RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 45, NUMBER 19 15 MAY 1992-I

Effects of nonparabolicity on collective intersubband excitations

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(Received 26 November 1991; revised manuscript received 21 January 1992)

We have used inelastic light scattering to study the collective charge- and spin-density excitations associated with electronic intersubband transitions in GaSb-AlSb superlattices. The density of the photoexcited two-dimensional electron-hole plasma was controlled by varying the incident laser power. A comparison of experiment with detailed calculations of the response functions shows that the nonparabolicity of the conduction band must be properly included in order to yield quantitative agreement. It is found that nonparabolicity, in addition to decreasing the resonance energies of the collective excitations, provides a mechanism for observing single-particle excitations at $q = 0$.

The two-dimensional electron gas (2DEG) has received an extraordinary amount of attention from semiconductor physicists for many years.[1] This is somewhat because of its technological importance in electronic devices but also due to the fact that it is an ideal system in which fundamental concepts of physics can be tested experimentally. In particular, important insights into electron-electron correlations have been obtained by optical studies of the elementary excitation spectrum of the 2DEG. Far-infrared absorption[2] and inelastic light scattering experiments[3] have both yielded information about collective excitations of the charge density (e.g., intersubband plasmons) which exist through the *direct Coulomb interaction*. In addition, collective excitations of the spin density,[4,5] which are manifestations of the *exchange and correlation Coulomb interaction*, are observable via inelastic light-scattering measurements. Although some of the early work concerning the *collective* charge-density excitations (CDE's) was performed using 2DEG's formed at the oxide-semiconductor interface in Si field-effect transistors,[1] much of the more recent work, particularly concerning the *collective* spin-density excitations (SDE's) has been performed in $GaAs/Al_xGa_{1-x}As$ heterostructures.[4,5] The interacting 2DEG in these systems has usually been described within a self-consistent field theory,[6,7] where the exchange and correlation Coulomb interactions are included through the local-density approximation (LDA).[8,9] It is found that, as a function of increasing density and for light scattering with zero momentum transfer (i.e., at $q \approx 0$), the energy of the CDE is depolarization-shifted to higher energy relative to the "bare" intersubband transition, while the energy of the SDE is shifted down by the exchange interaction. A comparison of the peak energies associated with the calculated response functions and experiment typically leads to good agreement.[10]

Recently, there has been some interest in using our detailed understanding of these collective excitations to probe the internal piezoelectric fields which are generated by misfit strains in (111) GaSb/AlSb strained-layer superlattices.[11,12] The GaSb/AlSb system is, however, slightly different than the $GaAs/Al_xGa_{1-x}As$ system in that the narrower band gap of GaSb leads to a more pronounced conduction-band nonparabolicity. This is expected to affect both the character and energies of the collective excitations. In this paper we present an experimental study of the collective charge and spin-density excitations in (100) GaSb/AlSb strained-layer superlattice samples. Unlike the (111) case, there are no piezoelectric fields in samples oriented along the (100) direction. A detailed comparison between self-consistent LDA calculations of the response functions and experiment shows that the nonparabolicity of the conduction band must be explicitly included in order to obtain good agreement. In addition, the theory predicts that nonparabolicity provides a new mechanism by which single-particle excitations can be observed at $q = 0$.

The three samples used in this study were grown by molecular-beam epitaxy on (100) GaAs substrates and consisted of a ~ 1-μm AlSb buffer layer followed by 20 periods of a GaSb/AlSb superlattice and a GaSb cap layer. All layers were unintentionally doped p-type in the $\sim 10^{15}$-cm^{-3} range. The samples were characterized by x-ray diffraction measurements which were performed on a powder diffractometer using reflections from (004) planes. A least-squares-fitting algorithm was used to fit a kinematic simulation of the diffraction process to the x-ray data. This allowed us to estimate the individual GaSb well (L_w) and AlSb barrier (L_b) thicknesses as well as the strains in the respective layers. These results are summarized in Table I. In these simulations we assumed coherent growth throughout the superlattice so that the

TABLE I. Sample parameters as measured by x-ray diffraction. The z axis is along the growth direction of the samples.

	GaSb			AlSb	
L_w (Å)	ϵ_{zz}	$\epsilon_{\parallel}$	L_b (Å)	ϵ_{zz}	$\epsilon_{\parallel}$
125	−0.0036	0.0039	163.0	0.0025	−0.0025
176	−0.0028	0.0031	160.1	0.0033	−0.0034
249	−0.0026	0.0029	234.0	0.0036	−0.0036

in-plane lattice constants in the GaSb and AlSb layers were the same. In addition, well and barrier thicknesses were fixed to integral numbers of monolayers and the interfaces were assumed to be perfectly abrupt and smooth. Although this is clearly an oversimplification, it is estimated that the individual layer thicknesses can be determined to within approximately ±2 monolayers, whereas the superlattice period is known to ±1 monolayer. The strains which result from the fitting procedure are estimated to be accurate to within approximately 10% or 15%. It should also be noted that the measured barrier widths are such that the superlattice is weakly coupled (i.e., the electron wave functions from adjacent wells do not overlap significantly) such that the samples are more correctly characterized as a set of isolated multiple quantum wells.

The Raman scattering measurements were performed near resonance with the GaSb quantum well $E_0 + \Delta_0$ band gap employing a Ti:sapphire laser pumped by an argon-ion laser. The scattered photons were dispersed with a Dilor triple monochromator and detected with an EG&G liquid-nitrogen-cooled CCD detector. All measurements were performed in the backscattering geometry so that wave-vector transfer was negligible (i.e., $q \approx 0$). The incident laser light is also used to photoexcite carriers in the quantum wells resulting in a two-component plasma with approximately equal densities of electrons and holes. These densities are controlled by varying the power incident on the sample. The density of carriers is determined *in situ* from the energy differences between the CDE and SDE.[4] The temperature of the sample was nominally 5 K with considerable local heating occurring at the highest laser powers. From the ratio of the Stokes to anti-Stokes phonon lines the local temperature at the highest powers was determined to be ~ 75 K. The effects of this higher temperature on the energy gaps and effective masses is very small and is not included in the calculations.

In Fig. 1 we show spectra for both the CDE and SDE in the 176-Å sample. The CDE are measured in the polarized Raman scattering geometry, where the polarizations of the incident and detected radiation are parallel, whereas the SDE are detected in the depolarized or crossed geometry.[3] At the lowest laser powers, where the plasma density is estimated to be ~ 10^{10} cm^{-2}, the polarized and depolarized resonances both occur at the same, single-particle or bare intersubband energy. As the plasma density is increased, the collective CDE and SDE split apart in energy. The CDE is shifted to higher energy by the net effects of the direct and exchange Coulomb interactions and nonparabolicity, while the SDE is shifted to lower energy by a combination of the exchange-correlation interaction and nonparabolicity.

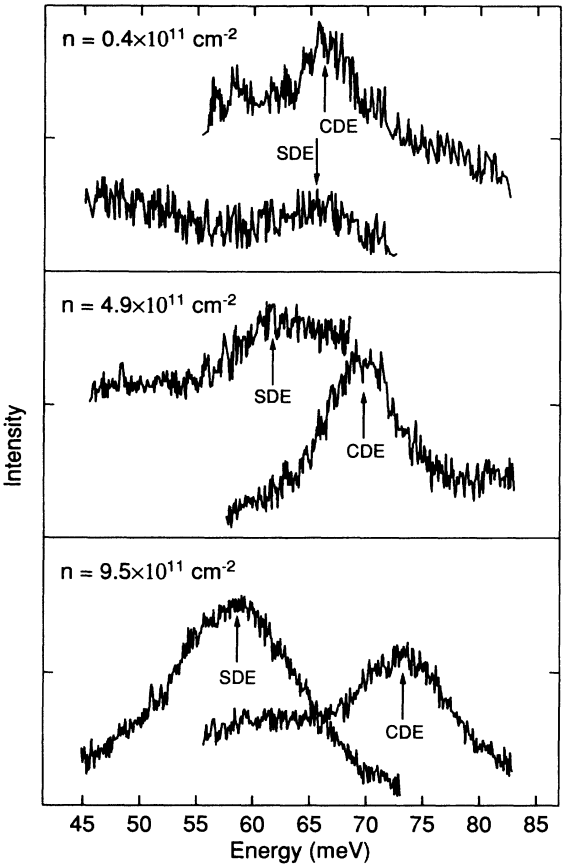

FIG. 1. Raman spectra from the collective charge and spin-density excitations in the 176-Å sample.

The collective excitations of the two-component electron-hole plasma are calculated within a time-dependent local-density approximation. The Schrödinger equation for electrons is solved self-consistently with the Poisson equation where the potentials due to both the photoexcited electrons and holes are taken into account. Because the envelope functions of the electrons and holes are essentially identical the Hartree term is insignificant. The response function of the electron gas is then calculated using the resulting electron wave functions and energy levels. In principle, the response functions should be calculated by including contributions from both the electron and hole collective excitations as well as their interaction terms. The interaction between electron and hole intersubband excitations would only be significant if their energies were comparable which would occur if, for example, the barrier heights and masses corresponding to the electrons and holes were the same. We have calculated the full response functions for the case of a parabolic band structure and found that the effects of including the hole states are small and that it is thus appropriate to include only the electron states in what follows.

The collective excitations of the 2DEG are obtained from the poles of the electron response function,

$$\chi_i(q,\omega) = \frac{\chi_0(q,\omega)}{1+\gamma_i(q)\chi_0(q,\omega)}\ , \tag{1}$$

where $i \equiv$ CDE or SDE. Here $\gamma_{\rm CDE} = \alpha_{\rm CDE} - \beta_{\rm CDE}$, and $\gamma_{\rm SDE} = -\beta_{\rm SDE}$, where the direct Coulomb interaction integral $\alpha_{\rm CDE}$ and exchange-correlation interaction integrals β_i are defined as in Ref. 5. The response function in the absence of interactions is given by $\chi_0(q,\omega) = D_{10}(q,\omega) + D_{01}(q,\omega)$, where

$$D_{ij}(q,\omega) = 2\sum_{k=0}^{\infty} \frac{f_j(E_j) - f_i(E_i)}{E_j(k+q) - E_i(k) - \hbar(\omega + i\Gamma)}. \tag{2}$$

Here $f(E)$ is the Fermi-Dirac distribution function and Γ is a phenomenological broadening parameter. The subband dispersion in a quantum well is approximated by

$$E_i(k) = E_i + \frac{\hbar^2 k^2}{2m_i^*(k)}, \tag{3}$$

where E_i is the energy of the ith subband and k lies in the plane of the quantum well. If the nonparabolicity of the conduction band of GaSb is ignored, then the masses in the ground ($i = 0$) and first excited ($i = 1$) subbands are identical and a calculation of the response functions as a function of plasma density yields the dashed curves shown in Fig. 2. The symbols in Fig. 2 represent the peaks of the observed collective excitations (e.g., see Fig. 1). The densities at which these peaks are plotted were determined by comparing the observed energy separation between the CDE and SDE to the separation in the calculated curves. It is seen that the SDE slopes down in energy with increasing density. What is striking is the large magnitude of the slope which is not observed in systems with a more parabolic band structure such as GaAs/$Al_xGa_{1-x}As$.[4,5] This large slope is attributed as being due to nonparabolicity as described below. It is also seen that the slope increases as the well width decreases. This is further evidence that nonparabolicity is responsible since it becomes more pronounced at smaller well widths.

The nonparabolicity of the GaSb conduction band can be included in the response functions by allowing the masses in Eq. (3) to depend on k. The mass thus not only varies from subband to subband but also varies within a subband with increasing in-plane wave vector. We calculate the in-plane subband dispersion along the lines of Załużny's[13] implementation of the empirical two-band model proposed by Nelson and co-workers.[14,15] In order to facilitate a closed form calculation of the sum in Eq. (2), approximate values for the in-plane masses of the subbands are determined from a least-squares fit of the energy dispersion from $k = 0$ to $k = k_F$ onto a parabola. Here k_F is the wave vector at the Fermi energy. The energies of the collective excitations calculated in this way are plotted as the solid curves in Fig. 2.

The decrease in energy of the SDE with increasing density is significantly underestimated by the dashed curves, where the small downward shift away from the bare intersubband transition is due to the exchange and correlation Coulomb interaction. At the same time, the upward shift of the CDE is seriously overestimated by the parabolic calculations. It is clear that the solid curves, where nonparabolicity is explicitly included, give much better agreement with experiment. Despite the excellent agreement exhibited in Fig. 2, particularly in the 176- and 125-Å samples, it is important to stress that the absolute energy of the bare intersubband transition cannot be taken too seriously. The estimated error in our knowledge of the quantum well widths (i.e., ±2 monolayers) does not allow us to believe the results of the calculation of the intersubband energy to better than several meV. For example, in the 125-Å sample, which is most sensitive to small changes in well width, the shift of the intersubband energy corresponding to a change in the well width of ±2 monolayers is approximately 8 meV. The dependence of the collective excitations on density is, however, quite independent of the well width. If we calculate the curves corresponding to the CDE and SDE for a well width of 118 Å (a decrease of 2 monolayers) we obtain curves which are nearly identical to those shown in Fig. 2 but translated *rigidly* upwards in energy by ~ 8 meV.

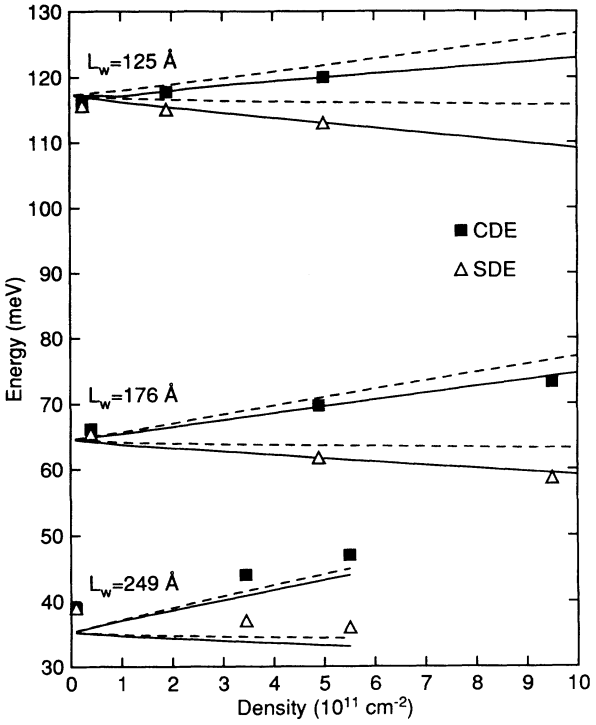

FIG. 2. Comparison between experiment and calculations ignoring (dashed curves) and including (solid curves) the nonparabolicity of the GaSb band structure. The densities corresponding to the experimental points were determined by comparing the experimental energy separation of the measured CDE and SDE to the solid curves although essentially the same density would be extracted from the dashed curves.

In order to understand the downward shift of the SDE in nonparabolic band structures, consider Fig. 3. We show the in-plane dispersion of the lowest two subbands, where the mass of the first excited subband is somewhat larger due to nonparabolicity. The single-particle excitations form a band of states consisting of all the vertical (i.e., $q \approx 0$) transitions between $k = 0$ and $k = k_F$. As

the density (and hence k_F) increases, *this band of single-particle excitations widens toward lower energy.* If we calculate the response function for the system we find that the peak corresponding to the collective spin density excitation always occurs at the lower energy edge of the single-particle band. In essence, the SDE is pushed down in energy by the effects of nonparabolicity. These results are also of interest in that they lead to a new way of observing the single-particle band. In a parabolic band structure the vertical ($q = 0$) single-particle transitions for all k occur at the bare intersubband energy (E_{01}) so that the single-particle band reduces to a delta function. This single-particle response can be shown to vanish and the single-particle excitations are only observable for light scattering with significant wave-vector transfer into the plane of the quantum well.[16] In a nonparabolic system the finite width of the single-particle band leads to a finite response at $q = 0$ so that it is now, in principle, possible to observe the single-particle band in the backscattering geometry. If this single-particle band were to be observed in the measurements described above, it would show up as a weak shoulder to the higher (lower) energy side of the SDE (CDE). Unfortunately, the samples are not of sufficient quality and the measurements are not of high enough sensitivity to observe the single-particle band directly. The indirect effects of this band where the collective excitations are pushed down toward lower energy is, however, clearly observed.

In conclusion, we have studied the collective excitations associated with intersubband transitions in GaSb/AlSb quantum wells. These excitations have been modeled within a self-consistent field theory and we have found that quantitative agreement can only be obtained if the nonparabolicity of the conduction band is explicitly included. The model suggests a way to observe the single-particle excitations in the backscattering geometry which has previously not been considered.

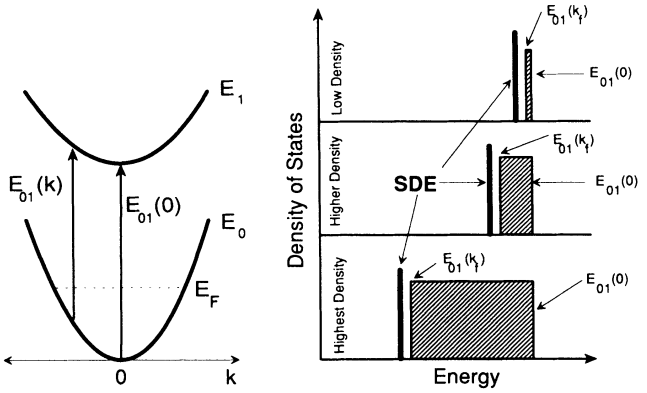

FIG. 3. A schematic diagram which demonstrates how nonparabolicity decreases the energy of the SDE. The cross-hatched region on the right represents the band of single-particle excitations shown as a function of increasing density. The heights of the SDE and single-particle bands are not to scale.

We would like to thank R.P. Leavitt for his help in the initial stages of the analysis of x-ray data and P.A. Knipp for a careful reading of the manuscript. This work was supported in part by the Office of Naval Research. G.B. acknowledges financial support from the National Research Council.

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