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Double Raman resonances by light and heavy magneto-excitons in GaAs/AlAs multiquantum wells

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Raman resonance profiles have been measured in GaAs/AlAs multiple quantum wells (MQW's) under an intense magnetic field perpendicular to the layers. We find a strong enhancement of the peak intensities when the difference between the incoming and outgoing transition energies are tuned by the field B to equal the GaAs LO-phonon. This result is interpreted as double Raman resonances involving either light-hole or heavy-hole magneto-excitons which differ in 2 units of the Landau index. These observations are better understood by assuming an exciton–phonon scattering mechanism, instead of the usual free electron–phonon interaction.

The usefulness of Raman spectroscopy for the investigation of the vibrational properties of low-dimensional systems has been widely demonstrated during the last decade. Resonant Raman scattering (RRS) by phonons has also been successfully applied to study the superlattice (SL) and multiquantum-well (MQW) electronic structure [1,2]. Recently, there is a growing interest in the mechanisms underlying these resonances, which take place whenever the incident or scattered light has the precise energy of an electronic interband transition of the system. Incoming and outgoing resonances can occur simultaneously when two of these transitions differ in energy that of one phonon. The study of the resulting double resonances [3–9] can help in the elucidation of the scattering mechanisms.

Conditions for doubly resonant Raman scattering (DRRS) can be attained by the proper choice of well dimensions in MQW structures, and DRRS due to the deformation potential mechanism [3] as well as induced by the Fröhlich interaction [4] have been reported. Else, DRRS can be produced by changing external conditions. This has been observed either in SL's in an external electric field [5] and in bulk materials under uniaxial stress [6] or magnetic fields [7,8]. In particular, the application of a magnetic field splits the continuum of states into discrete Landau levels (LL). Optical transitions between valence and conduction Landau states are allowed provided that the Landau index is conserved. A detailed theoretical study on magnetic-field induced DRRS, in the framework of the uncorrelated electron–hole approximation, can be found in ref. [9].

New interesting effects are expected when the DRRS is induced by a magnetic field in 2D systems [10], where excitonic effects are known to play an important role [11]. We report new observations of magnetic-field induced DRRS in GaAs/AlAs MQW's. Our results support the idea

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that excitons are scattered as a whole by the phonons in the Raman process.

We have studied MQW samples grown by MBE, consisting of 30 periods with 100 Å thick wells and barriers, as determined by X-ray diffraction. Photoluminescence (PL), PL excitation (PLE) and RRS measurements were performed using light from an LD-700 dye laser and a double monochromator. The sample was immersed in a liquid He bath-cryostat within a standard-coil superconducting magnet. Magnetic fields up to 13.5 T were applied in the Faraday geometry. Circular polarization configurations ($\sigma^{\pm}$, $\sigma^{\pm}$), in the laboratory frame, were obtained by means of achromatic $\lambda/4$ plates.

In order to determine the energy and character of the magneto-optical transitions which may participate in a DRRS, we have measured the PLE spectra for different fields in the four polarization configurations. In each spectrum, two sets of transitions can be distinguished, involving either heavy- or light-hole states, whose energies are displayed in fig. 1 for the $\sigma^+\sigma^+$ configuration. The heavy or light character is determined by the selection rules for absorption and emission of circularly polarized light [12,13]. H_0 and L_0 transitions are almost field independent due to

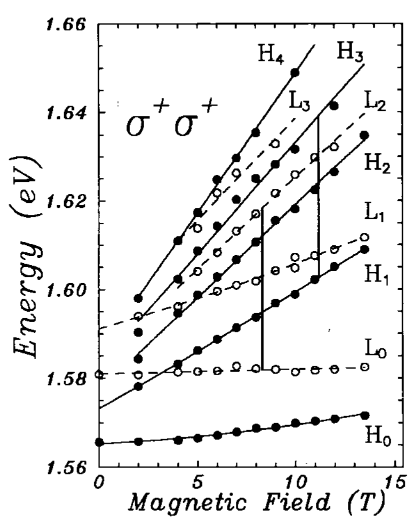

Fig. 1. Energies of the main magneto-optical transitions for a 100 Å GaAs–100 Å AlAs MQW structure as a function of magnetic field, from PLE experiments in the $\sigma^+\sigma^+$ polarization configuration.

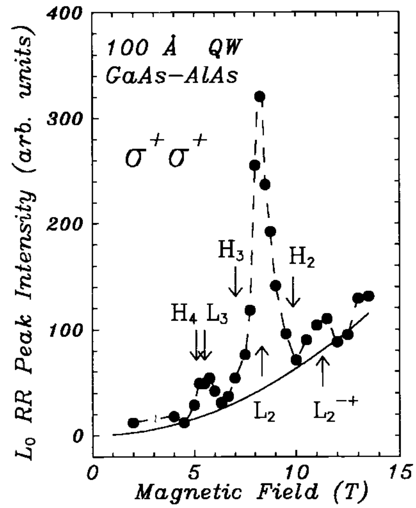

Fig. 2. Maximum intensity of the L_0 outgoing resonance vs. magnetic field, for the $\sigma^+\sigma^+$ polarization configuration. The arrows correspond to expected double resonances with the incoming transition H_n or L_n, taken from fig. 1. The solid line shows a B^2 law for the resonant background.

their marked excitonic character; actually, they correspond to the ground states of the heavy- and light-hole excitons, h(1s) and l(1s), respectively. The remaining transitions ($n \geq 1$), though still influenced by excitonic effects [14], can be described as transitions between electron- and hole-Landau states, since their energy vary almost linearly with B. The different slopes observed in fig. 1 allow to search for DRRS conditions, by tuning the energies of the LL's to differ in the GaAs LO-phonon energy ($\simeq$ 36 meV). The vertical bars in fig. 1 indicate some magnetic fields and the corresponding light frequencies at which DRRS are expected.

To look for magnetic-field induced DRRS we first choose an outgoing transition and then measure the full Raman resonances, as a function of the incoming energy, for different values of B [10]. Since the strong luminescence at H_0 prevents the distinct observation of the Raman signal, we have selected L_0 and H_1 as outgoing transitions. The respective intensities of these resonances are displayed in figs. 2 and 3, for the $\sigma^+\sigma^+$ configuration, as a function of B. The arrows indicate the values of B at which double resonances could be expected, for the indicated incoming channels, according to the energies observed in the PLE data of fig. 1.

Consequently, the strong peak at 8.2 T in fig. 2 is interpreted as a DRRS in which the incident photon produces the excitation L_2, while the outgoing photons originate from the recombination L_0 of the electron and light-hole ground states. This process involves a $\Delta n = 2$ change of the LL index between the incoming and outgoing transitions, whereas n is conserved in each of them. Similarly, a clear DRRS is obtained at 11.2 T for the H_1 outgoing resonance (fig. 3), when the incoming channel is resonant with the H_3 heavy-hole transition. Again, we find $\Delta n = 2$ in the dominant process. Smaller peaks, at 5.5 and 6.3 T in figs. 2 and 3, respectively, are found to correspond to the DRRS L_3–L_0 and H_4–H_1, both with $\Delta n = 3$. The small peak labelled L_2^{-+} in fig. 2 corresponds to the L_2–L_0 DRRS in the $\sigma^-\sigma^+$ configuration [10], which appears in the $\sigma^+\sigma^+$ one due to polarization leaks. In addition to DRRS, it is apparent from fig. 2 the increasing background, which can be described by a B^2 dependence as predicted for bulk GaAs [9].

The $\Delta n = 2$ change observed in the DRRS processes could be related to the mixing of light- and heavy-hole states and the variation $\Delta J_z = 2$ which takes place in the Raman process for bulk material [9,10], but further theoretical effort is

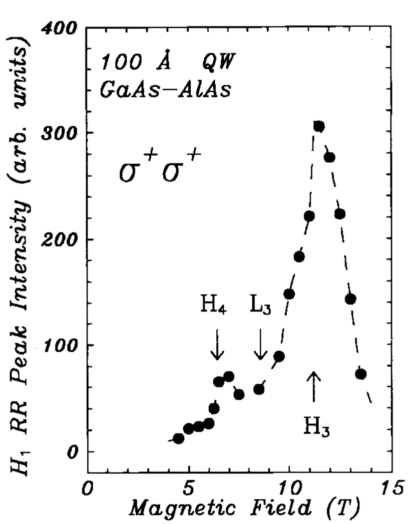

Fig. 3. Same as fig. 2 for the H_1 outgoing resonance.

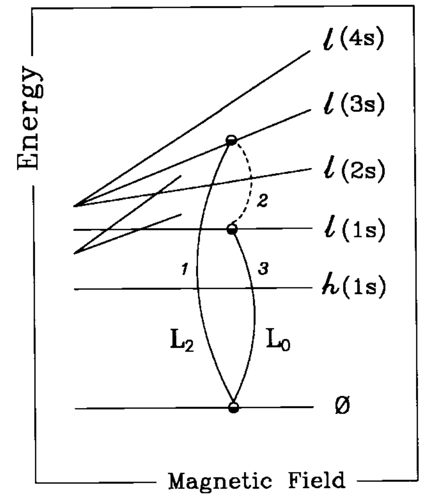

Fig. 4. Three-band scheme of the processes involved in the L_2–L_0 DRRS: L_2 excitation, exciton scattering by LO phonons and L_0 recombination. ∅ denotes the vacuum state of excitons.

needed to fully understand the present results. One can nevertheless make the following considerations. If n is to be conserved in the optical transitions, the sequence of individual interband and scattering processes in the independent particle picture would be: creation of the e–h pair in bands e_2 and l_2, relaxation of both free carriers to their respective ground states, and recombination of the e_0–l_0 pair. In this scheme, a DRRS event would require the simultaneous relaxation of the electron and the hole, sharing the phonon energy. A different and more likely process is proposed and illustrated in fig. 4, where only excitonic states are involved. Here the exciton is scattered as a whole by the phonon, and DRRS takes place between states of the same exciton: l(3s) and the ground state l(1s) for fig. 2 and h(4s) and h(2s) for fig. 3.

In summary, we have observed double Raman resonances between states of the same (heavy or light) exciton differing in two units their principal quantum number. The results point at the exciton–phonon coupling as the scattering mechanism. Though the experiment by itself can give very important information, our understanding of the processes and mechanisms involved in the observed DRRS will not be satisfactory until a theoretical description is developed.

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