below the SP intersubband continuum. (So the collective modes are free of Landau damping.) In the model used here we have $\tilde{\alpha}_{CD} \equiv \alpha_{CD} 2N_s/E_{10}(0) \sim N_s$ and $\tilde{\beta}_{CD} \equiv \beta_{CD} 2N_s/E_{10}(0) \sim N_s^{1/3}$. Consequently, at sufficiently small N_s, quantity $\tilde{\beta}_{CD}$ exceeds $\tilde{\alpha}_{CD}$ resulting in the shift CD mode and appearance of the steplike structure at N_s satisfying the relation $\tilde{\alpha}_{CD}(N_s)=\tilde{\beta}_{CD}(N_s)$. The wider the well, the lower the electron density, for which this effect occurs. It should be noted that the above-mentioned structure does not occur when the homogeneous line broadening is included (see Fig. 2).

Now we discuss the line shapes of the collective and single-particle ($\gamma_i=0$) excitations [calculated with the help of Eqs. (1)–(9)] for the previously specified quantum well when T and Γ are finite. We have performed calculations for $T=0$ and 300 K. The homogeneous line broadening Γ was taken to be 2.5 and 7.5 meV. At this point we should note that the total linewidth comes from the nonparabolicity of the subbands and the homogeneous broadening of the level which, in general, depends on the temperature, electron concentration, and the quality of the structure. In the case of GaAs/Ga_xAl_{1-x}As QW's with thickness $\lesssim 100$ Å, the experimentally determined linewidths are larger than 4–5 meV.[11,12,14] Thus the value $\Gamma=2.5$ meV is rather underestimated even for a high-quality sample.[11]

Plots of the peak positions and the linewidth versus the electron concentration are given in Figs. 2 and 3, respectively. We see that with increasing T the excitation spectrum becomes broader and its maximum shifts towards lower energy. (We should remember that in our model the temperature variation of the line shape is caused only by population effect.) In the sample considered here the increase of T from 0 to 300 K leads to a rather small shift of the peak (1–2 meV). This shift is not very sensitive to the electron concentration and the homogeneous level broadening.

Much more interesting is the dependence of the nonparabolicity correction to the FWHM (full width at half-maximum) on N_s. Figure 3 shows that this correction is very sensitive to the type of the excitation. The behavior of the FWHM for the SP and SD excitations is qualitatively similar. With increasing N_s the nonparabolicity-induced correction increases. At $T \approx 0$ this correction is significant only in the range of the high electron concentration ($\gtrsim 10^{12}$ cm^{-2}). When $T=300$ K the nonparabolicity gives substantial contribution (a few meV) to the linewidth even when N_s is small. The situation is quite different in the case of the CDE. Figure 3 shows that, in contrast with SPE and SDE, the increase of N_s leads not to an increase but to a decrease of the nonparabolicity-

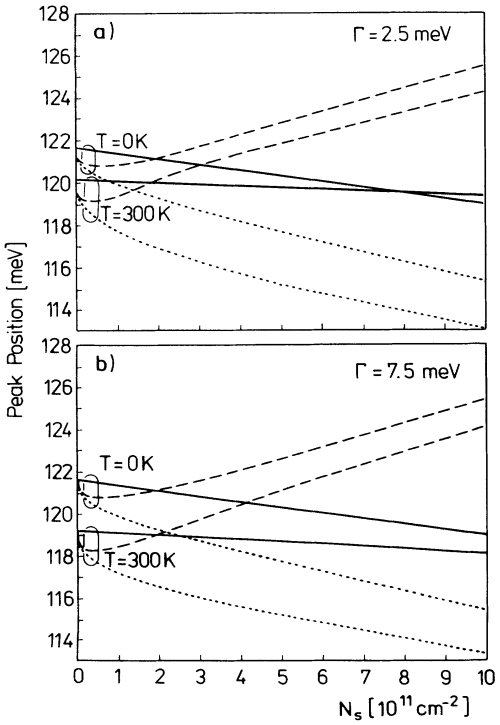

FIG. 2. Dependence of the peak position on the electron density in 75-Å GaAs/$Al_{0.35}Ga_{0.65}$As QW's with (a) $\Gamma=2.5$ meV and (b) $\Gamma=7.5$ meV at temperatures $T=0$ and 300 K. (——) SPE, (— — —) CDE, and (- - -) SDE.

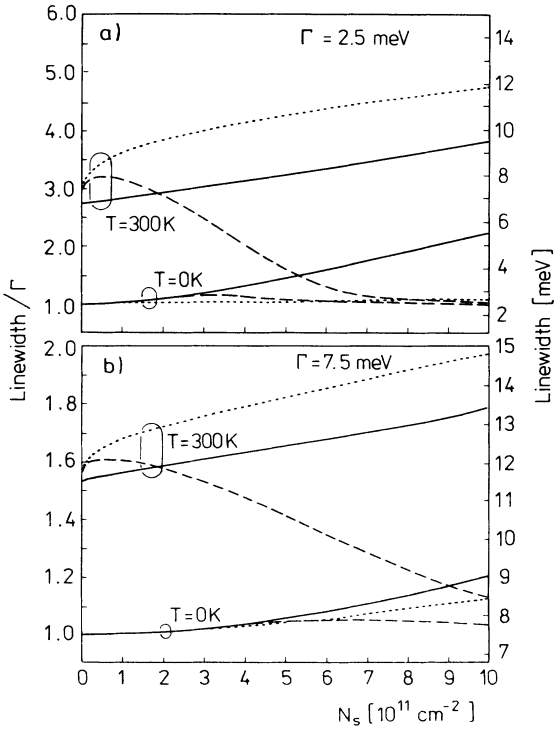

FIG. 3. Dependence of the linewidth on the electron density in 75-Å GaAs/$Al_{0.35}Ga_{0.65}$As QW's with (a) $\Gamma=2.5$ meV and (b) $\Gamma=7.5$ meV at temperatures $T=0$ and 300 K. (——) SPE, (— — —) CDE, and (- - -) SDE.

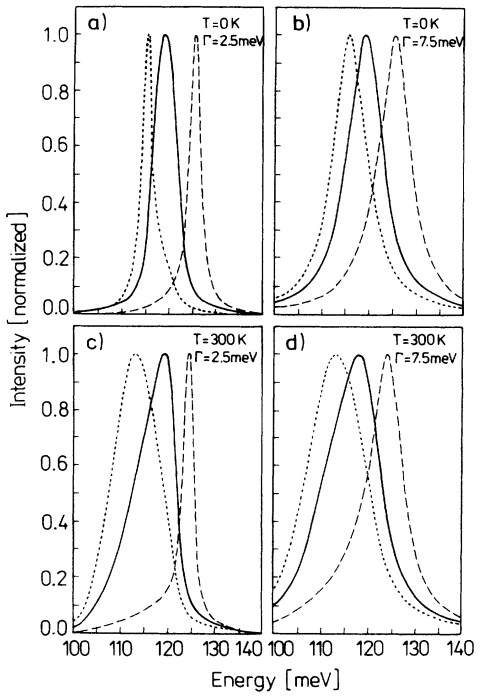

FIG. 4. Calculated single-particle and collective excitation spectra in 75-Å $GaAs/Al_{0.35}Ga_{0.65}As$ QW's with $N_s=10^{12}$ cm^{-2}, $\Gamma=2.5$, and 7.5 meV at temperatures $T=0$ and 300 K. (——) SPE, (— — —) CDE, and (- - -) SDE.

induced correction to the CD linewidth. This effect is particularly strong at room temperature. In highly doped systems ($N_s \geq 10^{12}$ cm^{-2}) the total linewidth is then practically determined only by the level broadening. Note that at $T \sim 0$ the reduction of the nonparabolicity correction to the CD linewidth (with increasing N_s) is observed for $N_s \geq 5\times 10^{11}$ cm^{-2}. In the range of the high electron concentration the correction to the CD linewidth is smaller than the correction to the SP (or SD) linewidth. The difference between the CDE and the SDE is especially large in high-quality barrier-doped structures where Γ is small ($\sim 4-5$ meV). Thus, if we fit (as was done in previous papers) the experimental intersubband absorption line shape to the theoretical SP line shape we get the value of Γ substantially smaller than that obtained from the fitting to the theoretical CD line shape. Taking, for example, $T=0$, $N_s=10^{12}$ cm^{-2}, and assuming that the experimental linewidth is 5.6 meV, we find that the fit to the theoretical SP spectrum gives $\Gamma \simeq 2.5$ meV, while the fit to the theoretical CD spectrum gives $\Gamma \simeq 5.5$ meV. We believe that this is the main reason why the values of the parameter Γ deduced by von Allmen *et al.*[12] from the line-shape analysis of the barrier-doped QW's are unusually small. Therefore, an estimate of the contribution of level broadening to the total linewidth performed in previous papers[12,14] should be revised.

Figure 4 shows the SD, CD, and SP excitation spectra calculated for the technically interesting value of the electron concentration $N_s=10^{12}$ cm^{-2}. At room temperature our model predicts a strongly asymmetric line shape for SP excitations (especially at smaller values of Γ). In the case of the collective excitations the asymmetry is rather negligible. The CD excitation spectrum has a nearly Lorentzian shape. It is confirmed very well by experiment.[14] Calculations show that the deviation from the Lorentzian shape can be substantial when the temperature is high and the electron concentration small, i.e., when the nonparabolicity-induced correction to the CD linewidth is largest.

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