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Multiple cyclotron resonances in GaAs-Al$_x$Ga$_{1-x}$As quantum wells detected by resonant inelastic light scattering

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Cyclotron resonance excitations in modulation-doped Al$_x$Ga$_{1-x}$As-GaAs quantum wells have been investigated by means of resonant inelastic light scattering. Besides the cyclotron resonance excitation $\hbar\omega_C$, we observe up to fifteen additional peaks at multiples of $\hbar\omega_C$. The constant energy separation between these resonances indicates that they are multiple excitations of $\hbar\omega_C$ (and not higher harmonics). These multiple resonances all have nearly the same strength. For an explanation, we propose a cascade model, where the photocreated hole in the valence band couples via direct and exchange Coulomb interactions to the Fermi sea. This allows the hole to relax resonantly by creating cyclotron resonance excitations in the conduction band. [S0163-1829(97)02827-0]

Although resonant inelastic light scattering has proven to be a powerful tool in the investigation of electronic excitations in two-dimensional (2D) electron systems, there are only a few Raman investigations with application of external magnetic fields. In 1987, Pinczuk *et al.* reported the observation of cyclotron resonance and combined intersubband inter-Landau-level excitations in multiple-quantum-well structures.[1] In these resonance experiments, where photon energies in the range of the E_0 gap have been used, the usually Raman forbidden inter-Landau-level excitations could be observed due to relaxation of parity selection rules by heavy-hole-light-hole mixing at the E_0 gap. Pinczuk *et al.* observed the roton density of states in two-dimensional Landau-level excitations applying very strong magnetic fields.[2] Brozak *et al.* reported the observation of combined intersubband inter-Landau-level excitations of collective spin-density and charge-density type. In their investigations at the $E_0+\Delta$ gap, the breakdown of selection rules has been achieved by a tilted magnetic field with respect to the sample normal.[3] In this paper we report the observation of multiple cyclotron resonances in 2D electron structures. To explain our experimental results, we propose a cascade model very similar to the model that was used to explain the multiphonon Raman scattering in CdS bulk material.[4–6] Recently, Stühler *et al.* observed multiple spin-flip Raman excitations in semimagnetic Cd$_x$Mg$_{1-x}$Te-Cd$_x$Mn$_{1-x}$Te quantum wells.[7]

Our investigations were performed on high-quality one-sided modulation-doped single quantum wells with a width of 25 nm. The GaAs well was separated from the Si-doped top barrier by a 20-nm-wide Al$_x$Ga$_{1-x}$As spacer. The carrier densities under illumination were in the range $(4.0-7.5)\times10^{11}$ cm^{-2}. The mobilities were about 7×10^5 cm^2 V^{-1} s^{-1}. At a temperature $T=2$ K, only one subband in the quantum wells was occupied. For the inelastic light scattering experiments with magnetic fields up to 10 T, glass fiber optics have been used.[9] With a microscope objective the light from a Ti:sapphire laser was coupled into a fiber with a 110 μm core diameter. The fiber guided the laser light into a 10 T axial magnet cryostat. Within the axial magnet the light was focused onto the sample via a graded-index (GI) lens. The sample was immersed in liquid superfluid helium at a temperature of 2 K. Another GI lens collected the scattered light and coupled it into a glass fiber with a 600 μm core diameter. The scattered light was analyzed in a triple Raman spectrometer and detected by a charge coupled device camera. Since the multimode glass fibers do not conserve polarization, all experiments in the axial magnet system were made using unpolarized light. For polarization dependent measurements, a 8.5-T optical split-coil magnet system has been used. The photon energies of the Ti:sapphire laser were in the range of transitions from various confined hole states to the Fermi energy in the 2D conduction-band ground state at the E_0 gap of the quantum-well structure. The power density was about 20 W cm^{-2}.

Figure 1 shows measured Raman spectra of a quantum-well sample in dependence on the magnetic field. At $B=0$ T, we observe no excitation in the displayed energy range. At $B=1$ T, twelve equally spaced peaks occur within the detected range where their energetic positions exactly differ by $\hbar\omega_C$. The numbers n above the peaks indicate the number of cyclotron quanta that contribute to the observed excitations. As a guide to the eye, dotted lines are drawn, which connect peaks with the same n. For Raman shifts $\geqslant 22$ meV the cyclotron excitations are obscured by the much stronger 2D intersubband excitations and luminescence. At magnetic fields larger than 2 T, also the combined intersubband inter-Landau-level excitations $E_{01}-\hbar\omega_C$ and $E_{01}-2\hbar\omega_C$ can be observed (indicated by arrows in Fig. 1). E_{01} is the 2D intersubband spacing. For a closer look, Raman spectra in the range $B=0-1$ T are displayed in Fig. 2. The multiple resonances can be resolved for magnetic fields $B>0.3$ T. These

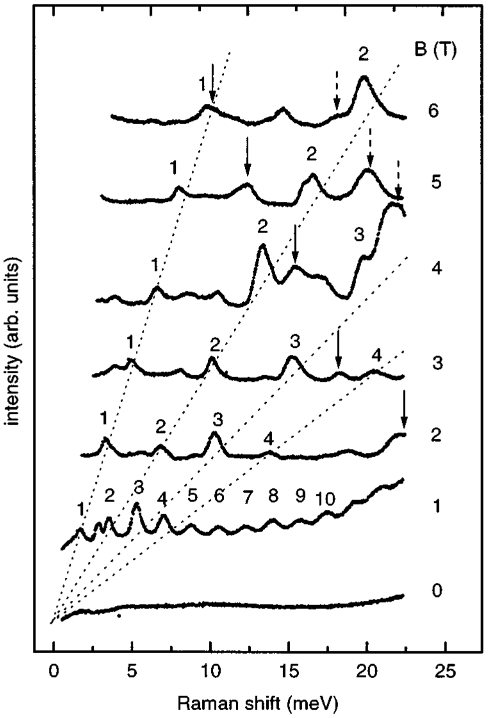

FIG. 1. Experimental cyclotron resonances in a one-sided modulation-doped single-quantum-well structure with 25 nm well width and carrier density $n_s = 7.5 \times 10^{11}$ cm^{-2}. The spectra are shown in dependence on the magnetic field. The laser energy was $E_L = 1570$ meV. The numbers n label the number of excited cyclotron quanta. The dashed and solid arrows indicate the combined intersubband inter-Landau-level transitions $E_{01} - \hbar\omega_C$ and $E_{01} - 2\hbar\omega_C$, respectively.

spectra (polarized spectra) have been recorded using the optical magnet cryostat with well-defined polarization directions of the incident and scattered light. In the experiments a finite wave vector $q = 0.8 \times 10^5$ cm^{-1} was transferred parallel to the plane of the 2D electron gas.

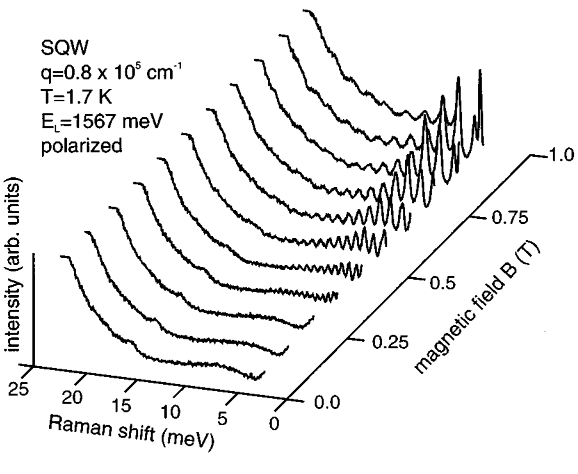

FIG. 2. Polarized Raman spectra of the same sample as in Fig. 1 for small magnetic fields $B = 0 - 1$ T.

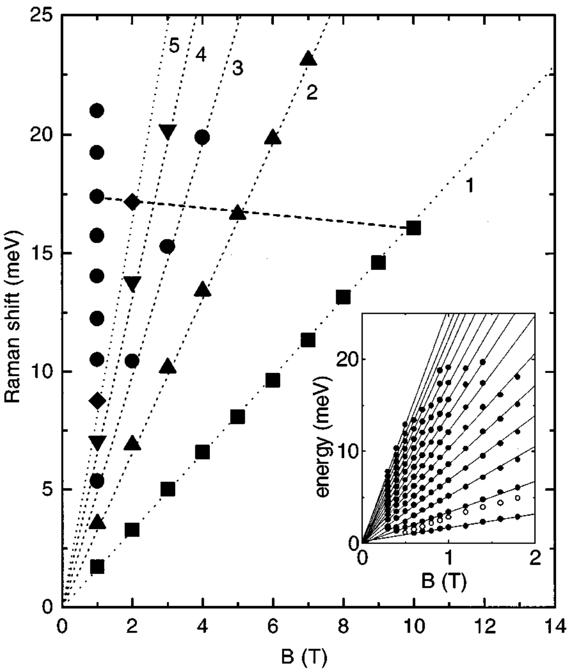

FIG. 3. Magnetic-field dispersion of the multiple cyclotron resonance excitations. The numbers n label the same excitations as in Fig. 1. We note that the energy of the first cyclotron resonance at $B = 10$ T is about 1.2 meV lower than the tenth resonance at $B = 1$ T. This is due to the increase of effective mass with the magnetic field.

We find that the cyclotron resonance excitations occur in the depolarized as well as in the polarized scattering configurations, where the observed spectra do not differ significantly in both polarizations. This indicates the single-particle character of the observed excitations. This single-particle character is also emphasized by the observation that the excitations show strong resonant behavior in the vicinity of the E_0 gap. If the laser energy is well above the effective band gap of the quantum well, only one single cyclotron resonance peak at $\hbar\omega_C$ is observed. The multiple resonances occur under conditions of extreme resonance in a range of laser energies, where in the quantum-well samples as well as in laterally structured samples, at $B = 0$ T, so-called single-particle excitations can be observed.[10]

The magnetic-field dispersion of the cyclotron resonances is shown in Fig. 3. The solid dots in the inset show the observed resonances in the range $B = 0 - 2$ T. The observed peaks, which correspond to the open symbols in the inset, can be attributed to impurity-related resonances from the GaAs buffer layer. The dotted lines in Fig. 3 represent the calculated cyclotron resonance dispersions $n\hbar\omega_C = n\hbar(eB/m^*)$, where the effective mass $m^* = 0.071$ m_e and $n = 1,2,3,\ldots$. The deviation from the linear dispersion relation in the range 0–10 T is about 9% and is due to the increase of effective mass with magnetic field. It is emphasised that for a fixed magnetic field, the energy spacing between the multiple resonances is the same to an accuracy of 1.5%. For clarity, the dashed line in Fig. 3 connects the tenth point at 1 T with the first point at 10 T. If the multiple resonances were higher harmonics, i.e., inter-Landau-level transitions with a change in Landau-level quantum number $\Delta l > 1$, the dashed line should be horizontal. This leads to the conclusion that the multiple cyclotron resonances all arise from Landau-level transitions with $\Delta l = 1$. On the other hand,

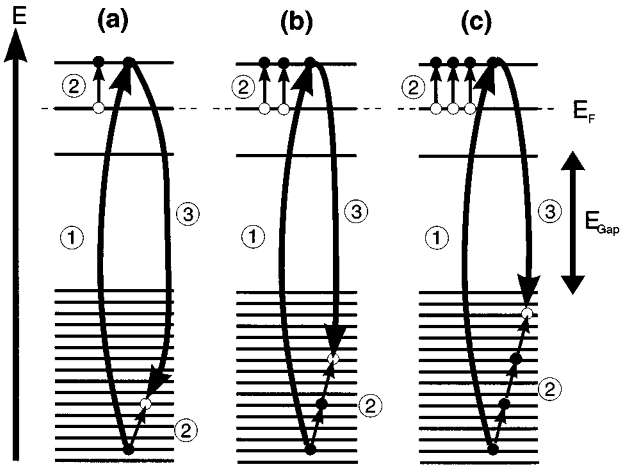

FIG. 4. Sketch of the proposed cascadelike scattering process, where (a) one, (b) two, or (c) three cyclotron resonance excitations are created.

if higher-order Raman scattering would be the reason for the observed peaks, the intensity of the $n=2$ excitation should be at least one order of magnitude less than the intensity of the first cyclotron resonance peak. Obviously, this is not the case. We suggest that the multiple cyclotron resonances are not created simultaneously, but in a cascade process. In one step of this process, the photoexcited electron hole pair is scattered via an interaction with the Fermi sea by creating a cyclotron resonance excitation. The number of cyclotron resonances created in one cascade seems to be arbitrary. Energy conservation is provided by the almost continuous distribution of hole states in the valence band and thus all the scattering processes can take place via real transitions. One necessity for such cascades to occur is that the time for creation of a cyclotron resonance excitation is much shorter than the lifetime for radiative recombination. Figures 4(a)–4(c) show a schematic picture of the proposed scattering process: In the first step (1) an electron-hole pair or exciton is created by absorption of the incident photon. In the second step (2) the hole is scattered into another state due to the Coulomb interaction with the Fermi sea, where during the scattering process a cyclotron resonance excitation in the conduction band is created. We assume these two steps of the scattering process to be very similar to the commonly known excitonic scattering mechanism proposed by Danan *et al.*[8] for electronic Raman scattering by 2D intersubband excitations. In the third step (3) the electron recombines with the scattered hole. The important point is that due to the almost continuous distribution of real states in the valence band at finite magnetic field, the hole may be scattered n times, where all intermediate states in the scattering or relaxation process may be real, before it recombines. During these n scattering processes, n cyclotron resonance excitations can be created in the conduction band by energy conservation [Figs. 4(b) and 4(c)]. In principle, the hole can resonantly relax until it reaches the top of the valence-band. To demonstrate the plausibility of this scattering process, we have calculated the splitting of the valence band states for an asymmetric quantum-well sample in a magnetic field. The results, plotted in Fig. 5, show that, due to heavy- and light-hole mixing, there are a large number of closely spaced Landau levels, which form a quasicontinuum of real states. The hole Landau levels are calculated within the axial approximation of a 4×4 Luttinger Hamiltonian.[11] As an input we have used the potential that has been calculated self-consistently within the local-density approximation of the Γ_6 conduction band. For a fixed magnetic field B, Fig. 5 displays the ten lowest eigenvalues for each Landau-level index N, ranging from -2 to 18.

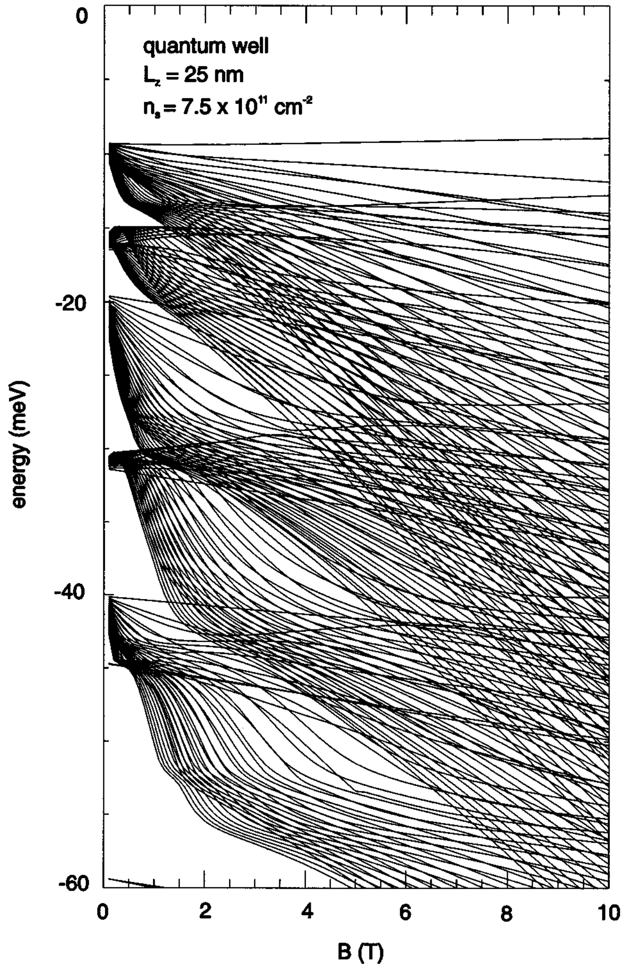

FIG. 5. Hole states in the axial approximation with Landau-level index $-2\leqslant N\leqslant 18$ for a quantum-well structure with electron density $n_s=7.5\times10^{11}$ cm^{-2}. We have used the Luttinger parameters $\gamma_1=6.85$, $\gamma_2=2.1$, and $\gamma_3=2.9$ for both GaAs and Al$_x$Ga$_{1-x}$As. For each N the ten lowest eigenvalues are displayed.

We like to note that in the past there was some controversy about whether or not the resonant multiphonon Raman scattering in CdS bulk material must be interpreted in terms of hot luminescence.[12,13] This point, which might be only a question of semantics,[14] is still not clear. Therefore, it may be regarded as an open question whether or not our spectra of multiple cyclotron resonance excitations have to be interpreted as hot luminescence.

In conclusion, we have observed multiple cyclotron resonance excitations in single-quantum-well samples by resonant Raman spectroscopy. The results could be reproduced in different samples with different densities in the range

$(4.0-7.5)\times10^{11}$ cm^{-2}. For explanation, we propose a cascadelike scattering mechanism where the photoexcited hole is resonantly scattered by exciting cyclotron resonance excitations in the conduction band.

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