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Collective intersubband spin- and charge-density excitations in tilted magnetic fields

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Collective intersubband excitations, from the ground to the first excited electronic subband, are studied via inelastic light scattering in modulation-doped GaAs/$Al_xGa_{1-x}As$ quantum wells. The application of a magnetic field, tilted with respect to the growth direction, allows coupling of electron motion in the plane of the quantum well to motion along the growth axis such that combined intersubband–cyclotron-resonance transitions are observed. Anticrossing behavior between the combined resonances and the collective charge- or spin-density excitations is exhibited in the polarized or depolarized Raman-scattering geometries, respectively. The combined resonances show weak collective effects allowing an extrapolation of the bare subband separation and hence measurements of the depolarization and exchange-correlation shifts. Comparison to a theory employing the local-density approximation yields excellent results.

Intersubband excitations in confined structures have long been a topic of keen research interest.[1] Measurements of the intersubband separation are burdened by electron-electron correlations which arise from the direct Coulomb and exchange-correlation interactions. Collective intersubband charge-density excitations (CDE's or plasmons), which are observable in both far-infrared absorption[2] and Raman-scattering measurements, exhibit shifts from the "bare" intersubband energy due to both depolarization[3] and exchange-correlation[4] effects. In addition to the CDE's, electronic Raman-scattering measurements[5–7] allow the observation of collective intersubband spin-density excitations (SDE's or spin waves). The SDE's are especially exciting because they are not affected by the direct Coulomb interaction and are thus a sensitive probe of the exchange-correlation interaction if the value of the bare intersubband transition energy is well known.

In this paper we use inelastic light scattering to study collective intersubband excitations in magnetic fields which are tilted with respect to the plane of modulation-doped GaAs-$Al_xGa_{1-x}As$ quantum wells. In this geometry there is coupling between the intersubband transitions and cyclotron resonance such that combined resonances[8,9] are observed. Because the amplitudes of the combined resonances are smaller than the amplitudes of the main intersubband transitions, they exhibit weaker collective effects.[8,10] In principle, if the amplitudes of the combined transitions are small enough, the energy corresponding to the bare subband separation can be obtained by extrapolating the energy of the combined transitions to zero magnetic field. A comparison of this energy to the energies of the collective excitations yields a measure of electron-electron interactions. This method has previously been used to measure the depolarization shift associated with charge-density excitations in far-infrared transmission studies.[10] In those measurements only the plasmon could be observed at zero magnetic field, while at higher, tilted fields combined resonances could also be observed.

In the present work both spin- and charge-density intersubband excitations and their associated combined resonances were studied. In addition, the predicted anticrossing behavior[8,10] between the collective intersubband transitions and combined resonances was clearly observed in both Raman polarizations. Two samples with relatively high electron densities were studied. In the lower-density sample the combined resonances corresponding to the spin- and charge-density geometries occurred at the same energies as a function of magnetic field, a sign that very weak collective effects were involved. In contrast, the higher-density sample exhibited a clear depolarization shift between the energies of the combined resonances observed in the two geometries. In addition, the plasmon-phonon interaction strongly affects the CDE combined resonances in this sample indicating that care must be taken in extracting the bare subband separation from the combined resonances. We will show that a more reliable experimental value for the bare subband separation is obtained from the SDE data. Experimental results for both samples were compared to a calculation based on a theory by Ando[8] employing the local-density approximation (LDA); excellent agreement was obtained.

The samples used in this study consisted of single GaAs quantum wells with 1000-Å $Al_{0.25}Ga_{0.75}As$ barriers. The lower (higher) density sample had a quantum-well width of 275 Å (285 Å) and a delta-doped Si layer in the top barrier separated by 300 Å (150 Å) from the quantum well. The electron density N_s was estimated[7] to be 2.8×10^{11} cm^{-2} and 6.5×10^{11} cm^{-2} in the two samples. All Raman measurements were performed in the backscattering geometry with the exciting dye laser energy in resonance with the $E_0 + \Delta_0$ gap of the GaAs quantum well. At the tilt angles used in these measurements the wave-vector transfer into the plane of the sample $q \approx 1 \times 10^5$ cm^{-1}. The scattered light was dispersed with a Dilor triple monochromator and detected with a liquid-nitrogen-cooled EG&G charge coupled device (CCD) detector. Magnetic fields from 0 to 7.2 T were applied by a superconducting magnet in an optical access cryostat where the sample was kept at 5 K. Raman spectra in tilted magnetic fields for the low- and high-density samples are shown in Fig. 1. The dotted curves show spectra obtained in the depolarized Raman geometry, where the polarizations of the incident and scattered light are perpendicular, and the solid spectra were obtained in the polarized Raman geometry where the incident and scattered polarizations are parallel.

The two-dimensional electron gas in a tilted magnetic field was first considered by Ando.[8] He showed that the parallel and perpendicular motions of the electron gas are coupled by the in-plane component of the magnetic field such that combined intersubband-cyclotron transitions are observed. The states can be labeled as $|n, N\rangle$, where n is the subband index and N the Landau-level index. The transitions observed in Fig. 1 occur between different Landau levels of the ground and first excited subbands, $|0, N\rangle \rightarrow |1, N'\rangle$, where $\Delta N = N' - N = 0, \pm 1$. The values of N and N' depend on the Landau-level filling factor $\nu(B) = hcN_s/eB_\perp$, which for the lower (higher) -density sample varies between more than 28 (62) at 0.5 T to less than 2 (5) at 7 T. Assuming that the in-plane component of the magnetic field can be treated as a small perturbation and ignoring electron-electron interactions, the states $|n, N\rangle$ correspond to energies[8]

$$E_n^N(B) = E_n + (N + \tfrac{1}{2})\hbar\omega_{c\perp} + E_{\text{dia}}^n. \tag{1}$$

Here E_n is the energy at the bottom of the nth subband in the absence of magnetic fields, $\omega_{c\perp} = eB_\perp/m^*c$ is the cyclotron frequency corresponding to the magnetic field normal to the sample, and E_{dia}^n is the diamagnetic shift[8] of the nth subband due to the in-plane component of the magnetic field. The transitions $|0, N\rangle \rightarrow |1, N'\rangle$ occur at energies

$$E_{10}^{\Delta N}(B) = E_{10} + \Delta N\hbar\omega_{c\perp} + \Delta E_{\text{dia}}, \tag{2}$$

where $\Delta N = 0, \pm 1$ and $E_{10} = E_1 - E_0$ is the bare subband separation.

Collective effects are included within what is often referred to as the time-dependent local-density approximation. Assuming a two-subband system where only the lowest subband is occupied at zero temperature, we obtain the energies of the combined resonances at $q = 0$ from[11]

$$0 = 1 + \gamma_i L_1(\omega, B), \tag{3}$$

where $\gamma_{\text{CDE}} = \frac{2N_S}{E_{10}}(\frac{\alpha}{\varepsilon(\omega)} - \beta)$ and $\gamma_{\text{SDE}} = \frac{2N_S}{E_{10}}(-\beta)$. Here α and β, which are the zero-field depolarization and exchange-correlation corrections, respectively, have been defined previously[6] and the phonon contribution is included via $\varepsilon(\omega) = \frac{\omega^2 - \omega_{\text{LO}}^2}{\omega^2 - \omega_{\text{TO}}^2}$, and

$$L_1(\omega, B) = E_{10}^2 \sum_{\Delta N} \frac{C^{\Delta N}(B)}{[E_{10}^{\Delta N}(B)]^2 - (\hbar\omega)^2}. \tag{4}$$

The coefficients

$$C^{\Delta N}(B) = \frac{2}{\nu(B)} \sum_{N}^{\text{occupied}} J_{N,N+\Delta N}^2[\Delta_{10}(B)] \tag{5}$$

are essentially the amplitudes of the combined transitions. The orbit center separation

$$\Delta_{10}(B) = \sqrt{\frac{eB\sin^2\theta}{\hbar c\cos\theta}}(z_{11} - z_{00}), \tag{6}$$

where $z_{nn} = \langle n|z|n\rangle$ are zero-field matrix elements between subband states. The functions

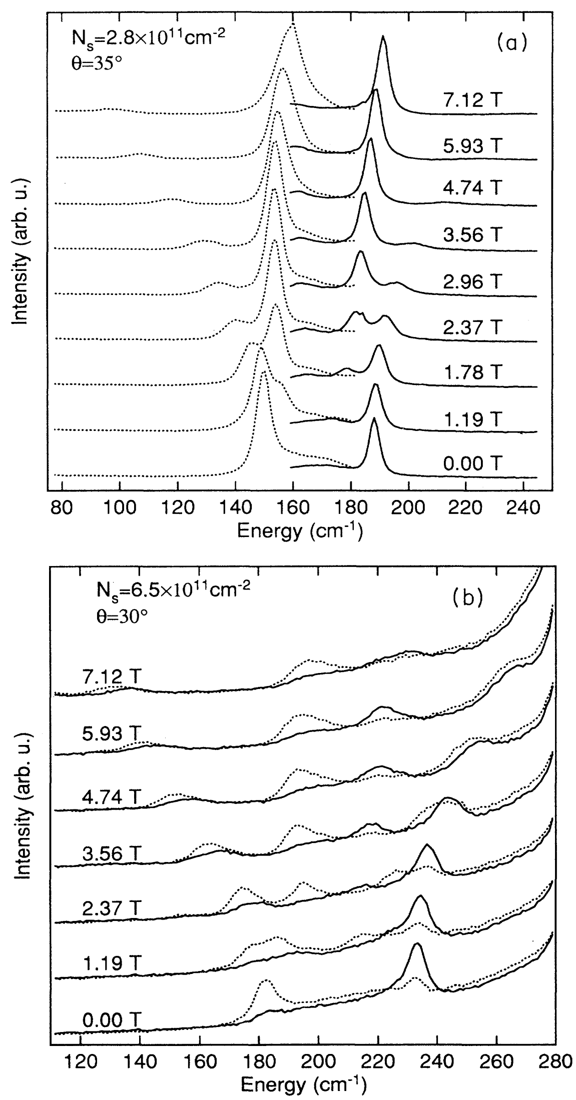

FIG. 1. Collective intersubband excitations as a function of magnetic field. The angle θ gives the direction of the magnetic field with respect to the sample normal. The dotted spectra are taken in the depolarized (SDE) Raman-scattering geometry and the solid spectra in the polarized (CDE) geometry. (a) Lower-density sample. Here the spectra have been truncated in order to facilitate ease of interpretation although all significant features remain intact. (b) Higher-density sample. The broad background is due to photoluminescence.

$$J_{N,M}(x) = \sqrt{\frac{M!}{N!}} \left(\frac{x}{\sqrt{2}}\right)^{N-M} L_M^{N-M}\left(\frac{x^2}{2}\right) \exp\left(-\frac{x^2}{4}\right) \tag{7}$$

are the overlap integrals of the Landau-level wave functions with $L_n^m(x)$ being the associated Laguerre polynomials.

The energy shifts of the observed transitions due to collective effects depend directly on the values of $C^{\Delta N}(B)$. These are plotted as dashed curves in the insets to Figs. 2 and 3. For our samples the amplitudes of the main intersubband transitions, $C^0(B)$, are virtually constant and of order unity at all magnetic fields of interest whereas the amplitudes of the combined transitions $C^{\pm 1}(B)$ are considerably smaller. At low magnetic fields the main $\Delta N = 0$ CDE (SDE) transitions exhibit larger shifts to higher (lower) energies than the $\Delta N = \pm 1$ combined transitions. As the magnetic field is increased the main transitions remain independent of field (except for a small diamagnetic shift) while the combined transitions move up or down in energy as $\pm\hbar\omega_{c\perp}$ and thus cross the main transitions. Since these resonances are coupled by the in-plane component of the magnetic field, an anticrossing results. This behavior is clearly exhibited in the calculated $\Delta N = 0, \pm 1$ transitions which were obtained from Eq. (3) and are shown in Figs. 2 and 3 as the solid (CDE) and dashed (SDE) curves. It should be stressed that the calculated curves constitute a simulation of the experiment using empirically measured input parameters and not a fit to the data. In order to obtain some of the quantities which are used in the theory, such as α, β, z_{00}, and z_{11}, it is necessary to calculate the electron wave functions in the quantum wells. This is done self-consistently within an effective-mass approximation using the growth parameters of our samples. This along with the determination of N_s has been described previously.[6,7] Nonlocal exchange-correlation[7,12] and nonparabolicity[13] effects on the collective excitations were not included. The in-plane effective masses used in the calculations, $m^* = 0.076m_e$ (low-density sample) and $m^* = 0.080m_e$ (high-density sample), were obtained from the in-plane cyclotron resonance of energy $\hbar\omega_{c\perp}$ (where $m^* = eB_\perp/\omega_{c\perp}c$) which were observed in the Raman data but are not shown. Extrapolating the combined resonances back to zero field yielded bare subband separations of $E_{10} = 161$ cm^{-1} and $E_{10} = 200$ cm^{-1}, for the lower- and higher-density samples, respectively. These values are 8 cm^{-1} lower than the peak positions of the single-particle bands measured at $k = 0$ and at zero magnetic field.[6,7,14] If one uses these bare intersubband energies, the magnitude of the exchange-correlation interaction is in excellent agreement with that predicted by the LDA.[7]

The peak positions of the CDE and SDE spectra are plotted versus magnetic field in Figs. 2 and 3. The assignments were made based on Raman selection rules

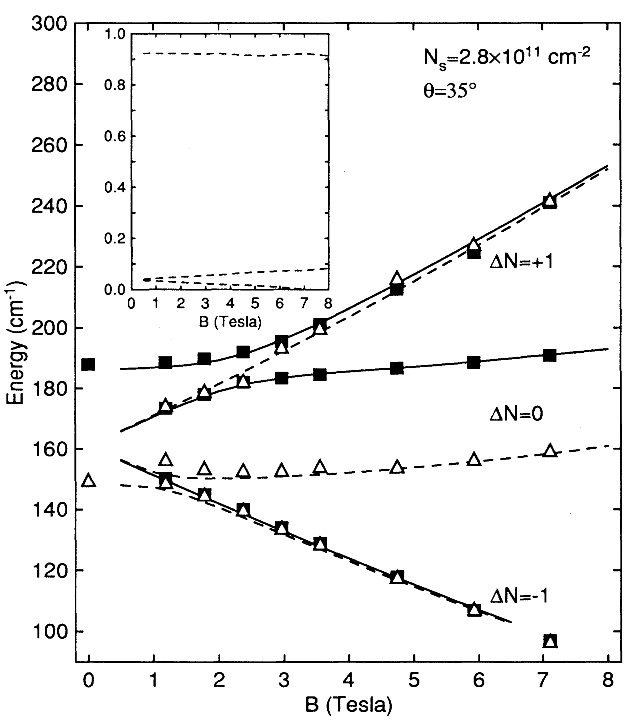

FIG. 2. A comparison between measured and calculated values for the energies of the peak positions of the combined resonances for the lower density sample as a function of magnetic field. ■ denotes peak positions measured in the polarized (CDE) Raman-scattering geometry and △ those measured in the depolarized (SDE) geometry. The solid curves correspond to the $\Delta N = 0, \pm 1$ combined resonances calculated in the CDE geometry and the dashed curves to calculations for the SDE geometry. The inset shows values of $C^{\Delta N}(B)$ with curves corresponding to $\Delta N = 0$, $\Delta N = +1$, and $\Delta N = -1$ displayed from top to bottom, respectively.

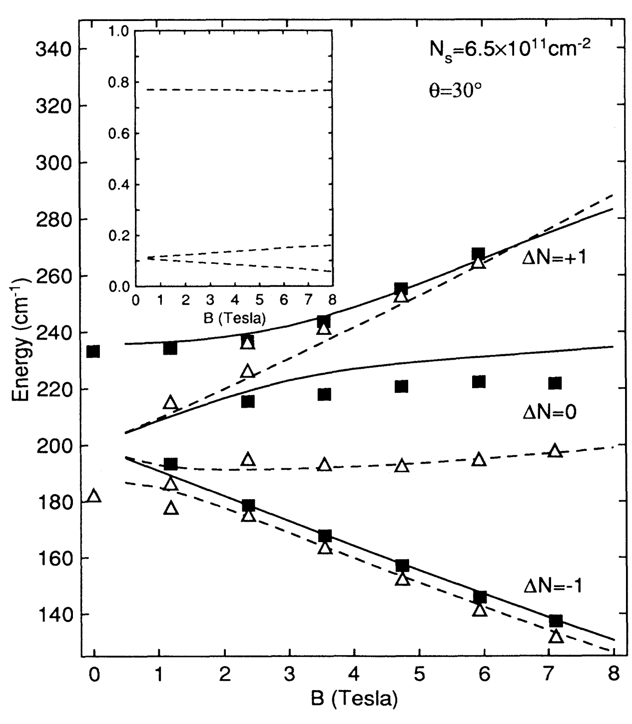

FIG. 3. Same as Fig. 2 but for the higher-density sample.

and the arguments above. In Fig. 2 a peak observed in both polarizations and identified as the single-particle excitation[14] was left out in order to simplify the figure. The magnetic-field dependence of this feature was previously described in Ref. 14. Similarly in Fig. 3, a peak previously identified[7] as the intersubband transition from the first excited to second excited subband (e.g., E_{21}) was left out of the analysis. The appearance of two peaks corresponding to the $\Delta N = +1$ SDE at 2.4 T is not understood but may be related to E_{21} which occurs at $\sim 223\ \mathrm{cm}^{-1}$. The small discrepancy between the calculated $\Delta N = 0$ CDE and experiment in Fig. 3 is also thought to be due to interaction with E_{21}. The small energy differences, which are both observed and calculated, between the SDE and CDE combined resonances are due to depolarization effects. For the lower-density sample these effects are small. This is because the amplitudes $C^{\pm 1}$ are quite small at all fields, as seen in the inset to Fig. 2. In fact, C^{-1} vanishes at ~ 7 T where the Landau-level filling factor drops below 2 and the $N = 1$ Landau level is completely depopulated. Referring to Fig. 2 we see that theory and experiment are in very good agreement and that the predicted small depolarization shifts are verified. In this sample it is thus possible to use either the SDE or CDE data in order to obtain the bare subband separation by extrapolating back to zero field.

From Fig. 3 we see that agreement between theory and experiment in the higher-density sample is again excellent. In this sample C^{-1} does not vanish until ~ 16 T so that both $C^{\pm 1}$ remain relatively large at all fields of interest. The depolarization shift between the $\Delta N = -1$ CDE and SDE combined resonances is essentially constant and clearly observable at all fields [see also Fig. 1(b)]. Interpretation of the $\Delta N = +1$ data is considerably more complicated due to interaction with the LO phonon which occurs at $\hbar\omega_{\mathrm{LO}} = 295\ \mathrm{cm}^{-1}$ [the tail of which is visible on the high-frequency side of Fig. 1(b)]. The resonance we have been associating with the charge-density excitation is actually the L^- branch of the coupled plasmon-phonon excitation[15] which is further coupled to cyclotron resonance. In essence, the $\Delta N = +1$ resonance, which we observe in the charge-density geometry, is depressed due to an interaction with the L^+ branch. In contrast, the spin-density excitation does not couple to the polarization field of the phonon and its energy is unaffected by the presence of the phonon. The expected large depolarization shift between the $\Delta N = +1$ CDE and SDE resonances is thus depressed due to the interaction between the CDE and phonon and, in fact, the calculation predicts that they will cross near the TO-phonon energy. In this sample, it is thus not possible to simply extrapolate the CDE data back to zero field[10] in order to obtain the bare subband separation. However, since the SDE is unaffected by the electron-phonon interaction, an extrapolation back to zero field of the SDE data would give a reliable value for the bare intersubband energy.

In conclusion, we have studied collective intersubband excitations associated with $\mathrm{GaAs/Al}_{1-x}\mathrm{Ga}_x\mathrm{As}$ modulation-doped quantum wells in tilted magnetic fields. We have observed combined intersubband-cyclotron transitions in both the CDE and SDE geometries and clear anticrossing behavior between the $\Delta N = 0$ and $\Delta N = \pm 1$ resonances. In addition, we have established that in order to obtain a reliable measure of the bare subband separation by extrapolating the combined transitions back to zero magnetic field, the SDE geometry should be used.

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[11] To our knowledge a theory which treats intersubband excitations in a magnetic field has not been developed for finite q. Our experimental results are obtained at $q \approx 1 \times 10^5$ cm^{-1} and compared to a theory which assumes that $q = 0$. Based on the q-dependent studies of collective excitations in Refs. 5 and 6 we assert that we are in the relatively small q limit and that our analysis is justified. Nevertheless a theory for finite q needs to be developed.

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