ELECTRONIC RAMAN SCATTERING IN QUANTUM WELLS: COUPLED LEVELS IN TILTED MAGNETIC FIELDS

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We report on a magneto-Raman scattering investigation of free and donor-bound electrons in GaAs-Al$_x$Ga$_{1-x}$As quantum wells. For fields perpendicular to the layers, the spectra show intersubband transitions of photoexcited electrons and 1s→1s' donor excitations. Tilted fields lead to subband-Landau level and 1s'-2p$^+$ coupling. Experimental results for the latter case agree very well with variational calculations. Data on combined intersubband-cyclotron resonances at arbitrary tilt angles are accurately described by expressions valid for parabolic wells. The parabolic approach is shown to provide a good approximation in situations where coupling to higher subbands can be neglected.

In quasi two-dimensional (2D) electron systems the motions in the confinement plane (x,y) and perpendicular to it are coupled for magnetic fields $\vec{B}$ at angles $\theta \neq 0$ with respect to z.[1,2] This leads to excitations of mixed character, such as combined intersubband-cyclotron resonances, exhibiting the well-known anticrossing behavior near degeneracy.[2-7] Subband-Landau level coupling has been extensively studied for 2D systems formed at semiconductor heterojunctions[4-7] and Si-accumulation layers[3] using the technique of cyclotron resonance. For small θ's these measurements provide a determination of the energies of intersubband transitions which are forbidden at $\theta = 0$.[2,4] Small tilt angles were also used in the far infrared (FIR) experiments of Jarosik et al.[8] to study crossing of donor levels in quantum-well structures (QWS's). In this work, we report on a Raman scattering (RS) investigation of tilted-field-induced mixing in GaAs-Al$_x$Ga$_{1-x}$As QWS for both free and donor-bound electron states. Our results complement those obtained from FIR-experiments;[3-8] transitions that are weak or forbidden in FIR dominate the Raman spectra. For donor excitations, we find good agreement between the experimental results and those of calculations based on a variational approach.[9] The data on free electrons reveal coupling between the cyclotron mode and $\varepsilon_0 \rightarrow \varepsilon_1$ transitions associated with the ground and first excited subband states. This coupling has been investigated in the range of parameters where perturbation theory applies, and beyond that range. Analytical expressions derived for parabolic wells[10] were found to describe extremely well the latter results, for arbitrary θ. This unlikely situation is explained by the close similarity between the coupled-mode equations for parabolic and square wells under conditions where the coupling to higher subbands can be ignored.

A QWS grown by molecular beam expitaxy on (001) GaAs was studied. It consisted of thirty uncoupled GaAs-wells of thickness L = 460 Å, with 125-Å-thick

$Al_{0.24}Ga_{0.76}As$ barriers. The sample was doped with Si- donors at the centers of the GaAs-slabs with a concentration $N_D = 5\times10^{15}$ cm^{-3}. The width of the donor spike was ≈ L/3. Free electron excitations were investigated on the same QWS. Carriers were generated either by photoexcitation[11] or by thermal ionization of donors. The laser beam used to obtain RS data also served to photopump the sample. Power densities were in the range $P = 2 - 10$ Wcm^{-2}, giving an estimated free electron density $\rho \sim 10^{9}$ cm^{-2}.[12] RS experiments were performed at fields $B \leq 7$ T (the maximum field provided by our split-coil superconducting magnet) and at angles Θ between 0° and 80°. The laser energy $\hbar\omega_L$ was tuned to resonate with the gap derived from the $E_0+\Delta_0$ gap of GaAs. At this resonance, the scattering involving states from the conduction band is strongly enhanced.[13] Data were recorded in the $z(x',x')\bar{z}$ and $z(x',y')\bar{z}$ backscattering configurations, with x' and y' denoting the [110] and [1$\bar{1}$0] directions and with z normal to the layers. The former geometry allows scattering by charge-density fluctuations while spin-density fluctuations are allowed in the latter.[13] No appreciable differences were found between spectra in the two configurations, indicating that depolarization effects are negligible. This is consistent with our estimate for a small value of ρ.

In Fig. 1, we show Raman spectra of the QWS for different values of T, Θ, and B. Features labeled D are donor-related, which is clear from their rapid quenching with increasing temperature due to impurity-ionization (data for Θ = 30°). The strongest line at 88 cm^{-1} in the spectra for Θ = 30° is mostly due to the transition 1s→1s' (1s' denotes the lowest donor state associated with the first-excited subband), while the weaker structures at 132 cm^{-1} and 137 cm^{-1} are due, respectively, to the 1s→2s and 1s→$2p^+$ transitions. The latter excitations shift to higher energies with increasing Θ (constant B) while 1s→1s' moves in the opposite direction. The assignment of these transitions is based on the comparison with calculations considered below (for 1s→1s', see also Ref. 12). As opposed to the 1s→1s' and the RS-allowed 1s→2s transitions, the 1s→$2p^+$ transition cannot be seen for fields perpendicular to the layers (Θ = 0). The $2p^+$-level transforms like {x,y} and its Raman activity at $\Theta \neq 0$ is derived from the coupling with 1s', which transforms like {z}. Strictly speaking, neither the 1s→$2p^+$ nor 1s→1s' transitions are allowed at $\vec{q} = 0$ because the states have different parities ($\vec{q}$ is the scattering wavevector). However, it

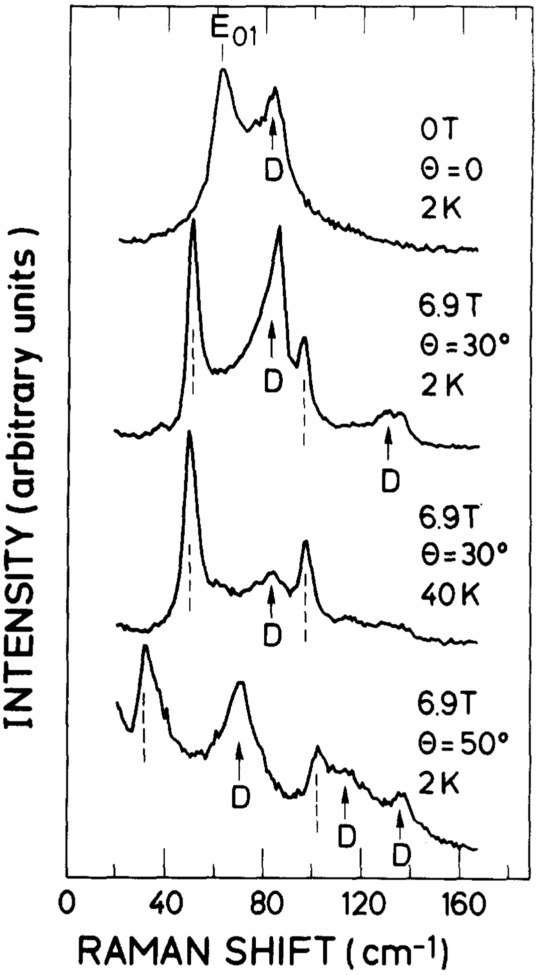

Fig. 1: Raman spectra of the structure with L = 460 Å as a function of T, Θ, and B. Features labeled D are donor-related. Dashed lines indicate the positions of the combined intersubband-cyclotron resonances. The scattering geometry is $z(x',y')\bar{z}$, $P \cong 5$ Wcm^{-2}, and $\hbar\omega_L = 1.875$ eV. Θ is the angle between the magnetic field and the z-axis of the QWS.

is a matter of fact that {z}-symmetry excitations (e.g., $\varepsilon_0\to\varepsilon_1$ intersubband transitions) can be observed in Raman spectra for $\vec{q}$ along z.[13] Mechanisms involving $\vec{q}$-dependent RS are required to account for these observations.[13]

The dashed lines in Fig. 1 indicate combined intersubband-cyclotron resonances. At 2 K, the intensity of the two

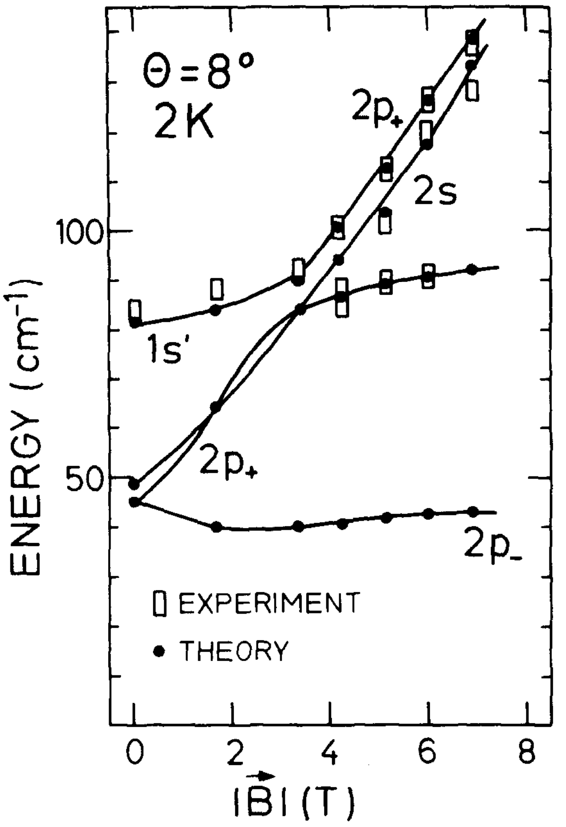

Fig. 2: Comparison between measured and calculated values of 1s→$2p^-$, $2p^+$ 2s, 1s' transition energies. The theoretical data are for a donor at the center of a 460 Å GaAs-$Al_{0.24}Ga_{0.76}As$ QWS. The solid lines are a guide to the eye.

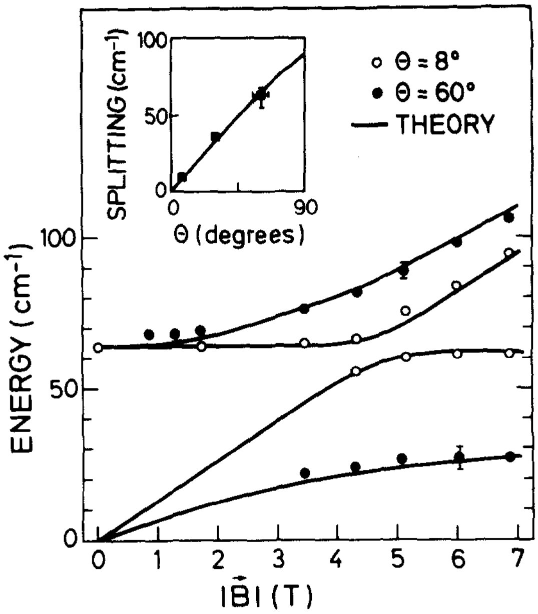

Fig. 3: The energy of the coupled inter-subband-Landau level excitations as a function of magnetic field. Θ is the tilt angle. The theoretical curves where obtained from the expressions for parabolic wells (Ref. 10). The inset shows coupled-mode splitting vs. Θ at a field of $|\vec{B}|$ = 4.8 T, which corresponds to $h\Omega_c = E_{01}$.

peaks (normalized to the intensity of optical phonons) increases roughly linearly with P as expected for scattering due to photoexcited carriers.[11,12] For Θ = 30°, the low-energy component at 53 cm^{-1} is mostly due to $\varepsilon_0 \rightarrow \varepsilon_1$ transitions while the peak at 98 cm^{-1} derives from the Δn = 1 inter-Landau level transition. This identification is supported by our results for small angles (≲ 10°), where the coupling can be neglected except near degeneracy.[2,4] For small angles, the position of the line ascribed to the cyclotron mode closely follows $h\Omega_c = ehB/mc$, with m = 0.07 m_0(Ω_c is the cyclotron frequency and m_0 is the free electron mass). The measured value of m is slightly larger than m = 0.0665 m_0, the mass for electrons at the bottom of the GaAs conduction band. The Raman shift of the line assigned to the $\varepsilon_0 \rightarrow \varepsilon_1$ transition, at small Θ's, is E_{01} = 64 cm^{-1} (it does not depend much on $|\vec{B}|$). A calculation using the band-gap discontinuities determined by Miller et al.[14] predicts E_{01} = 56 cm^{-1}, which is in reasonably good agreement with the experimental value. It is important to point out that we did not observe a peak at $h\Omega_c$ for $\vec{B}$ normal to the layers. This is unlike modulation-doped QWS which do show scattering by the cyclotron mode at Θ = 0.[15,16] We do not know of any reason that might explain this difference.

The positions of donor-related peaks, measured at Θ = 8°, are plotted in Fig. 2 as a function of $|\vec{B}|$. Also shown are results of variational calculations as described in Ref. 9, upon which our assignment of the donor transitions is based. The most interesting feature of these data is the coupling between 1s' and $2p^+$ induced by the tilted field. This is very similar to what has been reported in FIR measurements for donors at the edges of the wells.[8]

In Fig. 3, we show the measured

energies of the coupled intersubband-cyclotron modes for two different tilt angles. Level-repulsion is evident in the figure. $\theta = 8°$ is within the range where perturbation theory can be applied. The prediction[2,4] of a splitting proportional to θ for $\hbar\Omega_c = E_{01}$ has been confirmed by our experiments (see the inset). Departures from results of perturbative calculations can be seen in the data of the inset at large angles and in the results for $\theta = 60°$. In particular, we find that the lower branch of the coupled modes does not approach E_{01} for large B's, but deviates considerably from that value. The theoretical curves in Fig. 3 were obtained from expressions valid for parabolic wells.[10] The agreement between theory and experiment is quite remarkable. This is also the case for the asymptotic behavior of the lower branch which is predicted[10] to tend to $E_{01}\cos\theta$ at high fields. The reason why a square well can be approximated by a parabolic one is basically due to the fact that the corresponding ground and first-excited subband states are very similar. The matrix elements involved in the coupled-mode problem for the two cases differ by less than 2%. Hence, one can expect that parabolic solutions will apply to square wells if the coupling to higher subbands is not important. A detailed analysis of the coupled equations indicates that this condition is well-fullfilled for the range of parameters in our experiments.

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