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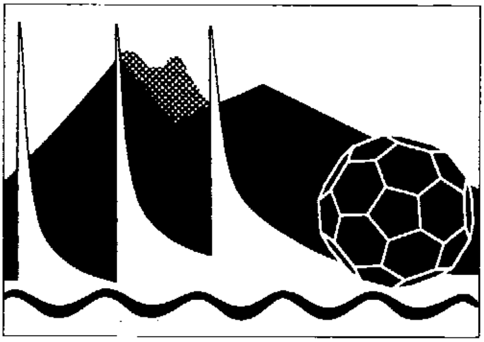

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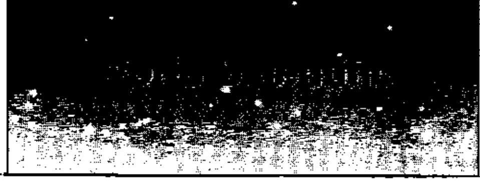

Double Raman Resonances in Semiconductor Quantum Wells: Excitonic Effects

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ABSTRACT

Magneto-Raman scattering by LO phonons has been measured on GaAs/AlAs multiple quantum wells under double resonance conditions We find an excellent agreement between the experiments and a calculation of the Fröhlich interaction considering the excitonic nature of the intermediate states in the Raman process and a non-zero in-plane phonon wave vector which accounts for the inter-Landau level transitions.

Resonant Raman scattering (RRS) occurs whenever the energy of the incident- (incoming resonance) or scattered-light (outgoing resonance) equals that of an electronic transition.[1] Up to date, the contributions of impurities[2] and excitonic effects[2,3] to the resonant behavior of the Raman cross section in semiconductors have not been satisfactorily clarified. A double-resonant Raman scattering (DRRS) process can occur when the energy difference between two electronic transitions in a system equals that of a LO phonon. In this case the enhancement of the Raman signal will be considerably larger than that of a single resonance, due to the simultaneous vanishing of the two energy differences appearing in the denominator of the description of the Raman cross section in third order perturbation theory.[1] DRRS conditions have been achieved in 2D systems by a precise choice of quantum well (QW) dimensions[4] and by the application of an external electric field.[5] Magnetic-field (**B**) induced DRRS have also been observed in bulk GaAs[6] and GaAs/AlAs QW's.[7]

In this work we present new data on DRRS in GaAs/AlAs QW's induced by magnetic fields up to 20 Tesla. The combination of our measurements and a calculation of the Fröhlich interaction taking into account the excitonic nature of the intermediate states in the Raman process clarifies the essential role of excitons in the resonant behavior of the Raman cross section in 2D semiconductors. GaAs/AlAs multiple quantum wells (MQW's) with different layer thicknesses were grown by MBE on (100) and (111)B GaAs substrates. Achromatic $\lambda/4$ plates were used to obtain circularly polarized light and

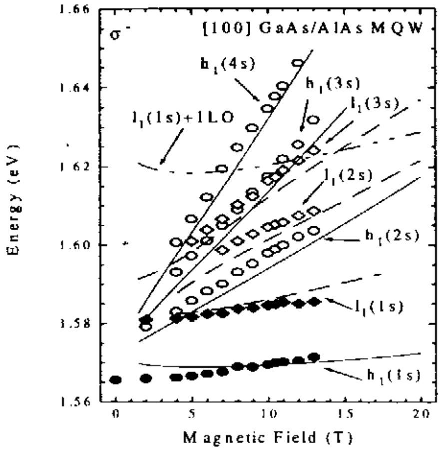

Figure 1. Energies of the ground (solid) and excited state (open) heavy-hole (circles) and light-hole (triangles) excitons as a function of the magnetic field for a GaAs/AlAs MQW. The lines represent the best fit of the magnetoexcitons (see text).

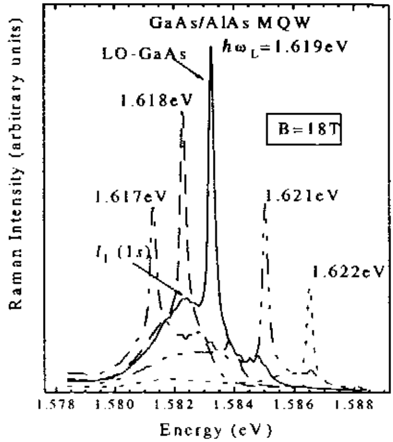

Figure 2.- Raman spectra of the sample in Fig.1 at 18T and $\sigma^+\sigma^+$ configuration for different laser energies. The sequence correspond to a double resonant with $l_1(1s)$ and $l_1(2s)$ as outgoing and incoming channels respectively

to analyze the scattered light. **B** was applied in the Faraday configuration.

Figure 1 shows the energies of the ground and excited exciton states as a function of **B**, obtained from photoluminescence excitation (PLE) with σ^- light, for a (100) sample. An accurate knowledge of these energies is crucial to find the conditions for DRRS. To fit the experiments we have calculated the VB energy dispersion taking into account the *hh-lh* mixing.[8] The electron in-plane effective mass (m^e_{xy}) has been taken as a free parameter.[9] Excitonic effects are included through a expression for the binding energy, which takes into account **B** and the confinement in the QW by a dimensionality parameter D.[10] The solid (dashed) lines in Fig. 1 represent the best fit of the ground and excited states of *hh* (*lh*) excitons, with D=0.6 and m^e_{xy}=0.075 m_0.

Raman spectra are shown in Fig. 2 at 18T for $\sigma^+\sigma^+$ polarization. The laser energies have been chosen to sweep the outgoing resonance condition with $l_1(1s)$, whose photoluminescence is seen as a broad background underneath the sharp GaAs-LO phonon peak. One observes clearly the outgoing resonant behavior of the LO phonon as it crosses $l_1(1s)$. **B** corresponds to a condition of double resonance, with $l_1(2s)$ as incoming channel, for a laser energy of 1.619 eV.

The maximum intensities of the LO-phonon, tuning the laser energy to follow an outgoing resonance with $l_1(1s)$, are plotted in Fig. 3 for $\sigma^+\sigma^+$ (solid circles) and $\sigma^-\sigma^-$ (open circles) as a function of **B**. These constitute the DRRS profiles, which clearly show peaks at the fields corresponding to a $l_1(ns, n>1)$-$l_1(1s)$ energy separation equal to the LO-phonon energy. The backgrounds and the intensity ratio between the different double-resonances follow a B^2 law as predicted theoretically.[6,8] One should emphasize that,

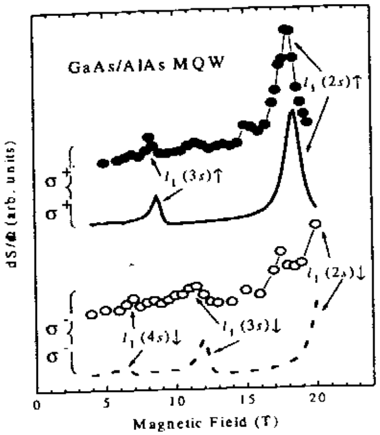

Figure 3. DRRS profiles (symbols) for $\sigma^+\sigma^+$ (full) and $\sigma^-\sigma^-$ (open) configurations. The lines represent the calculation of the Raman scattering efficiency including excitonic effects.

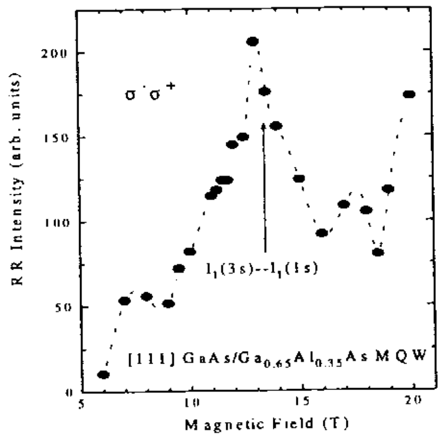

Figure 4. Double resonant Raman profile for a (111)-oriented GaAs/GaAlAs QW in $\sigma^-\sigma^+$ configuration. l_1(3s) and l_1(1s) act as incoming and outgoing channels, respectively.

when $l_1(1s)$ is selected as outgoing channel, only double resonances between <u>lh</u> states are observed and that any arbitrary change in the excitonic principal quantum number (n), which is related to the Landau quantum number, is allowed. Figure 4 shows the DRRS profile for a (111)-oriented QW in $\sigma^-\sigma^+$ configuration. The large enhancement corresponds to l_1(3s) and l_1(1s) acting as incoming and outgoing channels, respectively.

In order to calculate the Raman scattering efficiency per unit crystal length and solid angle one has to evaluate the following expression:

$$\frac{dS}{d\Omega}=\frac{\omega_L\omega_S^3\eta_L\eta_S}{(2\pi)^2c^4}\frac{V}{(\hbar\omega_L)^2}\left|W_{FI}(\omega_S,\vec{e}_S;\omega_L\vec{e}_L)\right|^2[n(\omega_q)+1] \quad (1)$$

where $\omega_{L(S)}$, $\eta_{L(S)}$ and $\vec{e}_{L(S)}$ are the angular frequency, refractive index and polarization wave vector of the incident (L) and scattered (S) photon, and W_{FI} is the probability amplitude of the process, which, for a one phonon emission process, in third order perturbation theory, can be written as:

$$W_{FI}=\sum_{\alpha,\beta}\frac{\langle 0|\hat{H}_{ER}|\beta\rangle\langle\beta|\hat{H}_{EP}|\alpha\rangle\langle\alpha|\hat{H}_{ER}|0\rangle}{\left(\hbar\omega_S-E_\beta+i\Gamma_\beta\right)\left(\hbar\omega_L-E_\alpha+i\Gamma_\alpha\right)} \quad (2)$$

where $|\alpha\rangle$ and $|\beta\rangle$ are the intermediate states with energy $E_{\alpha(\beta)}$ and lifetime broadening $\Gamma_{\alpha(\beta)}$. H_{ER} (H_{EP}) is the electronic-state/radiation (phonon) interaction. If electron-hole pairs are considered as intermediate states, even taken into account VB mixing, the theory predicts that the Landau quantum number is conserved and that the scattering process is intersubband, and obtains resonances whose intensities and **B** values are in discrepancy with the experimental findings. These results indicate that excitonic effects must be taken into account. The Hamiltonian of a 2D magneto-exciton may be written as:[11]

$$H_{exc} = -\frac{\hbar^2}{2}\frac{\partial}{\partial z_e}\frac{1}{m_z^e}\frac{\partial}{\partial z_e} + V(z_e) - \frac{\hbar^2}{2}\frac{\partial}{\partial z_h}\frac{1}{m_z^h}\frac{\partial}{\partial z_h} + V(z_h)$$
$$-\frac{\hbar^2}{2\mu_{xy}^h}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial}{\partial r}\right) - \frac{\hbar^2}{2\mu_{xy}^h}\frac{1}{r^2}\frac{\partial^2}{\partial\phi^2} + \frac{1}{2i}\hbar\frac{\mu_{xy}^h}{\gamma_{xy}^h}\omega_c^h\frac{\partial}{\partial\phi} + \frac{\mu_{xy}^h\left(\omega_c^h\right)^2}{8}r^2 + V_c(r, z_e - z_h) \quad (3)$$

The last term, V_c, is the electron-hole Coulomb interaction ($e^2/4\pi\varepsilon_0 r$), which we treat as a perturbation, up to second order, to the motion of the electron-hole pair in **B**. We found that, in strict back-scattering configuration, the interaction exciton-photon is not able to connect excitonic states with different quantum number n. Thus we must invoke a higher order process, which we assume to be roughness scattering. In this model, the double resonances develop in triple resonances. The results of this calculation are plotted in Fig. 3 as a solid (dotted) line for the $\sigma^+\sigma^+$ ($\sigma^-\sigma^-$) configuration. The complete theory predicts correctly the position of the resonances as a function of **B** and the relative intensities in both polarization configurations.

The experiments performed in (111) QW´s proof that the predominant scattering mechanism is the Fröhlich interaction, since under resonant conditions the enhancement of the Raman signal is absent for the TO modes and is only seen for the LO modes.

In summary, we have studied magnetic-field induced DRRS in (100)- and (111)-oriented GaAs MQW's for different polarization configurations. The conditions for DRRS agree very well with those predicted by PLE measurements. It appears that the Raman scattering is dominated by the Fröhlich mechanism and that excitonic effects are crucial to explain the experiments. A calculation of the Raman efficiency, taking into account the Coulomb interaction between electrons and holes and roughness scattering, reproduces exceptionally well the main features of the observed double Raman resonances.

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