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Intrasubband excitations and spin-splitting anisotropy in GaAs modulation-doped quantum wells

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We present Raman-scattering measurements of intrasubband excitations in the quasi-two-dimensional electron gas (2DEG) of an n-type modulation-doped GaAs/$Al_{0.33}Ga_{0.67}As$ quantum well. In depolarized spectra we observe spin-flip single-particle excitations between the spin-split conduction bands. Measurements with the Raman wave vector along different crystallographic orientations allow us to probe the anisotropy of the spin splitting, caused by the lack of inversion symmetry in GaAs. We discuss theoretical descriptions of the spin splitting in a 2DEG and compare calculated and measured Raman spectra. In polarized spectra we observe scattering with comparable intensity from non-spin-flip single-particle excitations and plasmons. In order to describe correctly the plasmon dispersion we have to take into account the effects on the Coulomb interaction of image charges in the sample surface.

The study by Raman scattering of electronic excitations in doped semiconductor heterostructures provides a wealth of information about the band structure and electron-electron interactions in quasi-two-dimensional electron gases (2DEG).[1-4] Excitations involving changes in spin (single-particle spin-flip and spin-density excitations) cause depolarized Raman scattering. For polarized scattering, spin has to be conserved and so non-spin-flip single-particle and plasmon scattering are allowed.[5] We have reported recently on the observation of intrasubband single-particle excitations and plasmons in a high-density quasi-2DEG confined in an asymmetric modulation-doped GaAs/$Al_{0.33}Ga_{0.67}As$ quantum well.[4,6] In the present paper we will readdress this work, taking a detailed theoretical approach, together with further experimental results, to provide a coherent picture of the excitations within this system.

The conduction-band spin splitting of order k^3 caused by the lack of inversion symmetry in zinc-blende structures such as GaAs has attracted considerable interest in recent years.[7-10] Of particular note in our earlier work was the observation in depolarized spectra of spin-flip single-particle excitations between the spin-split conduction bands.[4] There we compared our experimental spectra with calculations using an isotropic model for the spin splitting based on the expression for bulk.[11] In this paper we will discuss descriptions of the conduction-band spin splitting in quasi-2DEG systems. We are able to test the validity of the anisotropic models from comparisons of theoretical and experimentally determined light-scattering spectra for single-particle excitations with wave vectors in different crystallographic directions.

We have also previously reported on the polarized Raman spectra from the same sample, when we assigned the two modes observed to be due to coupled plasmons in the 2DEG and an additional low mobility 2DEG in the $Al_xGa_{1-x}As$ barrier.[6] This was in agreement with the established understanding that in the Raman-scattering process single-particle excitations are completely screened by electron-electron interactions.[5] However, recent work has shown that single-particle and collective excitations can coexist,[2,3,12,13] and so we now assign the low-energy mode to be due to non-spin-flip single-particle excitations and the high-energy mode to the plasmon of the high-mobility 2DEG, enhanced in energy by image charge effects from the sample surface. Hence, we can now arrive at a complete description of polarized and depolarized Raman scattering from electronic excitations in a quasi-2DEG.

The structure under investigation is a modulation-doped GaAs quantum well with an electron density of 1.3×10^{12} cm^{-2}, grown by molecular-beam epitaxy on a semi-insulating GaAs substrate. After a thick GaAs buffer layer a GaAs/$Al_{0.33}Ga_{0.67}As$ superlattice barrier was grown, followed by the 180-Å-thick GaAs quantum well layer. On top of this, a 100-Å-thick $Al_{0.33}Ga_{0.67}As$ spacer layer, a Si δ-doping layer, a further 100-Å $Al_{0.33}Ga_{0.67}As$ and a 50-Å GaAs capping layer were grown. The form of the self-consistent potential is very similar to that of a heterojunction. Raman-scattering measurements were taken close to the E_0 resonance of the GaAs quantum well, with the polarizations of the incident and scattered light both crossed and parallel, to give depolarized and polarized spectra, respectively. Changing the angle of the sample surface normal with respect to the incident and scattered wave vectors allows a probe of excitations in the structure with different in-plane wave vectors $\mathbf{q}$.[1] The sample was also mounted in different orientations to allow a probe of $\mathbf{q}$ along different crystallographic directions. We show in Fig. 1 depolarized spectra for an in-plane wave vector $q=0.49\times10^5$ cm^{-1} along the [10], 25° to [10], and [11] directions. Depolarized and polarized spectra for different $\mathbf{q}$ along [10] are shown in Fig. 2.

We will first address the question of the dispersions for the spin-split conduction bands of a quasi-2DEG, such as

that found in a heterojunction or modulation-doped quantum well. To obtain an expression for the spin splitting in a quasi-2D system from that for the bulk conduction band, we substitute for k_z, the wave-vector component in the confinement direction (the z axis), $\kappa=\langle \hat{k}_z^2 \rangle^{1/2}$ the root-mean-squared value of its associated operator $\hat{k}_z=-i\,d/dz$ onto the quantum-well ground state. This confinement perturbation should be included in the Hamiltonian[14,15] rather than in the expression for the bulk conduction-band spin splitting.[4] For the present structure, wave-function penetration into the barriers is very small and so we shall ignore any change in material parameters in our evaluation of the spin splitting. Within the envelope function formalism, from $\mathbf{k}\cdot\mathbf{p}$ theory the Hamiltonian with respect to the spin-up $|\phi(z)\uparrow\rangle$ and spin-down $|\phi(z)\downarrow\rangle$ conduction-band Γ-point states can be given by[14]

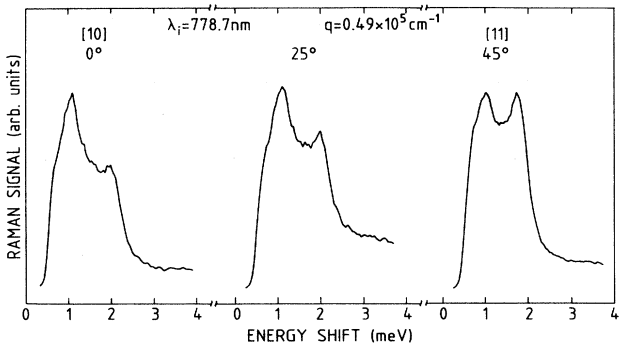

FIG. 1. Depolarized Raman spectra of spin-flip single-particle excitations for in-plane wave vector $q=0.49\times10^5$ cm^{-1} along the (a) [10], $\theta=0°$; (b) $\theta=25°$; and (c) [11], $\theta=45°$ directions.

$$\hat{H}(\mathbf{k}_\parallel)=\begin{bmatrix} \hat{H}^0(\mathbf{k}_\parallel)+\gamma\hat{k}_z(k_x^2-k_y^2) & \gamma k_x(\hat{k}_z^2-k_y^2)-i\gamma k_y(k_x^2-\hat{k}_z^2) \\ \gamma k_x(\hat{k}_z^2-k_y^2)+i\gamma k_y(k_x^2-\hat{k}_z^2) & \hat{H}^0(\mathbf{k}_\parallel)-\gamma\hat{k}_z(k_x^2-k_y^2) \end{bmatrix} . \tag{1}$$

$\hat{H}^0(\mathbf{k}_\parallel)$ is the conduction-band Hamiltonian neglecting spin splitting and includes the self-consistent Hartree potential. The in-plane wave vector $\mathbf{k}_\parallel=(k_x,k_y)$. Conduction-band nonparabolicity may be included in $\hat{H}^0(\mathbf{k}_\parallel)$.[16] As the spin splitting is small relative to the subband energy, we include spin splitting perturbatively. Using the fact that the mean value of k_z is zero for a bound state $\langle \hat{k}_z \rangle=0$, from a first-order perturbation of the inversion asymmetry terms in Eq. (1) on just the occupied ground state the spin splitting ΔE_s is found to be

$$\Delta E_s(\mathbf{k}_\parallel)=\pm\gamma[\kappa^4 k_\parallel^2-(4\kappa^2-k_\parallel^2)k_x^2k_y^2]^{1/2} . \tag{2}$$

As in the bulk, this spin splitting displays a large anisotropy, which now depends on the confinement through the parameter κ, as illustrated in Fig. 3.

Dresselhaus *et al.*[7] adopted a different approach and chose to ignore terms in the spin splitting corresponding to an effective magnetic field parallel to the wave vector $\mathbf{k}_\parallel$. This is achieved by considering the purely 2D band-structure problem (ignoring the confinement direction) and leads to the following expression for the spin splitting [given by Eq. (2) setting $\kappa=0$]:

$$\Delta E_s(\mathbf{k}_\parallel)=\pm\gamma k_\parallel k_x k_y . \tag{3}$$

Another contribution to the conduction-band spin splitting is the spin-orbit or Rashba term, linear in $k_\parallel$, which is caused by the macroscopic electric field of the quantum-well potential.[15] However, for the present structure this is only $\sim$0.02 meV at the Fermi level, which is negligible compared to the k^3 splitting.

The single-particle excitation spectra observed by Raman scattering are described by the electron intrasubband polarizability, which is essentially the density of states of single-particle excitations. The peak in the polarizability reflects single-particle transitions around the Fermi level from states with momentum along the direction of Raman wave-vector transfer $\mathbf{q}$. Hence, the form of the Raman-scattering peak will depend on anisotropies of the band structure, and so a rigorous comparison between experimental spectra and theory should allow a test of the above models. We perform a numerical calculation of the electron polarizability,[17] with subband dispersions given by

$$E_s(\mathbf{k}_\parallel)=\frac{\hbar^2k_\parallel^2}{2m^*}+\alpha k_\parallel^4+\beta k_x^2k_y^2+\Delta E_s(\mathbf{k}_\parallel) . \tag{4}$$

The effective mass m^* and nonparabolicity factors α,β have been obtained from self-consistent subband calculations:[16] $m^*=0.0695$, $\alpha=-2118$ eV Å^4, $\beta=-2684$ eV Å^4. These values correctly predict the Fermi velocity $v_F=1/\hbar(dE/dk)_{k=k_F}$, which is determined from the dispersion of the single-particle excitation peak.[4] For these same calculations a value of $\kappa=1.85\times10^6$ cm^{-1} was obtained.

From the electron polarizability we have calculated the single-particle spectra for in-plane wave vectors of different magnitude and in different crystallographic directions, for comparison with the experimental spectra shown in Figs. 1 and 2. There exists in the literature a wide spread of values for the spin-splitting parameter γ/eV Å^3: 17.0,[18] 27.6,[19] 20.9, 24.1, 24.5,[20] and 26.1$\pm$0.9.[7] Hence, we will use γ as a fitting parameter. Using Eq. (2) with the above calculated value of κ we obtain excellent agreement with experiment for the [10] direction, with $\gamma=23.5$ eV Å^3. However, a peak spacing is hardly resolved for the [11] direction, contrary to experiment. From Fig. 3 it can be seen that the spin-splitting anisotropy depends critically on the ratio κ/k_F.

If $\kappa = k_F/2$ the spin splitting is isotropic. Hence, if we also use κ as a fitting parameter we obtain excellent agreement with experiment for all directions, taking an electron temperature $T = 17$ K and setting $\kappa = 1.55 \times 10^6$ cm^{-1} (cf. the calculated value of 1.85×10^6 cm^{-1}) with $\gamma = 34.0$ eV Å^3. However, even taking into account the large spread among other published values for γ (see above) and the uncertainty in the calculation of κ, involving the determination of the second derivative of the wave function, which can be quite inaccurate, our fitted values are somewhat surprising. This leads us to suspect that the expression given by Eq. (2) for the spin splitting is incomplete. We find the 2D spin-splitting model[7] [Eq. (3)] is unable to describe the observed spectra. This corresponds to $\kappa = 0$ in Fig. 3, for which value there exists a large anisotropy with no splitting along [10].

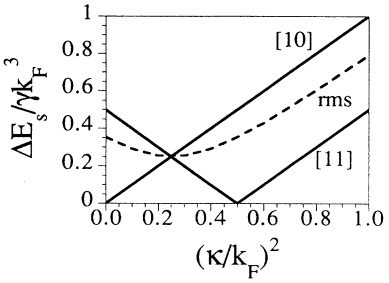

FIG. 3. The dependence of the spin-splitting anisotropy on the confinement wave vector κ. The spin splitting ΔE_s is evaluated at the Fermi wave vector k_F. Solid lines give the spin splitting for the [10] and [11] directions and the dashed line shows the variation of the average over all crystallographic directions.

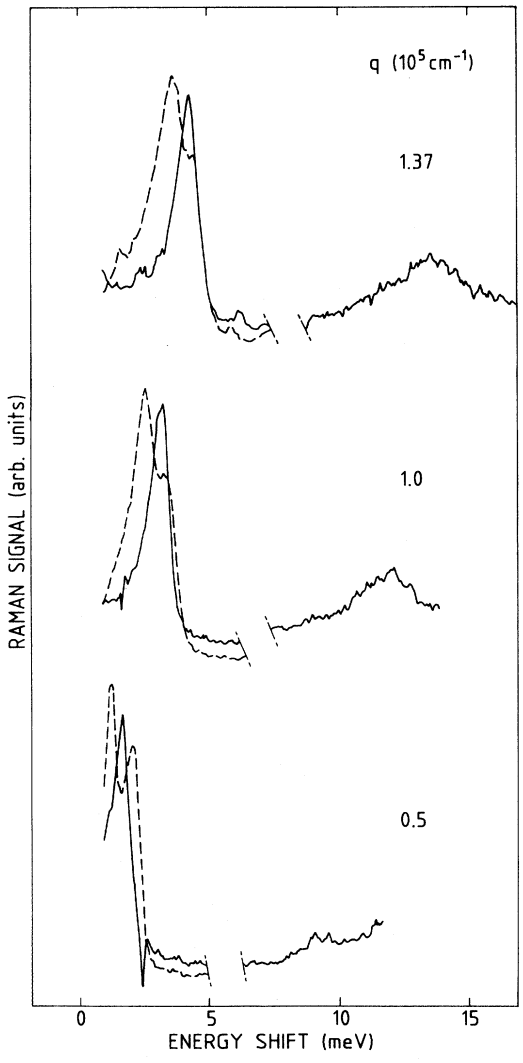

FIG. 2. Depolarized (dashed lines) and polarized (solid lines) spectra for different in-plane wave vectors **q** along [10]. The two peaks in the depolarized spectra are due to spin-flip single-particle excitations. The high-energy peak in the polarized spectra is due to the intrasubband plasmon and the low-energy peak is caused by *non-spin-flip* single-particle excitations.

Now let us turn to the polarized spectra. As can be seen from Fig. 2 there are two modes present, the dispersions of which are plotted in Fig. 4. We assign the low-energy feature in our polarized Raman spectra to non-spin-flip single-particle excitations. To support this argument we have calculated, using the same parameters as for the spin-flip excitations, the expected line shapes which gave an excellent agreement with experiment. Note also from Fig. 2 that the single-particle peak in the polarized spectra lies between the two spin-flip peaks in the depolarized spectra. In Fig. 4 the dispersions of the calculated and experimental peak positions are compared. We assign the high-energy mode in the polarized spectra to the intrasubband plasmon mode of the 2DEG. If we take the standard long-wavelength expression $\omega_p^2 = N_s e^2 q/2\varepsilon\varepsilon_0 m^*$ for the energy ω_p of a plasmon of wave vector q in a 2DEG of density N_s,[17] we find it is necessary to take a density of 1.6×10^{12} cm^{-2} to explain the experimentally observed plasmon dispersion. This is clearly at odds with measurements of single-particle excitations, intersubband plasmons, photoluminescence and

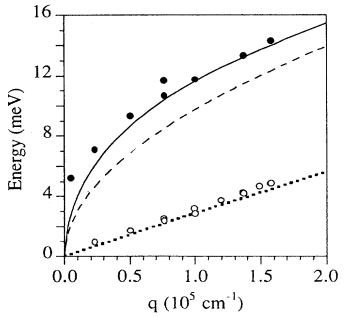

FIG. 4. Dispersion curves for excitations observed in polarized Raman spectra. Solid circles (●) give the experimentally determined plasmon dispersion. The solid and dashed lines show the calculated plasmon dispersion determined by including, and neglecting, surface image charge effects, respectively. The open circles (○) and dotted line give, respectively, the experimental and calculated dispersions of the single-particle peak.

the Hall effect, and self-consistent subband calculations, which all predict a density of $\sim 1.3\times 10^{12}$ cm^{-2}.[4,6,13] If, however, we include changes to the electron-electron Coulomb interaction due to the proximity of the sample surface (so-called image charge effects),[21] only 250 Å from the GaAs/$Al_{0.33}Ga_{0.67}As$ interface, we obtain a good fit for a 2DEG density of 1.3×10^{12} cm^{-2}. We show in Fig. 4 the calculated plasmon dispersion with and without image charge effects (solid and dashed lines, respectively). These curves have been determined from a full random-phase approximation calculation, employing self-consistently determined wave functions.[22] Plasmon energies are determined by the zeros of the dielectric response function, which depends on the intraband polarizability. The polarizability of an electron gas at small wave vectors describes excitations across the Fermi level and depends on the electron velocity there. Hence, in our calculations for the plasmon energy we have taken for the electron effective mass m^* the "velocity mass" at the Fermi level $m^*_{v_F}=0.075$, which is defined by $\hbar k_F/v_F$ where k_F and v_F are the Fermi wave vector and velocity, respectively. Taking into account the nonparabolic nature of the conduction band, we found from numerical calculations that the 2DEG intrasubband polarizability, and hence plasmon energy, is indeed determined by $m^*_{v_F}$. It should be noted that this value is considerably larger than the bulk GaAs zone center conduction-band mass of 0.0665. As can be seen in Fig. 4, the agreement for the plasmon dispersion is very good between experiment and theory, when image charge effects and nonparabolicity are included.

To conclude, we have presented a full quantitative description of Raman scattering from a single quasi-two-dimensional electron gas in a GaAs modulation-doped quantum well. We obtain spectroscopic information for different crystallographic orientations on the conduction-band spin splitting due to the lack of inversion symmetry in GaAs. Comparison with calculations of the Raman-scattering spectra gives a quantitative description of the spin-splitting anisotropy in the present sample. Calculated Raman line shapes are in good agreement with experiment and the values we determine for γ and κ are reasonable enough to support our interpretation of the Raman line splitting. However, the exact numerical values suggest that the theory we have presented here for the conduction-band spin splitting in quasi-two-dimensional electron gases is still incomplete. The intrasubband polarizability, and hence Raman line shape, may be modified by exchange-correlation effects.[12] However, we have not taken this effect into account as this correction is expected to decrease with increasing density.[23] We note that our calculations predict polarized Raman line shapes in close agreement with experiment with the only variable parameter, electron density, consistent with other independent measurements.[13] Let us finally point out that in polarized spectra we observe Raman scattering with comparable intensities from both single-particle excitations and plasmons. This is consistent with previous observations,[12,13,24] although the scattering mechanism is still not fully understood.

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