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Coupling of intersubband charge-density excitations to longitudinal-optical phonons in modulation-doped GaAs quantum wells

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Abstract

We have investigated the interaction between the longitudinal-optical (LO) phonon and the lowest intersubband charge-density excitation of the two-dimensional (2D) electron gas formed in a modulation-doped single quantum well structure. The intersubband spacing is continuously increased by applying dc voltages up to about 200 V between a top gate and a back contact. The frequency of the LO-phonon mode confined to the quantum well exhibits an electron-density-dependent redshift which is attributed to enhanced screening effects by the 2D electron gas. Inelastic light-scattering spectra reveal a clear anticrossing behavior of the energies of LO-phonon and charge-density excitation, for which a coupling constant of 1.5(3) meV is obtained.

Keywords: A. Quantum wells; D. Phonons

Nowadays, epitaxial-growth technology enables the fabrication of extremely high-mobility two-dimensional (2D) electron gases in semiconductor quantum well structures, which are particularly suitable for studies of phenomena associated with electronic correlations in reduced dimensions [1]. Coulomb interaction effects are apparent in the spectrum of elementary excitations of the electron gas, which can be probed by inelastic light scattering [2,3]. Electron–electron interactions give rise to collective excitations of the charge (CDE) and spin density (SDE), which are distinguished in light-scattering spectra by their different polarization selection rules [4]. Many-body effects manifest themselves in the energy shift of the collective modes with respect to the so-called single-particle excitations (SPE) of the non-interacting electron gas [5–9]; the observation of the latter still being a controversial matter [10,11]. In the long wavelength limit, the frequencies of the collective modes associated with electron–hole pair transitions between an occupied subband i with 2D density n_i and an empty one $_j$ are expressed within the local-density approximation as [5–9]:

$$\omega_{\mathrm{SDE}}^2 = E_{ij}^2 - 2n_i E_{ij}\beta_{ij} \tag{1}$$

$$\omega_{\mathrm{CDE}}^2 = E_{ij}^2 + 2n_i E_{ij}(\alpha_{ij}/\tilde{\epsilon} - \beta_{ij}) \tag{2}$$

where E_{ij} is the intersubband spacing energy. The parameter β_{ij} represents the so-called excitonic shift of the collective SDE resulting from exchange-correlation vertex corrections. In contrast, the CDE is shifted upwards from the spin-density excitation due to the Hartree interaction or depolarization shift α_{ij}, which is associated with the macroscopic electric field created by the charge-density fluctuations themselves. The factor $\tilde{\epsilon}$ represents the phonon contribution to the dielectric function.

Due to the polar character of the GaAs lattice, there is a strong coupling between electron gas and vibrational degrees of freedom. In the frequency range of the optical phonons, for instance, the Coulomb interaction between electrons is largely screened by the lattice. This effect is taken into account in the function $\tilde{\epsilon}(\omega_{\mathrm{CDE}})$ of Eq. (2). The electron gas, in turn, might also have an influence on the

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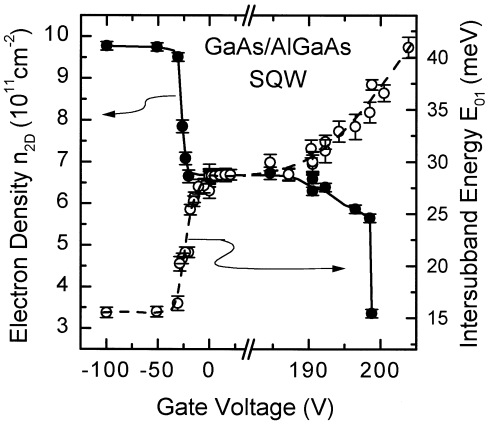

Fig. 1. Dependence on gate voltage of the total electron density n_{2D} and intersubband spacing E_{01} of a modulation-doped single quantum well at 5 K, as determined from PL spectra. The curves are a guide to the eye.

longitudinal–transverse splitting of the optical modes. Moreover, in heavily doped bulk semiconductors the LO phonon interacts with the macroscopic electric field of the plasmon, which can be regarded as compression wave of the electronic charge density, leading to the formation of coupled L_+ and L_- modes [12,13]. In doped quantum wells, a similar mode mixing is expected for LO phonon and intersubband charge-density excitation [4,14,15], in spite of the transverse character of the latter.

In this work we show that the intersubband spacing energy in a modulation-doped GaAs quantum well can be tuned towards degeneracy with the optical phonons by applying a large dc voltage between a gate and a back contact on the sample. The depolarization as well as the excitonic shift of the collective modes of the electron gas are determined as a function of bias from the energies of the excitations measured in light-scattering experiments. We find that the LO-phonon frequency in the well becomes renormalized (softened) with respect to that of the bulk due to additional screening by the electron gas. With increasing intersubband spacing the renormalized LO phonon and the collective CDE exhibit a clear anticrossing behavior of their energies. Using a simple two-level model and taking into account the anomalous dispersion of the dielectric function in the optical phonon region we obtained for the strength of the coupling a value of 1.5(3) meV.

The sample consists of a modulation-doped 245 Å-wide GaAs single quantum well (SQW) with $Al_{0.33}Ga_{0.67}As$ barriers grown by molecular-beam epitaxy. The growth sequence is given elsewhere [16]. Without bias only the lowest subband is occupied with electrons with Fermi energy $E_F \approx 20$ meV. The energy separation to the second subband is $E_{01} \approx 28$ meV. A large dc bias ranging from -100 to 200 V is applied between a gate electrode evaporated onto the top surface of the sample and a back contact on the substrate side. Photoluminescence (PL) and inelastic

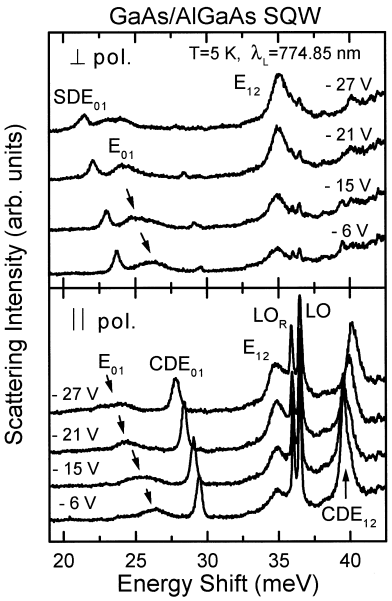

Fig. 2. Depolarized (⊥) and polarized (∥) light-scattering spectra of intersubband excitations at different negative voltages. The assignment of peaks to charge-density (CDE), spin-density (SDE), and single-particle excitations (E_{ij}) as well as the LO phonons is indicated.

light-scattering spectra were excited with a tunable Ti:sapphire laser and recorded with optical multichannel detection. Light-scattering measurements were performed in backscattering geometry with incident photon energies in resonance with excitonic transitions between higher excited states of the SQW.

The dependence on applied voltage of the 2D electron density n_{2D} as well as the intersubband energy E_{01} have been determined from a quantitative analysis of PL lineshapes as described elsewhere [16,17]. The values obtained for n_{2D} and E_{01} are plotted in Fig. 1 as a function of voltage. Depending on polarity we were able to increase or reduce n_{2D} roughly by a factor of two. In addition, the tilt of the band edges caused by the applied electric field tends to decrease or enhance the intersubband spacing, respectively. At high negative voltages above -30 V a filling of the second subband of the SQW is attained, which is revealed by a strong redshift of the corresponding PL line due to bandgap renormalization effects [17,18].

Fig. 2 shows representative inelastic light-scattering spectra in the energy range of intersubband excitations for different voltages measured at 5 K in backscattering geometry using different linear polarizations of incident and scattered beam. The sharp peaks observed in spectra with parallel polarization below and above the energy of the LO phonons correspond to the collective CDEs associated with electronic transitions between either the lowest occupied subband (0) and the first (1) excited one or between the first and the second (2) excited state of the SQW,

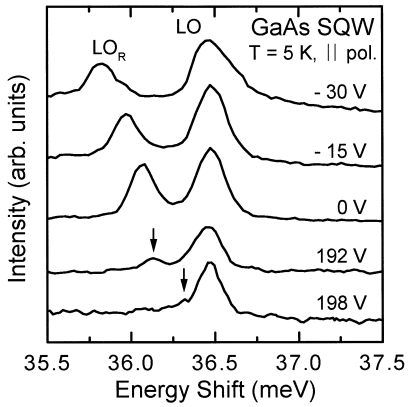

Fig. 3. Polarized light-scattering spectra in the narrow energy range of the longitudinal optical phonons for different negative and positive applied voltages. LO_R denotes the renormalized longitudinal vibrational mode of the doped quantum well (see text for details).

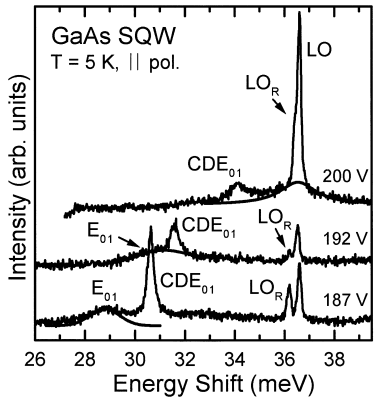

Fig. 4. Polarized light-scattering spectra for different positive bias showing the coupling between the intersubband charge-density excitation CDE_{01} and the LO_R phonon. The fitted Gaussian to the E_{01} peak is depicted in each spectra for clarity.

respectively. On the contrary, in depolarized spectra only the SDEs are active. These excitations are known to possess extremely sharp resonant enhancement profiles [13,19]; thus, in our case SDE_{12} remained unobserved. The broader peaks that appear in spectra with both polarizations are assigned to single-particle excitations. At long wavelengths they are centered at the intersubband spacing energies E_{01} and E_{12}. We point out that despite the fact that at low bias the Fermi energy lies below the bottom of subband (1), there is always a certain population of this subband due to thermal and photon-assisted excitation of carriers. For instance, the luminescence signal corresponding to recombination processes from subband (1) is at the laser powers used in light scattering as strong as the ground-state emission. This explains the observation of the features denoted as E_{12} and CDE_{12}.

We now turn to the discussion of the effects of the applied bias on the different elementary excitations observed in light-scattering spectra. With increasing 2D density, i.e. negative voltage, the energies of the excitations associated with the 0–1 transitions decrease due to the reduction of E_{01}. In contrast, the subband spacing E_{12} remains unchanged, whereas its corresponding charge-density excitation CDE_{12} shifts to higher energies indicating an enhancement of the macroscopic depolarization field at higher electron densities. We note that this peak is sometimes mistakenly assigned to the upper CDE–LO-phonon coupled mode [15]. The different behavior of both CDEs as a function of bias clearly speaks against such an interpretation. We show below that the excitation that couples to the CDE_{01} corresponds to the feature labeled LO_R having phonon character.

Fig. 3 displays light-scattering spectra in the energy range of the optical phonons for different negative as well as positive voltages. The peak denoted LO is centered at the energy of the longitudinal optical phonon of GaAs and its position is totally independent of bias. It originates most likely from Raman scattering processes taking place at the GaAs buffer layer and the substrate. The energy position of the LO_R feature, on the contrary, exhibits a redshift from the LO frequency, which is the larger the higher the electron density. Thus, we assign this peak to the LO-phonon mode corresponding to vibrations confined to the modulation-doped quantum well. The observed softening of its frequency (even at zero bias) might result from the screening by the free carriers of the transverse effective charge which gives rise to the TO–LO splitting in polar lattices. Within the Born–Oppenheimer approximation one can think of the electrons of the 2D gas rearranging instantly around the atomic cores of the lattice in such a way that the net negative charge of the anions and the positive one of the cations is effectively reduced. As an alternative explanation for the reduction of the TO–LO splitting we consider the increase of ε_∞, the contribution to the dielectric function arising from electronic interband transitions, due to bandgap renormalization effects at high electron densities. In any case, the softening is proportional to the electron gas density, as demonstrated by the spectra of Fig. 3.

In the following, we focus on the coupling of the intersubband CDE_{01} to the renormalized phonon LO_R. With increasing positive bias the intersubband energy E_{01} is tuned towards degeneracy with the LO_R phonon and finally a crossover, as shown by the spectra in Fig. 4. The associated collective charge-density excitation CDE_{01} cannot follow the bias dependence of E_{01} because of the coupling of its macroscopic electric field with that of the LO_R vibration. This coupling manifests itself in an anticrossing behavior of the mode energies, which can be appreciated better in Fig. 5, where we have plotted the CDE_{01} and LO_R peak positions (data points) as a function of the subband spacing E_{01}. The dashed lines correspond to the energies of the uncoupled modes. For LO_R we have taken the measured value at high negative voltages, where the coupling to the

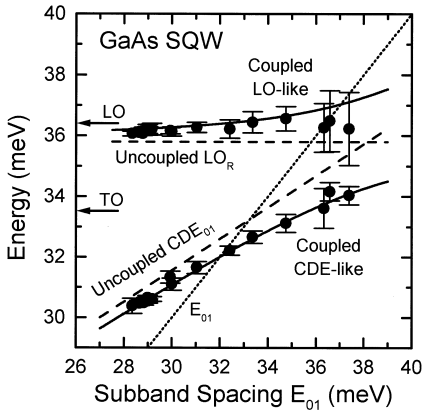

Fig. 5. Peak energy position of excitations measured in parallel polarization as a function of the spacing energy E_{01} between the two lowest subbands. Solid (dashed) curves correspond to the calculated coupled (uncoupled) LO-phonon–CDE modes within a two-level model. The dotted line represents the variation of the intersubband single-particle excitation.

CDE_{01} is negligible. The CDE_{01} values were calculated using Eq. (2) with constant Coulomb interaction parameters $2\alpha_{01} = 2.7(2) \times 10^{-11}$ meV cm^2 and $2\beta_{01} = 0.7(1) \times 10^{-11}$ meV cm^2 [17] and by equating $\tilde{\epsilon}(\omega_{CDE})$ according to the expression for a single Lorentz oscillator with damping $\Gamma = 0.1$ meV given by [20]

$$\tilde{\epsilon}(\omega) = \frac{(\omega_{LO}^2 - \omega^2)(\omega_{TO}^2 - \omega^2) + \omega^2\Gamma^2}{(\omega_{TO}^2 - \omega^2)^2 + \omega^2\Gamma^2} \tag{3}$$

We notice that after Eq. (3) one expects the second term in Eq. (2) corresponding to the depolarization shift to vanish due to the very efficient screening of the Coulomb interaction in the region of the so-called *reststrahlen band*, i.e. between the TO and LO frequency, as a consequence of the strong anomalous dispersion of the dielectric function. In fact, we observe the crossing of collective charge-density and single-particle excitations very close to the TO-phonon frequency and the subsequent broadening of the CDE_{01}, as it enters the reststrahlen band (see Figs. 4 and 5).

In order to obtain the strength of the coupling between both collective modes we have used a simple two-level model in which the degeneracy is lifted by introducing an interaction constant V. The solid curves in Fig. 5 represent the result of fitting the solutions of the 2×2 secular determinant to the data points having only V as the adjustable parameter. We obtain a good description of the coupled-mode energies with a value of $V = 1.5(3)$ meV, i.e. the coupling is weak but finite. This is an indication that mode mixing takes place although the macroscopic electric fields associated with the LO phonon and the transverse intersubband charge-density excitation are, a priori, mutually orthogonal. A coupling occurs because of the breakdown of translational symmetry caused by residual disorder from the ionized remote donors and well-width fluctuations [21].

In summary, we have varied the electron density and intersubband spacings in a modulation-doped GaAs single quantum well by applying a dc gate voltage to the 2D electron gas. From their dependence on bias we were able to identify the elementary excitations pertaining to different intersubband transitions between the two lowest electronic subbands and between the first and second excited ones. The energy of the LO phonon of the quantum well appeared to be renormalized due to the reduction of the TO–LO splitting by the electron gas. This phonon couples to the lowest intersubband charge-density excitation of the 2D electron plasma exhibiting an anticrossing behavior, as the intersubband spacing energy E_{01} is tuned across the reststrahlen band by the applied voltage. We determined the coupling constant to be 1.5(3) meV.

References

[1] For recent contributions, see Proceedings of the 11th International Conference on Electronic Properties of Two-Dimensional Systems, Nottingham, UK, August 1995, Surf. Sci. (1996) 361–362.
[2] A. Pinczuk, S. Schmitt-Rink, G. Danan, J.P. Valladares, L.N. Pfeiffer, K.W. West, Phys. Rev. Lett. 63 (1989) 1633.
[3] D. Gammon, B.V. Shanabrook, J.C. Ryan, D.S. Katzer, M.J. Yang, Phys. Rev. Lett. 68 (1992) 1884.
[4] A. Pinczuk, G. Abstreiter, in: M. Cardona, G. Güntherodt (Eds.), Light Scattering in Solids, Vol. V, Springer, Berlin, 1989, p. 153.
[5] D.A. Dahl, L.J. Sham, Phys. Rev. B 16 (1977) 651.
[6] T. Ando, J. Phys. Soc. Jpn 51 (1982) 3893.
[7] A.C. Tselis, J.J. Quinn, Phys. Rev. B 29 (1984) 3318.
[8] L. Wendler, V.G. Grigoryan, Solid State Commun. 71 (1989) 527.
[9] S.L. Chuang, M.S.C. Luo, S. Schmitt-Rink, A. Pinczuk, Phys. Rev. B 46 (1992) 1897.
[10] S. Das Sarma, D.-W. Wang, Phys. Rev. Lett. 83 (1999) 816.
[11] M. Sassetti, B. Kramer, Phys. Rev. Lett. 80 (1998) 1485.
[12] S. Ernst, A.R. Goñi, K. Syassen, M. Cardona, Phys. Rev. B 53 (1996) 1287.
[13] G. Abstreiter, M. Cardona, A. Pinczuk, in: M. Cardona, G. Güntherodt (Eds.), Light Scattering in Solids, Vol. IV, Springer, Berlin, 1984, pp. 5–150 (and references therein).
[14] E. Burstein, A. Pinczuk, D.L. Mills, Surf. Sci. 98 (1980) 451.
[15] G. Danan, A. Pinczuk, J.P. Valladares, L.N. Pfeiffer, K.W. West, C.W. Tu, Phys. Rev. B 39 (1989) 5512.
[16] S. Ernst, A.R. Goñi, K. Syassen, K. Eberl, Phys. Rev. Lett. 72 (1994) 4029.
[17] A.R. Goñi, M. Danckwerts, U. Haboeck, K. Eberl, C. Thomsen, Phys. Status Solidi (B) 215 (1999) 347.
[18] A.R. Goñi, U. Haboeck, M. Danckwerts, C. Thomsen, K. Eberl, F. Guinea, unpublished.
[19] D.C. Hamilton, A.L. McWorther, in: G.B. Wright (Ed.), Light Scattering Spectra of Solids, Springer, Berlin, 1969, p. 309.
[20] A.R. Goñi, K. Syassen, Semicond. Semimetals 54 (1998) 247.
[21] J.K. Jain, S. Das, Sarma, Phys. Rev. B 36 (1987) 5949.