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## Direct observation of the intersubband Bernstein modes: Many-body coupling with spin- and charge-density excitations

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The spectrum of collective intersubband magneto excitations in a quasi-two-dimensional electron system confined in a single quantum well has been studied by inelastic light scattering spectroscopy. Intersubband Bernstein modes with a change of the Landau level index up to three were observed in a perpendicular magnetic field. It is demonstrated that Bernstein modes couple with intersubband charge and spin density excitations through many-body Coulomb interaction, and the coupling strength is governed by the in-plane momentum. Detailed measurements of the coupling strength dispersion were performed and compared with theoretical simulations. [S0163-1829(99)50320-2]

Intersubband collective excitations in a quasi-twodimensional electron system (2DES) confined in a semiconductor quantum well (QW) have long been a subject for theoretical and experimental interest.<sup>1,2</sup> Two essentially different intersubband collective modes have been detected in the infrared absorption and resonance inelastic light scattering, which are the charge and spin density excitations (CDEs and SDEs).<sup>1,2</sup> The former is associated with a macroscopic polarization of the quasi-2DES perpendicular to the QW plane (depolarization shift), while the latter is associated with the Coulomb interaction between the electron in the upper subband and the hole left behind in the lower subband (excitonic shift). The excitonic shift reduces the SDE energy relatively to the single-particle-excitation (SPE) energies. The CDE could be shifted both up and down from the SPE, depending on the parameters of the quasi-2DES.<sup>3–5</sup>

When a magnetic field is applied perpendicular to the QW interface, the quasi-2D energy spectrum becomes completely discrete. This results in a different type of intersubband collective excitations, intersubband Bernstein modes (or combined resonances), involving transitions with a simultaneous change of the quantum subband and Landau level (LL) indexes. While the intrasubband magneto excitations in quasi-2DES have been extensively discussed both theoretically and experimentally,<sup>1,2</sup> the properties of intersubband magneto-excitations are not so well understood up to now, mainly due to a lack of the experimental information. A direct observation of the intersubband Bernstein modes (ISBMs) had long

been a considerable experimental problem, as the optical transitions with an excitation of the ISBMs do not conserve the LL index, being therefore dipole forbidden in both infrared absorption and inelastic light scattering. One possible way to overcome the problem of LL index conservation is to couple the in-plane and transverse electron motion by an in-plane magnetic field  $(B_{\parallel})$ .<sup>6</sup> Using a tilted magnetic field, the ISBMs with  $\Delta = \pm 1$  (*n* is an LL index) were detected in infrared absorption and inelastic light scattering.<sup>7,8</sup> The experiments demonstrated, however, that the parallel component of the magnetic field modified the spectrum of magnetoexcitations considerably. The effect of  $B_{\parallel}$  could be accounted for at zero in-plane momentum (q), when the energy spectrum of ISBMs in a  $B_{\perp}$  is trivial.<sup>6</sup> At nonzero q the ISBMs energies are renormalized by the many-body Coulomb interaction.9 In this case two different contributions due to the Coulomb interaction and  $B_{\parallel}$  could not be easily separated.

In the present work, we report on a direct observation of ISBMs at nonzero q in a *perpendicular* magnetic field. This allows us to extract the many-body contribution to the spectrum of ISBMs. As a manifestation of the many-body Coulomb interaction, a coupling of ISBMs with both principal intersubband excitations, SDE and CDE, occurs. The coupling becomes stronger, while q increases, demonstrating an increasing role of Coulomb correlations. The spectrum of intersubband collective magneto excitations is modeled within a framework of the time-dependent local density ap-

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proximation (TDLDA) treating the direct and exchangecorrelation Coulomb terms self-consistently. A close agreement between experiment and theory is found.

A set of high quality asymmetrically doped  $Al_xGa_{1-x}As/GaAs$  heterostructures with a single QW of 250 Å has been studied. The electron density  $(n_s)$  and mobility  $(\mu)$  in different samples vary from 0.5 to  $6.8 \times 10^{11}$  cm<sup>-2</sup> and from 1 to  $7 \times 10^6$  cm<sup>2</sup>/(V s), respectively. The high quality of the samples is confirmed by extremely narrow luminescence and inelastic light scattering lines (~0.3 and 0.15 meV, respectively). Samples were immersed in a cryostat with a superconducting solenoid operating from 0 to 11 T at a base temperature of 1.5 K.

To study ISBMs at nonzero q we employed resonant inelastic light scattering, a method having been proved to be a most powerful tool to measure the dispersion of collective excitations in quasi-2DES.<sup>2</sup> The inelastic light scattering spectra were obtained with a Ti-sapphire laser tunable above the fundamental band gap  $E_0$ . The excitation power density was about 0.1-1 W/cm<sup>2</sup>. The back-scattering geometry was applied for the measurements, being performed with a twofiber optical system.<sup>10</sup> One fiber was used for photoexcitation while through another fiber the scattered light was collected. To distinguish inelastic light scattering from CDE and SDE type excitations copolarized and cross-polarized incident and scattered beams were used. Polarization of the light was achieved with linear polarizers operating at liquid helium temperatures, which were attached to the ends of the fibers just nearby the sample. The in-plane momentum was transferred into the quasi-2DES via the light scattering process. Its value was defined by an arrangement of the fibers relative to the sample surface. The maximum accessible q in our configuration was  $1.2 \times 10^5$  cm<sup>-1</sup>. The scattered light was detected by a CCD camera and a double grating monochromator Ramanor U-1000, which provided a spectral resolution of 0.04 meV. The in situ Hall measurements were employed to control the electron density under quasiequilibrium illumination conditions. Part of the polarization measurements were performed in an optical cryostat with a split-coil magnet producing a magnetic field up to 7 T.

The problem of LL index conservation in a perpendicular magnetic field has been circumvented by using incoming and outgoing resonances involving the light and heavy hole states near the top of the valence band in the QW. The states of heavy and light holes with different LL indexes are coupled in a perpendicular magnetic field,<sup>11,12</sup> and as a result the usually forbidden inelastic light scattering by ISBMs becomes allowed. It has to be noted, however, that the inelastic light scattering efficiency in a magnetic field is a function of two independent parameters, an applied magnetic field and laser excitation energy  $(E_I)$ , as the spectral density of incoming and outgoing resonances is nonuniform.<sup>1</sup> It is not necessarily that scattering by a particular resonance will be observable, if both parameters are fixed. To detect the maximum possible number of ISBMs in the whole range of magnetic fields under consideration, we performed a large set of measurements varying the magnetic field and  $E_L$  indepen*dently*. In fact, the  $E_L$  was tuned through 140 meV above  $E_0$ in a step of about 2 meV. For each step the inelastic light scattering spectra were measured in the whole range of magnetic fields.



FIG. 1. Inelastic light scattering spectra of a sample with QW width of 250 Å and  $n_s = 6.8 \times 10^{11} \text{ cm}^{-2}$  measured at  $q = 0.4 \times 10^5 \text{ cm}^{-1}$ ,  $E_L = 1.587$  eV, and different magnetic fields.

An example of inelastic light scattering spectra of a sample with  $n_s = 6.8 \times 10^{11}$  cm<sup>-2</sup> measured at  $q = 0.4 \times 10^5$  $cm^{-1}$  and  $E_L = 1.587$  meV is shown in Fig. 1. The zero magnetic field spectrum consists of two narrow peaks at 25.7 and 30.7 meV arising from SDE and CDE, and a broad band situated between them, which is commonly assigned to the scattering by SPE continuum [Fig. 1(a)].<sup>3</sup> Proper selection rules for SDE and CDE were checked using standard polarized and depolarized configurations for incident and scattering beams.<sup>2</sup> Upon a sweeping of magnetic field (B) the SPE band splits into a set of distinct spectral components, marked in Fig. 1 as  $B_{+1}$ ,  $B_{-1}$ ,  $B_{-2}$ ,  $B_{-3}$ , and  $L_0$ .  $B_{+n}$  and  $B_{-n}$ shift upward and downward from the maximum of the SPE band, while B increases. At a small but finite magnetic field  $B_{+n}$  crosses the CDE, and  $B_{-n}$  crosses SDE. While  $B_{+1}$  and  $B_{-1}$  approach CDE and SDE, respectively, their energies and intensities undergo a modification, clearly manifesting a coupling between  $B_{\pm 1}$  and CDE, and  $B_{-1}$  and SDE [Figs. 1(b) and 1(c)]. At large B the principal intersubband resonances, SDE and CDE, and positioned between them broad  $L_0$  band dominate the spectra [Fig. 1(d)].

The experimentally determined energies of the inelastic light scattering lines acquired in the whole range of  $E_L$  and B have been combined in a single plot. Figure 2 illustrates the magnetic field dependence of the Raman shift measured for all the observed lines. It is seen that the energies of both CDE and SDE are almost independent on B beyond the crossing range with  $B_{\pm 1}$  and  $B_{-1}$ . The energies of  $B_{\pm n}$  step from the intersubband gap and form positive and negative LL fan diagrams corresponding to an effective mass of about  $0.071m_0$  ( $m_0$  is the mass of the free electron). Since this effective mass nearly coincides with the electron effective mass in GaAs ( $0.069m_0$ ),  $B_{\pm n}$  is to be associated with scattering by ISBMs, whose energies have to converge to the single-particle excitation energies at  $q \rightarrow 0$ ,

$$E_{B\pm n} = |\hbar\Omega_{10} \pm n\hbar\omega_c|, \qquad (1)$$



FIG. 2. Fan plot of the intersubband magneto excitations at  $q = 0.4 \times 10^5$  cm<sup>-1</sup>, experiment (dots) and simulations (dashed lines), measured in a sample with a QW width of 250 Å and  $n_s = 6.8 \times 10^{11}$  cm<sup>-2</sup>. The simulated curves above the  $\hbar \Omega_{10}$  demonstrate only charge-density-type excitations, while below  $\hbar \Omega_{10}$  spindensity-type excitations are shown. A large open circle at B = 0 T shows the maximum of SPE continuum. The dash-dotted line is a guide for the eye pointing the  $\hbar \omega_{10}$  as determined by the extrapolating of ISBMs energies to B = 0 T.

for  $n \neq 0.^{1}$  Here,  $\Omega_{10}$  and  $\omega_{c} = eB/m_{e}c$  are the electron intersubband and cyclotron frequencies. We stress that ISBMs with  $|\Delta n| \ge 1$  are *directly* observed in a perpendicular magnetic field for the first time.

The formula (1) which is a quasi-2D analog of the Kohn theorem states that the energies of combined intersubbandcyclotron excitations are not modified by the Coulomb interaction at  $q=0.^{1}$  At small, but nonzero, q it describes the ISBM energies satisfactorily far from the crossing range of  $ISBM_{+1}$  (ISBM\_{-1}) with CDE (SDE). When the energy of  $ISBM_{+1}$  (ISBM\_{-1}) approaches that of CDE (SDE) a resonance splitting occurs (see Fig. 2). A theoretical description of the resonance coupling between  $ISBM_{+n}$  and CDE in perpendicular magnetic field was previously achieved within a framework of the random-phase approximation (RPA).<sup>9</sup> The RPA approach could be naturally extended to a more general case by including the exchange-correlation interaction within TDLDA approximation.<sup>6,13,14</sup> Using the TDLDA, the spectrum of collective excitations involving intersubband transitions between the ground and first excited electron subbands has been simulated. It is proportional to the imaginary part of the polarization function  $\tilde{\chi}_i^{10}(q,\omega)$  (the subscript refers to the charge or spin-density response), which results in

$$1 - \operatorname{Re} \gamma_{i}(q)\chi^{10}(q,\omega) = 0,$$

$$\operatorname{Im} \gamma_{i}(q)\chi^{10}(q,\omega) = 0,$$
(2)

where  $\chi^{10}(q,\omega)$  is the intersubband part of the polarization function of noninteracting quasi-2DES in a magnetic field.<sup>9,15</sup>  $\gamma_i(a)$  represents many-body effects and is expressed through the depolarization shift and the exchangecorrelation contribution  $[\alpha(q) \text{ and } \beta_i]$ .<sup>3,4</sup> The dynamical screening of the direct Coulomb interaction by long-wave LO phonons is included through a frequency-dependent di-



FIG. 3. The energies of the  $\omega_{+1}^+$  and  $\omega_{-1}^-$  (a), and  $\omega_{-1}^-$  and  $\omega_{-1}^+$  (b) coupled modes at two different q,  $0.4 \times 10^5$  cm<sup>-1</sup> (open circles) and  $0.8 \times 10^5$  cm<sup>-1</sup> (solid circles), measured in a sample with a QW width of 250 Å and  $n_s = 6.8 \times 10^{11}$  cm<sup>-2</sup>. Dispersions of  $\Delta_1$  and  $\Delta_2$  are shown in the left and right insets, respectively. The dashed lines are simulated dispersion curves.

electric function.<sup>2</sup> Following Ref. 9 the long-wave approximation  $\chi^{10}(q,\omega) \sim (ql_B)^6$   $(l_B = \sqrt{\hbar c/eB}$  is a magnetic length) is used, which implies neglecting electron transitions with  $\Delta n > 3$ . The result of the simulations is shown in Fig. 2 by dashed lines. The resonance splittings between  $ISBM_{+n}$  $(ISBM_{-n})$  and CDE (SDE) naturally appear in the simulated spectrum (Fig. 2). At  $|\Delta n| > 1$  they are too small to be compared with the experiment, but that between  $ISBM_{+1}$  $(ISBM_{-1})$  and CDE (SDE) is seen to be in perfect agreement with the experimental result (Fig. 2). One may thus conclude that the experimentally observed resonance splittings are indeed due to the *many-body coupling* between  $ISBM_{+1}$  and principal intersubband collective excitations, CDE and SDE. The ISBM<sub>+1</sub> and CDE are coupled by the direct Coulomb interaction, while ISBM-1 and SDE are coupled by the exchange-correlation Coulomb interaction. Further, we will refer to ISBM<sub>+1</sub> and CDE coupled modes as  $\omega_{+1}^+$  and  $\omega_{+1}^-$ , and to ISBM<sub>-1</sub> and SDE coupled modes as  $\omega_{-1}^-$  and  $\omega_{-1}^+$ .

Experimentally the coupling strength between  $\omega_{\pm 1}^+(\omega_{-1}^-)$ and  $\omega_{+1}^{-}(\omega_{-1}^{+})$  can be varied in two different ways: (i) by changing q and (ii) by changing  $n_s$  (both direct and exchange-correlation Coulomb terms are monotonic functions of  $n_s$ ).<sup>5,16</sup> Figure 3 demonstrates energies of  $\omega_{+1}^+(\omega_{-1}^-)$  and  $\omega_{+1}^-(\omega_{-1}^+)$  measured at two different values of q, 0.4 and  $0.8 \times 10^5$  cm<sup>-1</sup>, and fixed  $n_s = 6.8 \times 10^{11}$  cm<sup>-2</sup>. The energy gap between  $\omega_{+1}^{-}(\omega_{-1}^{+})$  and  $\omega_{+1}^{+}(\omega_{-1}^{-})$  coupled modes  $[\Delta_1(\Delta_2)]$  is nearly twice as large at  $q=0.8\times 10^5$ cm<sup>-1</sup> than that at  $q=0.4\times10^5$  cm<sup>-1</sup> (Fig. 3). Summarized dispersions of the  $\Delta_1$  and  $\Delta_2$  are shown in the left and right insets to Fig. 3, respectively. Both  $\Delta_1$  and  $\Delta_2$  increase with increasing q. At small q  $(ql_B \ll 1)$  they are *linear* functions of q in agreement with theoretical expectations. A quantative discrepancy between the experimental and simulated dispersion curves for  $\Delta_2$  (see right inset to Fig. 3) appears most probably due to the static and dispersionless character of the exchange-correlation functional used by the TDLDA theory.

Using the samples grown with equal QW widths and different doping levels we have also studied the  $\Delta_1$  as function of  $n_s$ . Figure 4 presents experimental values of  $\Delta_1$  measured R12 754



FIG. 4. Dependence of the  $\Delta_1$  on the  $n_s$  at fixed q of  $1.1 \times 10^5$  cm<sup>-1</sup>, experiment (circles) and theory (dashed line).

at fixed  $q = 1.1 \times 10^5$  cm<sup>-1</sup>, and different  $n_s$ . As one may expect,  $\Delta_1$  decreases monotonously with decreasing  $n_s$ . At large  $n_s$  ( $n_s \ge 2 \times 10^{11}$  cm<sup>-2</sup>)  $\Delta_1$  is effectively reduced due to the optical phonon screening of the direct Coulomb interaction (Fig. 4). It is interesting to note that  $\Delta_1$  vanishes at small, but finite,  $n_s$  close to the critical electron density  $n_s^c$  $\approx 4 \times 10^{10}$  cm<sup>-2</sup>, at which the direct and exchangecorrelation Coulomb terms become equal.<sup>5,14</sup> Below  $n_s^c$  the ISBM<sub>+1</sub> does not cross the CDE anymore. Indeed, the crossing between ISBM<sub>+1</sub> and CDE has not been detected below  $5 \times 10^{10}$  cm<sup>-2</sup> in the experimental spectra. We have not so far considered the  $L_0$  band, dominating the inelastic light scattering spectra at high magnetic fields. This line was discussed previously;<sup>8</sup> however, its nature was not completely understood. Our results could hardly support an assignment of  $L_0$  band to intersubband single-particle excitations (as was proposed in Ref. 8), since such an explanation would demand a violation of the translation symmetry in the studied samples. We did not find, however, any correlation between the sample quality and scattering efficiency of  $L_0$ . Since the nature of the  $L_0$  band is still under debate, and it does not have a direct connection to the matter discussed in the present paper, we plan to consider the  $L_0$  band separately in the future.

In conclusion, we have measured the spectrum of intersubband collective excitations in a perpendicular magnetic field. The intersubband Bernstein modes with  $\Delta n = \pm 1,2,3$ , are observed and investigated. It have been found that at nonzero in-plane momentum the intersubband Bernstein mode with  $\Delta n = +1$  couples with the charge density excitations through the direct Coulomb interaction, while the intersubband Bernstein mode with  $\Delta n = -1$  couples with the spin density excitations through the exchange-correlation Coulomb interaction. Energy gaps between the coupled modes are linear functions of the in-plane momentum as long as the in-plane momentum is small if compared to the reciprocal magnetic length.

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