

Double Raman resonances induced by a magnetic field in GaAs-AlAs multiple quantum wells

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A strong enhancement in the intensity of the Raman resonance profile has been observed in GaAs-AlAs multiple quantum wells, when the energies of different transitions are tuned by an external magnetic field. This finding is interpreted as double Raman resonances corresponding to some excited states and the ground state of the light-hole exciton as incoming and outgoing channels, respectively. Our results are best understood by assuming an exciton-phonon scattering mechanism instead of the usual free-electron-phonon interaction.

To date, Raman scattering studies of undoped semiconductor superlattices (SL) and multiple quantum wells (MQW) have mainly been devoted to the vibrational modes resulting from the new periodicity.¹ Raman spectroscopy has also proved to be a very sensitive technique to study the electronic structure of low-dimensional systems, due to the resonant enhancement of the Raman signal whenever the energy of the incident or the scattered light equals that of an electronic transition of the system. The way in which photons are scattered by phonons in a resonant Raman scattering (RRS) process has been a matter of controversy in the last decade,² and different mechanisms, involving either excitonic effects³ or impurities,⁴ have been invoked to explain the observed asymmetries between the incoming and outgoing channels in the resonant profiles. The study of doubly resonant Raman scattering (DRRS), which occurs when both the incident and the scattered photon energies are resonant with electronic transitions of the system, may contribute to clarify some aspects of these problems. On the other hand, excitonic effects could play an important role in DRRS, as they do in the other optical properties of two-dimensional (2D) systems.^{5,6}

DRRS has been observed in bulk GaAs when its band structure was altered externally by an uniaxial stress,⁷ in a way that the energy splitting of the heavy- and light-hole states matched the LO phonon frequency. In 2D systems, like SL's and MQW's, conditions for DRRS have been achieved by the proper choice of well dimensions^{8,9} and by means of an external electric field.¹⁰

The application of an external magnetic field allows us to adjust the energy difference between light- and heavy-hole Landau levels to the LO phonon energy, thus leading also to DRRS conditions. It has the advantage, compared to the use of electric fields, that it does not perturb the original band structure of the system, and only splits the continuum states into discrete Landau levels. Magnetic-field-induced DRRS have recently been ob-

served in bulk GaAs,¹¹ and explained by a detailed theory on the electron-phonon coupling.^{12,13}

In 2D systems, the lower dimensionality of the density of states, further reduced by the magnetic field, produces stronger singularities of the response, enhancing the sensitivity of RRS for the observation of interband transitions. Therefore DRRS are expected to be more intense for MQW than for bulk GaAs. In this work we report on the observation of magnetic-field-induced DRRS in GaAs-AlAs MQW, and discuss its possible implications in the scattering mechanisms.

We have investigated GaAs-AlAs MQW's with different layer thicknesses grown by molecular beam epitaxy on (100) undoped GaAs substrates. The periodicity and homogeneity of the samples were controlled by x-ray diffraction. Photoluminescence (PL), PL excitation (PLE), and RRS measurements were performed at 2 K. Magnetic fields up to 13.5 T were applied in the Faraday configuration, with B along the growth axis. The incoming light was provided by a LD-700 dye laser, and for the detection a double monochromator with standard photon-counting electronics was used. The different circular-polarization configurations (σ^\pm, σ^\pm) were obtained by means of achromatic $\lambda/4$ plates in the incoming and outgoing light paths, where $+$ and $-$ refer to the fixed direction of the magnetic field (i.e., we use the laboratory frame to define the helicity of the light).

The PL and PLE spectra in a crossed configuration ($\sigma^- \sigma^+$) for a GaAs-AlAs MQW structure with 100-Å-thick wells and barriers, are shown in Fig. 1 (solid curves) together with the RRS profile (dotted curve), for $B = 10$ T. All the spectra are plotted in a logarithmic scale, with rigid vertical displacements for a clear display. The peaks labelled H_n (L_n) are associated with transitions between the n th Landau electronic state and the n th heavy- (light-) hole state. Note the emission at higher energies than the main luminescence band (H_0) originating from higher magneto-transitions (L_0, H_1, L_1), which is favored

by the magnetic field.¹⁴

The use of circularly polarized light allows for the separation of transitions between states with different spin orientations, thus leading to well-established selection rules.¹² This is clearly observed in Fig. 1, where the PL transitions (selected by the outgoing polarization σ^+) and those observed in PLE (governed by the incoming polarization σ^-) show the Zeeman splittings of the ground states. The inset of Fig. 1 explains why the L_0 emission ($e_{+1/2}$ - $lh_{-1/2}$) is seen at lower energy than its corresponding excitation ($lh_{+1/2}$ - $e_{-1/2}$). This is not observed in the H_0 transition, because its Zeeman splitting at 10 T (≈ 2 meV) is smaller than the Stokes shift (4 meV). The resonant profile, plotted versus the scattered photon energy, shows the same transitions as the PL emission. This coincidence clearly indicates the dominance of the outgoing channel over the incoming one.

The ideal DRRS event would involve two Landau states, associated with the heavy- and light-hole subbands, with the *same* Landau number n , differing in energy by the GaAs LO phonon (about 36 meV). Since the difference $E(L_n) - E(H_n)$ decreases with the magnetic field, the light-hole-heavy-hole splitting at zero field should be larger than the phonon energy. This condition imposes an upper limit on the well width ($L_z \leq 45$ Å) for the observation of DRRS involving the GaAs LO phonon. However, in QW's of these dimensions, the inhomogeneous broadening of the transitions is usually an impor-

tant fraction of the light-hole-heavy-hole splitting, so that it is expected to prevent the clear observation of DRRS. Therefore, we have chosen to investigate the feasibility to obtain DRRS involving electronic transitions between states with different Landau numbers in wider QW's. For this purpose we have concentrated on a sample containing 30 GaAs wells with $L_z = 100$ Å, separated by 100-Å AlAs barriers. In the absence of field one can roughly estimate that three electron states are confined in the quantum wells, at energies close to 50, 200, and 500 meV above the bottom of the conduction band, the fourth one being close to the barrier top.

A theoretical prediction of the conditions for DRRS involving magneto-optical transitions would require a cumbersome numerical calculation. On the other hand, this information can be also obtained directly from experiment. Therefore, as a previous step to the RRS experiments, we have measured the pseudoabsorption spectra for different magnetic fields between 0 and 13.5 T by PLE spectroscopy, in order to identify the transitions.¹⁵ Their energies are plotted in Fig. 2 for the $\sigma^- \sigma^+$ configuration. For a given polarization of the exciting light, the observed diamagnetic shifts of the different transitions depend strongly on the character of the involved states. The H_0 and L_0 transitions hardly shift with the field due to their strong excitonic character;⁶ actually, they correspond to the ground ($1s$) states of the heavy- and light-hole excitons, respectively. On the other hand, the ener-

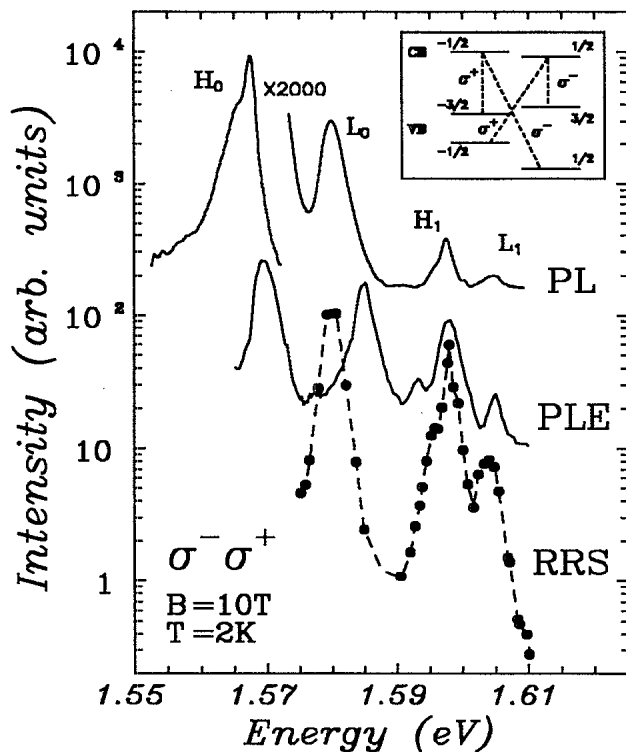


FIG. 1. Comparison of PL, PLE, and outgoing RRS for a 100 Å GaAs-100 Å AlAs MQW under magnetic field (10 T), in the $\sigma^- \sigma^+$ configuration. H_n, L_n refer to transitions between the n th Landau states.

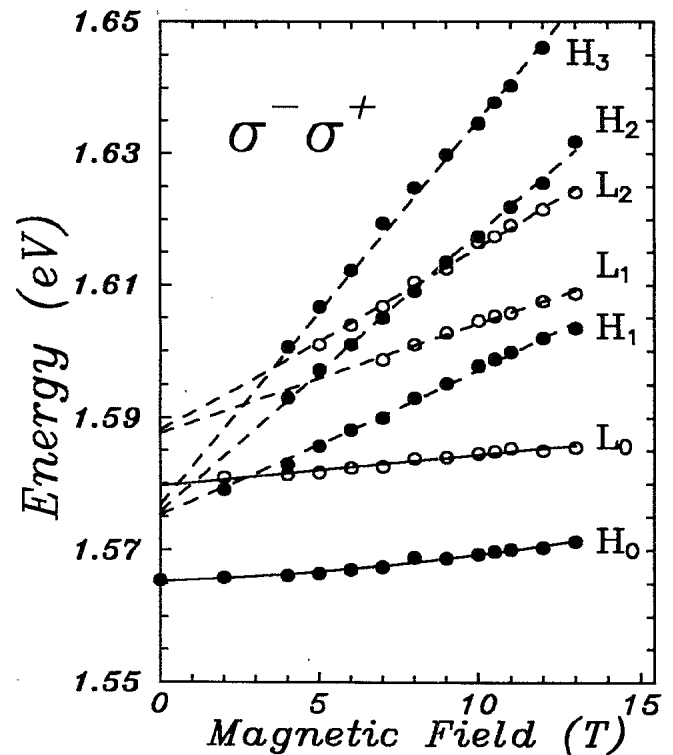


FIG. 2. Energies of the main magneto-optical transitions for a 100 Å GaAs-(100 Å AlAs) MQW structure, from PLE experiments in the $\sigma^- \sigma^+$ polarization configuration, as a function of magnetic field.

gies of higher transitions ($n \geq 1$), which are also influenced by excitonic effects, vary almost linearly with B for moderate fields, so that it is a good approximation to describe them as transitions between electron and hole Landau states. The magnetic-field dependence of the different states provide an easy method to produce DRRS conditions, by tuning the energies of the Landau levels with the magnetic field.

A way to investigate DRRS is to fix the laser energy, set the monochromator to the position of the Stokes Raman phonon, and vary the strength of the applied magnetic field, as was done in Ref. 11. Performing these experiments we have obtained magneto-Raman oscillations corresponding to incoming and/or outgoing resonances. However, this procedure does not allow us to distinguish between the true Raman enhancement and changes in the PL background. Actually, the presence of hot luminescence due to high-energy transitions,¹⁴ with maximum intensity for DRRS conditions, makes the interpretation of the results difficult. Consequently, we have measured the full Raman resonances as a function of the laser energy for different values of B .

The outgoing channel was set at the L_0 transition, since the strong H_0 PL emission prevents a clear observation of the Raman signal. Under these conditions, the resonance only moves slightly (≈ 3 meV) to higher energies when the field is increased from 0 to 13.5 T, whereas its peak intensity changes dramatically. The L_0 outgoing-resonance intensity is displayed in Figs. 3(a) and

3(b) for $\sigma^- \sigma^+$ and $\sigma^+ \sigma^+$ configurations, respectively, as a function of the magnetic field. The arrows indicate the values of B for which double resonances in-out (H_4-L_0 , H_3-L_0 , L_3-L_0 , H_2-L_0 , and L_2-L_0) are expected, according to the energies observed in the PLE spectra, taking into account that the PLE transitions observed in the incoming (outgoing) polarization correspond to the incoming (outgoing) resonant channels. The strong peaks at 11.4 T in Fig. 3(a) and at 8.2 T in Fig. 3(b) are identified as DRRS in which the incident photon produces the excitation of L_2 , while the outgoing photon originates from the recombination of the ground-state electron and light-hole L_0 . The difference between the peak positions in Figs. 3(a) and 3(b) is due to the Zeeman splitting of the L_2 transition excited with σ^- and σ^+ polarizations. The whole process implies a change of $\Delta n = 2$ between the up and down transitions, each of them conserving n . In a similar way, the shoulder at 7.3 T in Fig. 3(a) [5.5 T in Fig. 3(b)] can be interpreted as a DRRS involving the light-hole transitions L_3 and L_0 , with change of three units in the Landau index. No indication of H_2-L_0 or H_3-L_0 DRRS is apparent. Besides the DRRS peaks, one can observe an increasing background in Fig. 3(b), which is well described by the B^2 dependence shown by the solid line and predicted in Ref. 12 for bulk GaAs.

A model in which n has to be conserved in each interband transition is shown in Fig. 4(a), where the processes involved in the observed DRRS are schematically depicted

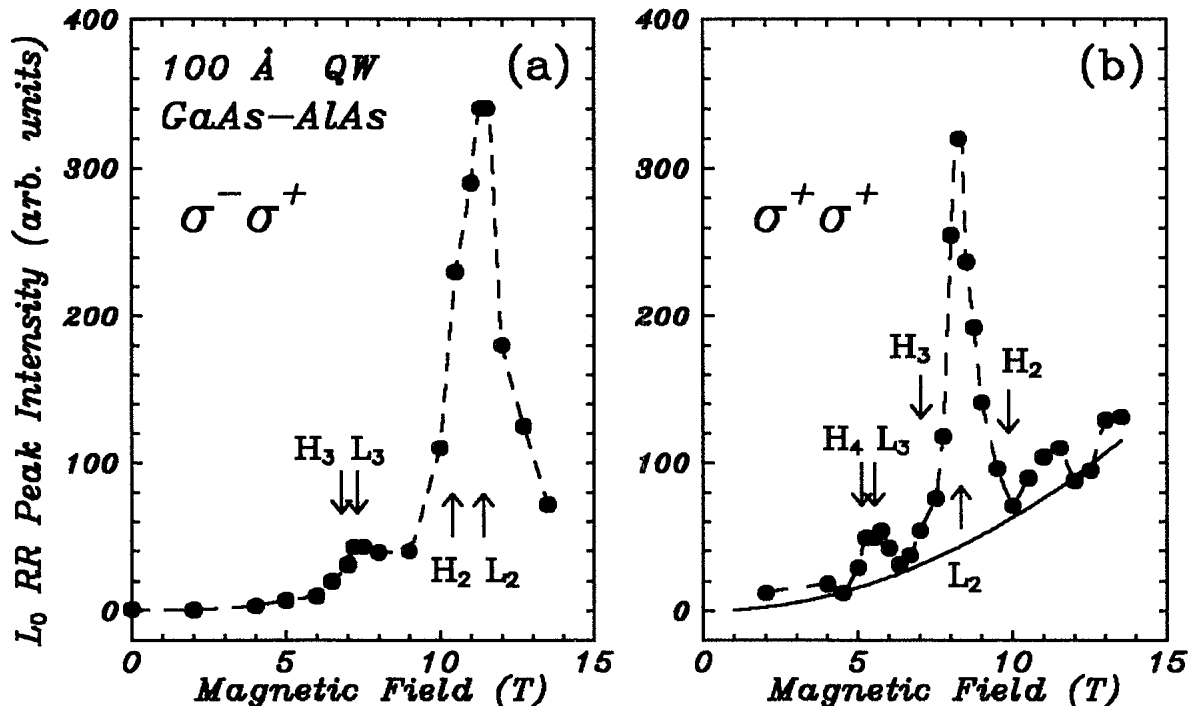


FIG. 3. Maximum intensity of the L_0 outgoing resonance vs magnetic field, for (a) $\sigma^- \sigma^+$ and (b) $\sigma^+ \sigma^+$ polarization configurations. The arrows correspond to expected double resonances with the incoming transition H_n or L_n , as identified from PLE experiments. The solid line in (b) shows a B^2 law (see text).

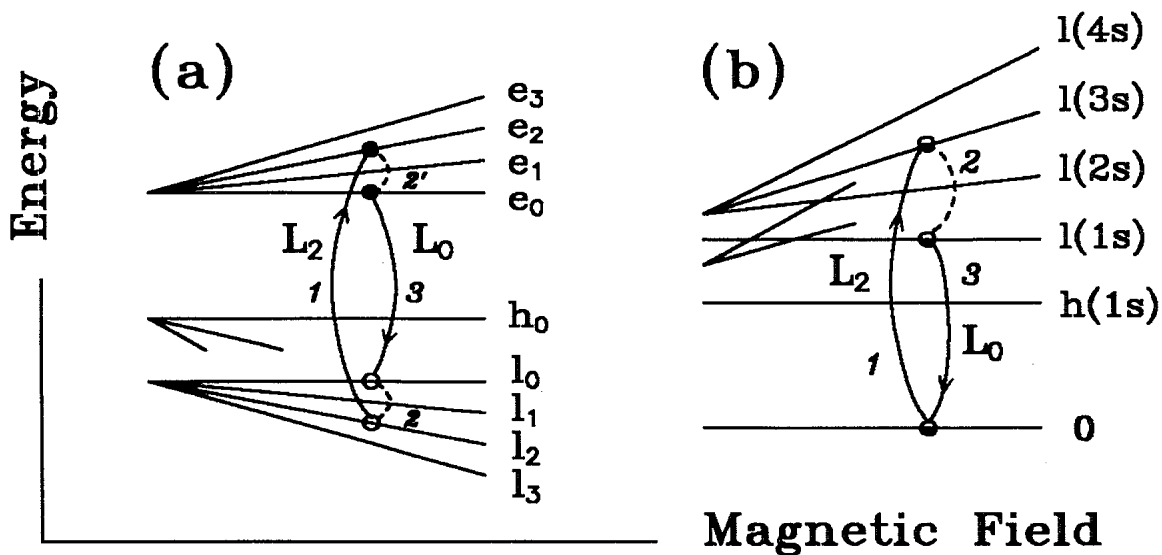


FIG. 4. Diagrams showing the possible processes involved in the observed DRRS: excitation L_2 , scattering by LO phonons (dashed lines), and recombination L_0 . (a) Four-band diagram for free particles. (b) Three-band scheme for exciton states. 0 denotes the vacuum state of excitons.

ed by arrows and numbered according to their occurrence. An electron-hole pair is created in bands e_2 and l_2 , respectively. In the scattering process both free particles relax independently to their respective ground states, e_0 and l_0 , before the recombination of the e_0 - l_0 electron-hole pair takes place.

Let us discuss first the change in the Landau index occurring in the DRRS event. This result differs essentially from that obtained in bulk GaAs, where the DRRS involves incoming and outgoing transitions corresponding to the same Landau index.¹¹ Some understanding of the reason why we observe Raman processes with $\Delta n = 2$ is obtained if one takes into account that bulk hole states are not pure light and heavy ones. In fact, they present a mixing given by [Eq. (42) of Ref. 12]

$$\begin{aligned}\psi_1^\pm &= a_1^\pm |n-2, \frac{3}{2}, +\frac{3}{2}\rangle + a_2^\pm |n, \frac{3}{2}, -\frac{1}{2}\rangle, \\ \psi_2^\pm &= b_1^\pm |n-2, \frac{3}{2}, +\frac{1}{2}\rangle + b_2^\pm |n, \frac{3}{2}, -\frac{3}{2}\rangle,\end{aligned}$$

as a result of the $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian separation in 2×2 blocks for the Γ_8^v valence bands. Since in the Raman process the optical-phonon-hole coupling implies a variation of the third component of the angular momentum $J_z = \pm 2$, this manifests in a $\Delta n = 2$ change from the functions $\psi_{1,2}^\pm(n)$ to the ones $\psi_{1,2}^\pm(n \pm 2)$ [see Fig. 3(a) of Ref. 12]. In other words, the simultaneous effects of phonon-hole coupling and magnetic field can be responsible for the $\Delta n = 2$ feature observed. Nevertheless, it is not clear how the above argument of plausibility would explain the observation of DRRS with a $\Delta n = 3$ change in the whole process.

On the other hand, the scheme of Fig. 4(a) in terms of

the free-particle-phonon interaction implies the simultaneous but separated relaxation of the electron and the hole to their respective ground states, e_0 and l_0 . Since the phonon energy is not likely to be shared in both independent processes, the observed DRRS suggest the exciton-phonon coupling as an alternative for the scattering mechanism. A scheme where only excitonic states are involved is illustrated in Fig. 4(b). The $l(3s)$, or L_2 , exciton created by the incoming light would be scattered as a whole to its ground state $l(1s)$, or L_0 , by the phonon. Thus, DRRS would be a three-band process in which all the states are real and excitonic in character. A heavy-light excitonic mixing, which is known to play an important role in the magneto-optical properties of GaAs QW's,⁶ would allow for the $\Delta n = 2$ change in the scattering event. The alternative explanation in the free particle scheme, namely, DRRS being a four-band process with at least one virtual intermediate state, is also possible, even if less likely to produce so much intense resonances as observed.

In conclusion, DRRS has been observed in a GaAs MQW at certain values of the laser energy and magnetic field. The results presented suggest the exciton-phonon coupling as the scattering mechanism, in accordance to the prominent role played by excitons in the optical properties of 2D systems, even if the single-particle mechanism cannot be ruled out. More theoretical work will be needed to fully characterize the microscopic nature of the observed resonances.

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