Zero-Magnetic-Field Spin Splitting in the GaAs Conduction Band from Raman Scattering on Modulation-Doped Quantum Wells

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We present Raman scattering spectra of intrasubband excitations in an n-type modulation-doped single quantum well. We attribute a double peak in the depolarized spectra to be due to spin-flip single-particle transitions. This gives direct spectroscopic evidence at zero applied magnetic field of the spin splitting of the conduction band of GaAs due to the lack of inversion symmetry in zinc-blende compounds.

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The spin-split character of the conduction band in zinc-blende compounds has attracted considerable theoretical attention for some time [1–3]. This effect arises from the polar nature of the III-V materials, eventually augmented by the existence of an electric field in the studied structures. In the moving reference frame of an electron, the total electric field is transformed into a magnetic field, which interacts with the electron spin. Experimental estimations have been presented, mostly using extrapolations to zero field of magnetotransport data [4] and, with a poorer accuracy, of electron-spin resonance [5] results. Other evidence for this splitting can be found in the spin relaxation [6] and spin precession [7] processes in bulk compounds and quantum wells. Very recently, a new observation was reported on bulk GaAs using nearly zero-field magnetoconduction measurements [3]. However, a direct spectroscopic observation in the absence of any applied magnetic field is still not available. We present in this Letter electronic Raman scattering spectra of excitations between the two spin-split conduction bands of a modulation-doped GaAs single quantum well. We thereby obtain a novel determination of the zero-magnetic-field spin splitting averaged on the Fermi surface of the two-dimensional electron gas (2DEG) which can be compared to the calculation of a similar quantity in GaAs heterojunctions by Malcher, Lommer, and Rössler [8].

Electronic Raman scattering in parallel polarizations is well known to probe the charge-density fluctuations in an electron gas. With crossed polarizations, scattering from spin density fluctuations and spin-flip excitations of the electrons are observed [9]. The Raman spectra are then directly proportional to the imaginary parts of the relevant response functions of the 2DEG [10–12]. In the absence of screening mechanisms, they simply reflect the integrals of well-defined single-particle excitation (SPE) processes within the conduction band. With crossed polarizations this will be the case if exchange-correlation effects are negligible. Processes involving spin flip can contribute only to the crossed polarization scattering, while non-spin-flip transitions are allowed in both configurations [12]. Neglecting the spin splitting, these transitions are degenerate and so the intrasubband spectra should be identical in both polarizations and display the characteristic narrow shape of the imaginary part $I(q,\omega)$ of the polarizability of a noninteracting 2DEG [13]. At $T=0$ and assuming infinite lifetimes of the electronic states, the spectra extend up to a sharp cutoff $\omega_c(q)$ which equals $v_F q$ as long as the wave-vector transfer q is much smaller than the Fermi wave vector k_F (v_F is the Fermi velocity). The corresponding electron transition is schematically shown in Fig. 1(a). Changing the angle of the sample surface normal with respect to the incident and scattered wave vectors then allows a probe of $I(q,\omega)$ over a wave-vector range extending from 0 to about 1.6×10^5 cm^{-1}. From the dependence of the Raman spectra on this angle, one thus obtains proof of the 2D character of the electron gas and from the linear dependence of the cutoff frequency on the corresponding in-plane wave vector, a determination of the electron density.

Including the k^3 terms in the conduction-band dispersion results in an anisotropic spin splitting [1]. This will not significantly affect the non-spin-flip process and one expects to observe in parallel polarization a single cutoff at $v_F q$ due to the almost identical $I_{++}(\omega,q)$ and

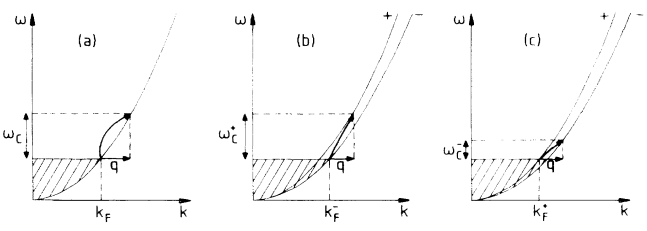

FIG. 1. Schematic representation of the highest energy processes associated, respectively, with (a) intrasubband, (b) inter-spin-split-subband, and (c) single-particle excitations with in-plane wave vector q.

$I_{--}(\omega,q)$ functions. However, in crossed polarization, new intrasubband processes should now be resolved, associated respectively with the spin-up to spin-down and the spin-down to spin-up transitions. The shape of the associated functions $I_{+-}(\omega,q)$ and $I_{-+}(\omega,q)$ is not essentially modified, except for the addition of anisotropy effects, but they exhibit two different cutoff frequencies as illustrated in Figs. 1(b) and 1(c). One obtains

$$\omega_c^{-+}(q)=\Delta E(k_F)+v_F q, \quad \omega_c^{+-}(q)=-\Delta E(k_F)+v_F q$$

when neglecting the band anisotropy and the very small difference between the Fermi wave vectors and velocities associated with both spin-split conduction bands. One thus expects to observe by Raman scattering the emergence in crossed polarizations, as compared to parallel, of two similar structures with different cutoff frequencies. These frequencies vary linearly with q with the same slope but are separated by a constant offset $2\Delta E(k_F)$. The sensitivity of Raman scattering to this quantity is therefore very high because one measures $2\Delta E/\hbar v_F q$ which exceeds $\Delta E/E_F$ by a factor k_F/q, which amounts to 100 for a typical Raman wave-vector transfer. An estimate of $2\Delta E$, averaged over the wave-vector directions in the layer plane, can be found in Ref. [8]. It is of the order of 1 meV for a modulation-doped GaAs/GaAlAs heterojunction with an electron density around 1×10^{12} cm^{-2} ($E_F\sim35$ meV) and should be therefore easily observable by electronic Raman scattering.

We performed Raman scattering experiments on a 180-Å-thick modulation-doped GaAs quantum well grown by molecular-beam epitaxy. It is followed by a 100-Å-thick spacer layer and a silicon-doped delta layer. This results in a rather large density of electrons in the quantum well (1.3×10^{12} cm^{-2}), as estimated from transport and luminescence measurements and from Raman scattering determinations of the intersubband excitations [14,15]. Because of the asymmetric doping, the shape of the self-consistent potential is very similar, up to around the Fermi energy, to that of a single heterojunction. Raman scattering experiments were performed around liquid-helium temperature with an incident photon energy in close resonance to the fundmental gap of the sample.

We present in Fig. 2(a) Raman spectra taken with crossed polarizations for several different in-plane wave vectors. They clearly exhibit two contributions with the expected line shapes for SPE transitions. Remarkably this splitting is well resolved at small wave vector. From these spectra we can get an estimate of the associated cutoff frequencies from the position of the peaks in the SPE spectrum, which we plot in Fig. 3 as a function of the in-plane wave vector. They exhibit linear and parallel variations. The parallel nature of the dispersion curves is

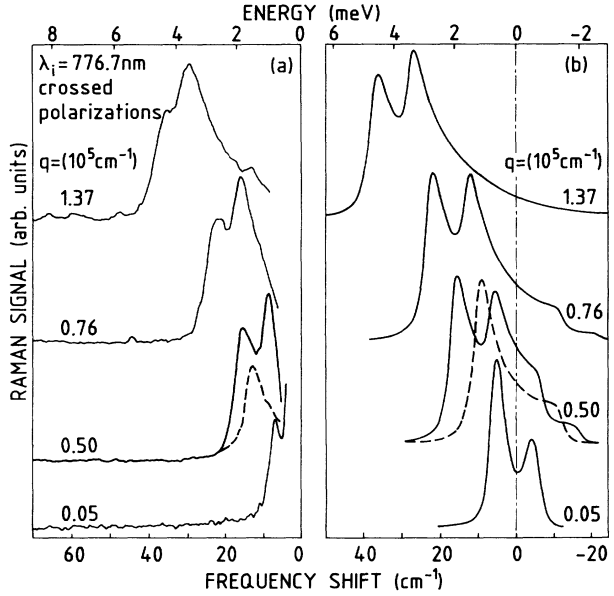

FIG. 2. Low-frequency Raman scattering spectra (a) measured in crossed polarizations for several different in-plane wave vector q and (b) calculated for the same values of q according to the model described in the text. For $q=0.5\times10^5$ cm^{-1}, we also show the Raman spectrum in parallel polarization and the calculated non-spin-flip SPE profile (dashed lines).

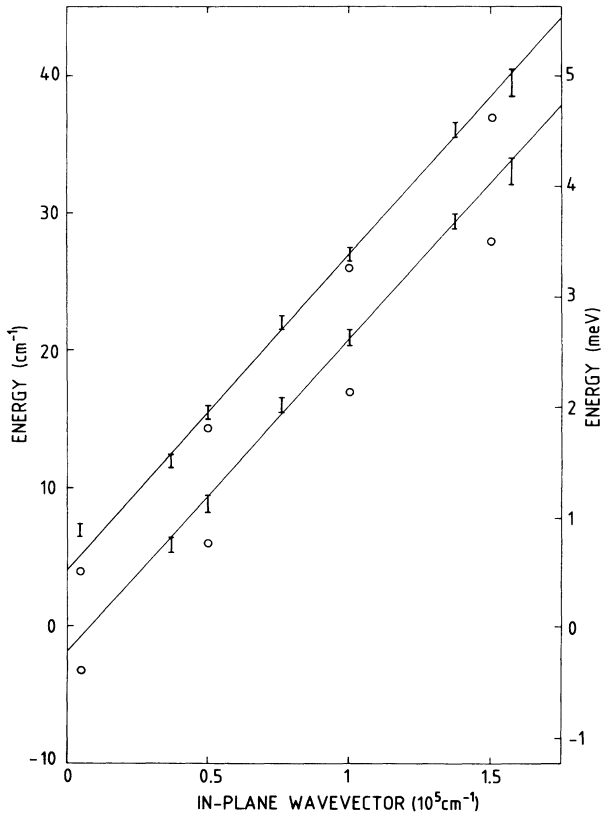

FIG. 3. Experimental (bars) and calculated (circles) spin-flip SPE peak positions shown as a function of the in-plane wave vector q. The lines are linear interpolations of the experimental results.

a very strong argument to support their assignment to inter-spin-subband transitions. Other possible origins, associated with purely intrasubband excitations, imply converging energies at very small wave vector, which is clearly not observed here. From these experimental results, one is able to extract accurate values for the slope and the distance between the two linear dispersion curves. Because of experimental uncertainties, both straight lines do not extrapolate exactly to $\pm\Delta E$ at vanishing wave vector. One deduces from these fits $\Delta E = 0.37$ meV, which is somewhat smaller than predicted in Ref. [8] (0.6 meV) for a heterojunction with a similar density, and $v_F = 4.33 \times 10^7$ cm/s. For a density of 1.3×10^{12} cm^{-2} (and hence $k_F \sim 2.86\times10^6$ cm^{-1}), we obtained from a self-consistent Poisson Schrödinger calculation (taking nonparabolicity into account [16]) $v_F = 4.39\times10^7$ cm/s and $E_F \sim 43.2$ meV in reasonable agreement with other optical results [14].

We previously attributed [17] the two lines observed in parallel polarization from the same sample to two different, strongly coupled plasmons. In this scattering configuration, only collective charge density models (plasmons) are expected to be observed, with the SPE completely screened [9]. However, this does not appear to be the case for this type of experiment: Intersubband SPE and charge-density waves have already been observed simultaneously in similar samples [10] and display comparable intensities in the sample used in this work [14]. This leads us to present a new assignment of the lowest component of the parallel spectra to the non-spin-flip SPE. It indeed displays a linear dispersion and peaks, whatever the wave vector, between the two maxima in crossed polarization with a comparable intensity. This result is illustrated in Fig. 2 for a typical in-plane wave vector. We are now able to describe all the Raman lines in both polarizations on the basis of a detailed description of the single high-mobility gas present in the sample. This further supports our evidence of spin-splitting effects.

Let us now model the different contributions to the SPE line shapes using the classical expression for the Raman signal [9] and the well-known intersubband Lindhard polarizability for a noninteracting electron gas [18]. We further introduce a phenomenological elastic lifetime of the electron states by replacing ω by $\omega + i\delta$ in the equations. The Raman cross section associated with the spin-split part of the SPE signal should then be straightforwardly obtained from a numerical integration over the Fermi disk of the two similar expressions for $I_{-+}(\omega,q)$ and $I_{+-}(\omega,q)$, depending on the spin-split conduction-band dispersion relations $E_{\pm}(k)$. In a quantum well, the displacement along z is quantized and k_z has to be replaced by its associated operator $-id/dz$. In our self-consistent Poisson Schrödinger calculation, the nonparabolicity k^4 terms were included explicitly in the conduction-band Hamiltonian [16]. This provides the 2D electron subband dispersion, parametrized by an in-plane effective mass $m^* = 0.0695$ and nonparabolicity $\alpha = -2587$ eV Å^4. The spin-split term [8] is included as a perturbation to the lowest order, k_z replaced by the average $\kappa = 1.75\times10^6$ cm^{-1} of its associated operator onto the fundamental quantum state. We neglect for simplicity the additional term associated with the average electric field in the asymmetric quantum well which was predicted to be small for large gap materials [8]. To make a comparison with our experimental results, which involve excitations from k states over all the directions in the layer plane, we further simplified the dispersion by averaging over the wave-vector direction [8]. The final energy dispersions read

$$E_{\pm} = \hbar^2 k_{\parallel}^2/2m^* + \alpha k_{\parallel}^4 \pm \gamma_0 \{k_{\parallel}^2 \kappa^4 + \tfrac{1}{8} k_{\parallel}^6\}^{1/2}.$$

We illustrate in Fig. 2(b) the typical results of our model. For the coefficient γ_0 of the spin-splitting term we have taken the value of Ref. [8] $\gamma_0 = -27.57$ eV Å^3. We also used an electron temperature of 10 K. As this parameter has only a moderate influence on the Stokes part of the Raman spectra, which we only recorded, the chosen value is only indicative. We roughly adjust the broadening parameter δ to 0.2 meV, corresponding to a lifetime of 3 ps, in good agreement with other Raman scattering results [13]. The SPE shapes obtained in these conditions look very similar to the experimental ones, thus giving further support to our interpretation. We indicate in Fig. 3 the peak positions deduced from the calculated signals. The agreement is excellent as regards the slopes and the parallel nature of the dispersions. However, the calculation overestimates the spin-splitting $2\Delta E$ by 50%. Taking into account a smaller splitting would also improve the description of the relative intensity of both SPE signals. This discrepancy could be attributed to a lack of accuracy in the previously reported values of γ_0 or to the approximations of the model, such as the in-plane averaging, or the use of the Lindhard function instead of an extension of the Mermin one [19] to spin-split transitions. In addition, inclusion of a non-spin-flip component due to spin-density fluctuations may also give better agreement. Further experiments involving different electron densities and different crystal orientations as well as quantitative estimations of the different possible Raman processes will be needed to conclude.

Moreover, the exact nature of the Raman processes involved in both intersubband and intrasubband excitations has been readdressed recently, in particular, the importance of exchange-correlation contributions to the electron-electron interactions [10,11]. For intersubband excitations one expects to observe in crossed polarizations not necessarily the SPE signal but rather the collective spin-density wave [10,11]. The intrasubband response function, and hence the Raman line shape, may also be modified, as has been indicated by a recent investigation of a low-density modulation-doped multi-quantum-well [20]. However, the exchange-correlation corrections to

the irreducible polarizability [21] are predicted to decrease strongly with increasing k_F/q. Therefore, for the high-density sample of this work, such corrections may well be negligible.

To summarize, we have presented in this Letter the first direct spectroscopic evidence in the absence of any applied magnetic field of the spin-split character of the conduction band of GaAs. It provides a unique explanation of the very unusual parallel dispersion observed in our spectra. The observation of the spin splitting was facilitated by the large 2D electron density in our sample. On the basis of sample parameters adjusted on independent experimental results and of a published value of the spin-splitting coefficient, we obtained a reasonable description of our Raman scattering results, but a significant overestimation of the spin splitting. This experiment is a new demonstration of the great power of electronic Raman scattering as a sensitive probe of semiconductor band structures. It opens the way to a more critical test of the available band-structure models and to more systematic studies on different semiconductor systems at different doping levels and eventually under magnetic field. It also points out the insufficiencies in the present understanding of the electronic Raman scattering mechanisms under strong resonance conditions.

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