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RESONANCE RAMAN SCATTERING IN CdTe/CdMnTe SUPERLATTICES UNDER A MAGNETIC FIELD

L. Viña[1,2], F. Calle[2*], J.M. Calleja[2], L.L. Chang[3], and J. Yoshino[3**]

[1]Instituto de Ciencia de Materiales de Madrid, Consejo Superior de Investigaciones Científicas. Madrid. SPAIN
[2]Departamento de Física Aplicada, C-IV. Universidad Autónoma. Cantoblanco. 28049 Madrid. SPAIN
[3]IBM T.J. Watson Research Center. P.O. Box 218, Yorktown Heights, NY 10598, U.S.A.

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We present a comparative study of the pseudo-absorption and resonance Raman scattering in CdTe/$Cd_{1-x}Mn_xTe$ superlattices in the presence of an external magnetic field. Under excitation with circularly polarized light, the resonance Raman profiles separate into two components corresponding to the Zeeman-split ground state excitons. A further analysis of the polarization of the scattered light proves that the efficiency of the Fröhlich mechanism is larger by a factor of ~ 10 than the corresponding deformation-potential one.

1. Introduction

$Cd_{1-x}Mn_xTe$ belongs to a group of materials known as semimagnetic or dilute magnetic semiconductors. The interest in these materials comes from the unique magnetic and magnetooptic properties they exhibit due to the presence of the ion Mn^{2+}, which has a large magnetic moment of 5.92 Bohr magnetons.[1] The exchange interaction between the localized $3d^5$ electrons of the Mn^{2+} ions and itinerant band electrons results in large g-factors, strongly circularly polarized luminescence, and other interesting phenomena which link the field of semiconductor physics and magnetism. The variation of the band gap in $Cd_{1-x}Mn_xTe$ with increasing Mn concentration can be used to build superlattices (SL's) and quantum wells, by the successive deposition of CdTe and $Cd_{1-x}Mn_xTe$ layers, thus giving the possibility of a precisely controlled variation of band schemes and dimensionality.[2] This system represents the first heterostructure realized with semimagnetic materials,[3,4] and it has been the most extensively studied.[5] The application of an external magnetic field makes possible to tune the electronic states, and therefore to study the effects of dimensionality on magnetic behavior.[6] Photoluminescence (PL) and PL excitation (PLE) have been used to investigate the effects of the magnetic field on the optical properties.[7,8]

Resonance Raman Scattering (RRS) by LO phonons is an optical method which provides information on both lattice-dynamical and the electronic properties of crystals. This technique has been widely used to investigate SL's and heterostructures in the last years.[9] RRS in the presence of a magnetic field has recently been used to enhance sensitivity for the observation of interband transitions.[10] The reduction of the dimensionality of the density of states by the field leads to a sharpening of the resonant peaks. It has also been observed that the field increases the strength of the Raman resonance of the LO confined modes and especially of the interface modes in GaAs-$Ga_{1-x}Al_xAs$ SL's.[11] In CdTe-$Cd_{1-x}Mn_xTe$ SL's RRS studies have been performed only at zero magnetic field.[12-15] The Raman efficiency for LO phonon scattering in bulk $Cd_{1-x}Mn_xTe$ near the fundamental gap, in the presence of magnetic fields, has been investigated recently.[16]

In this work we investigate the electronic properties of [111]-oriented CdTe/$Cd_{0.87}Mn_{0.13}Te$ SL's in the presence of an external magnetic field by means of RRS. The results are compared with the pseudo-absorption obtained through PLE measurements. The polarized Raman profiles show clearly that the Fröhlich mechanism predominates over the deformation-potential one under resonant conditions.

2. Experiments

The samples were grown by molecular beam epitaxy on (001)-GaAs substrates. A buffer layer of CdTe, (111)-oriented, was followed by a CdTe/$Cd_{0.87}Mn_{0.13}Te$ superlattice, consisting of 25 periods of 100Å-wide wells and barriers. The period and individual thicknesses were obtained from X-ray diffraction measurements and from small-angle X-ray interference.[2] The Raman spectra were obtained with a LD700 dye-laser pumped by a Kr^+-ion laser. The scattered light was analyzed with a Jarrel-Ash 1-m double-grating spectrometer and detected with standard photon-counting techniques. The power density was kept below $5 Wcm^{-2}$ and $0.1 Wcm^{-2}$ for the Raman and PLE measurements, respectively. The incident light was circularly polarized by means of a $\lambda/4$ plate. The scattered (or emitted) light was analyzed by means of a second $\lambda/4$

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* Present address: E.T.S.I.T. Universidad Politécnica, Ciudad Universitaria, 28040 Madrid. SPAIN.
** Present address: Tokyo Institute of Technology. 4259 Nagatsuda, Midori-ku, Yokohama 227. JAPAN

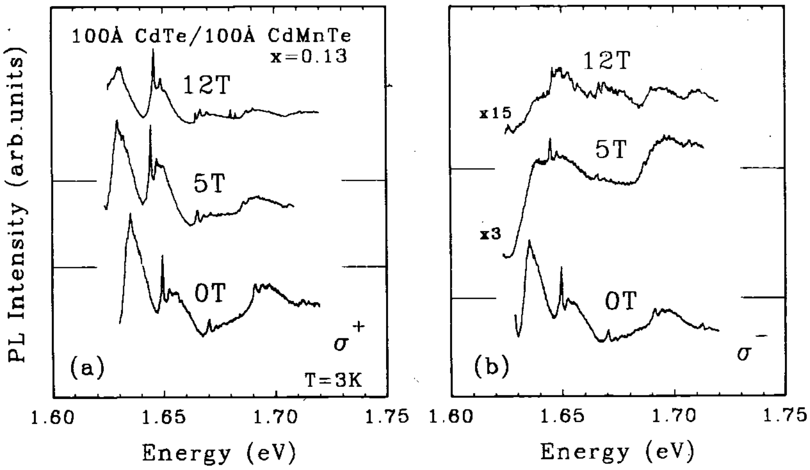

Fig. 1(a): Photoluminescence excitation spectra for a 100Å wide $CdTe/Cd_{0.87}Mn_{0.13}Te$ superlattice at different magnetic fields. The spectra were obtained at 3K, exciting with σ^+ circularly polarized light. (b): Same as (a) for excitation with σ^- polarized light.

plate and a linear polarizer. The measurements were performed in a back-scattering configuration, at 3K, with the sample immersed in a liquid-He bath-cryostat within a standard-coil superconducting magnet. The fields, up to 13T, were applied in the Faraday configuration

3. Results and discussion

We show in Fig. 1 PL excitation spectra at different magnetic fields obtained with σ^+ (a) and σ^- (b) polarized exciting light. Two different kind of structures are clearly observed in the spectra: the broad features correspond to excitonic transitions between quantum confined levels in the SL's; the sharp peaks correspond to first and higher-order Raman scattering by LO and interface (IF) phonons. The presence of this periodic structure in the PLE spectra of bulk CdTe has been reported previously.[17] Recently, it has been shown that light scattering by phonons dominates the PLE spectra in $CdTe/Cd_{1-x}Mn_xTe$ SL's when the detection is made on the high energy tail of the photoluminescence.[13] Let us concentrate now on the excitonic transitions. A clear shift towards the red of the two lowest excitonic transitions is observed between 0T and 5T when exciting with σ^+ light. At higher fields the excitons shift back towards higher energies and new features are resolved in the spectra. On the other hand, a blue shift is obtained for the lowest-lying exciton under σ^- excitation, while the energy of the second structure decreases with increasing magnetic field. Note that, in both polarizations, the strength of the second exciton, relative to that of the first one, increases monotonically with field. This fact, together with the polarization selection-rules,[15] indicates that the second exciton is probably associated with excited states. An analysis of the polarization of the emission, exciting with circularly polarized light, also reveals that the lowest exciton is composed of a strong mixing of heavy-hole and light-hole states:[15] the peak observed with σ^+ excitation originates mainly from $|3/2,-3/2\rangle$ and $|3/2,-1/2\rangle$ states ($|J,J_z\rangle$ denotes the total angular momentum and its third component), while with σ^- an exciton with a strong coupling of $|3/2,3/2\rangle$ and $|3/2,1/2\rangle$ hole states is observed.

Figure 2 compiles the shift with magnetic field of the lowest transitions observed with σ^+ (open circles) and σ^- (full circles) excitation. An almost symmetric splitting of the lowest exciton is seen, amounting to ~10meV at 7T. The usual Landau level and Zeeman splittings in non-magnetic superlattices give rise to small effects in the $CdTe/Cd_{1-x}Mn_xTe$ system, the main effect in this case arises from the exchange interaction in the barriers and the exciton-ion interaction when the carrier penetration into the barriers is substantial. From the shift of the lowest exciton

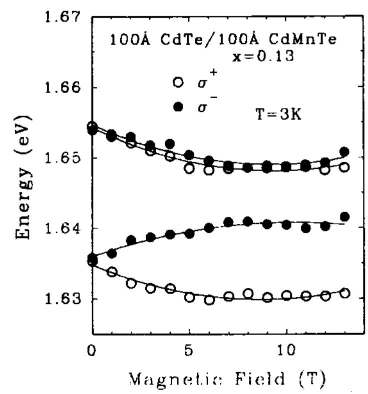

Fig. 2. Excitonic energies versus magnetic field for σ^+ (open circles) and σ^- (full circles) polarized incident light.

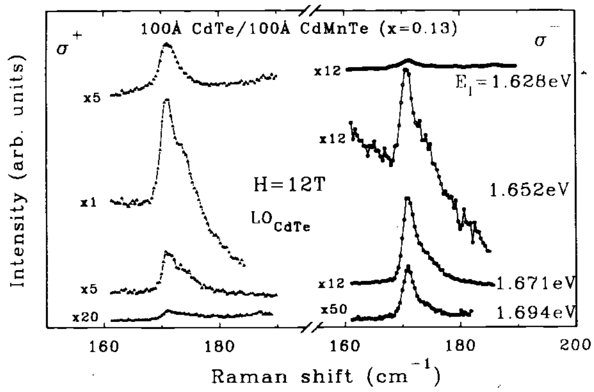

Fig. 3. Raman spectra in the energy range of the LO-CdTe phonon at 12T for different laser energies. Left (right) spectra were obtained with σ^+ (σ^-) polarization, the scattered light was not analyzed. The intensities have been multiplied by the factors on the left of the spectra.

with magnetic field, we have obtained an effective g-factor of ~25. The saturation of the magnetization in the CdMnTe layers at ~7T produces that the splitting remains constant beyond this field. The behavior of the second lowest state, with a shift towards the red for both polarizations, is at present not understood.

Several phonons corresponding to lattice vibrations in the CdTe (LO-CdTe) and $Cd_{1-x}Mn_xTe$ layers (LO_1-CdTe-like and LO_2-MnTe like), as well as IF vibrations, have been observed in the Raman spectra of CdTe-$Cd_{1-x}Mn_xTe$ SL's.[12,15]. Close to resonance with the ground state exciton of the SL's the LO-CdTe and the IF modes dominate the spectra. Since the resonant behavior of both phonons is quite similar,[15] we will concentrate here in the resonance of the LO-CdTe mode in the presence of the magnetic field. Figure 3 shows Raman spectra for LO-CdTe at 12T for σ^+ (on the left) and σ^- (on the right) incident polarized light and depolarized scattered light at different laser energies. The spectra are multiplied by the factors depicted on their left. A strong enhancement of the signal is observed when the energy of the scattered light (E_l - E_{LO}) approaches the lowest excitons observed in the PLE spectra at 1.631eV and 1.641eV for σ^+ and σ^- polarizations, respectively. At the same laser energy, the intensities of the peaks close to resonance are considerably larger for σ^+ excitation. The resonance corresponds to the outgoing channel and therefore the Raman spectra are superimposed on a large background due to the PL.

The complete resonant behavior at 12T is shown in Fig. 4 for both polarizations. A comparison of the Raman profiles with the corresponding PLE spectra allow us to identify the low energy and the high energy peak in the resonance curves, separated by one LO-phonon energy, with the incoming and outgoing channels of the resonance, respectively. The larger strength of the outgoing channel may be attributed to impurity induced one-LO-phonon scattering, which presents a double resonant behavior in the denominator of the scattering amplitude.[16,18] The asymmetry in the line shape of the outgoing resonance also points to impurity effects, however the influence of bound excitons as intermediate electronic states cannot be ruled out.[14,19] Actually, from the anisotropy of the excitonic shifts in the presence of a magnetic field applied in the Faraday or the Voigt configurations, it has been concluded that the excitons in CdTe/$Cd_{1-x}Mn_xTe$ are mainly bound to defects at the interfaces.[20] The resonance is a factor of ~10 larger for σ^+ polarization than for σ^- excitation, approximately the same factor which is obtained in the PLE experiments (see uppermost curves in Figs. 5 (a) and (b)). The difference in the energy of the resonance maxima, 10 meV, agrees with the result obtained in the pseudo-absorption measurements (see Fig. 2). A study of the dependence of the resonance on the field-strength shows that a sharpening and a further enhancement of the resonance profiles is obtained with increasing magnetic field.

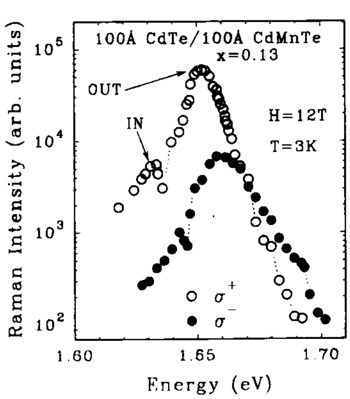

Fig. 4. Resonance Raman profiles at 12T for σ^+ (open circles) and σ^- (full circles) polarized incident light. The incoming and outgoing channels of one resonance with the ground state exciton are indicated by arrows.

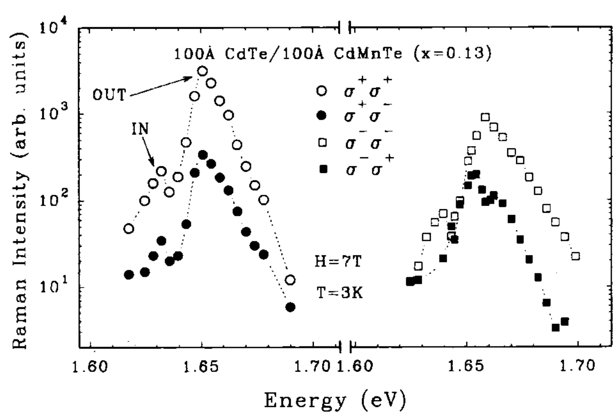

Fig. 5. Resonance Raman profiles for the four possible scattering configurations at 7T. The incoming and outgoing channels of one resonance with the ground state exciton are indicated by arrows.

This behavior is consistent with the reduction of the dimensionality of the density of states by the field.[21] For σ^+ (σ^-) incident light the peaks of the resonance shift to lower (higher) energy with increasing magnetic field by an amount which agrees with the results depicted in Fig. 2. A much weaker enhancement is also obtained in the experiments for the second exciton observed in PLE experiments.

Further information about the scattering mechanisms can be obtained when the scattered light is also analyzed into its circularly polarized components. Figure 5 depicts resonance Raman profiles in back-scattering configuration for the four possible combinations of incident and scattered light at 7T. For a [111] surface and LO-phonon scattering in zinc-blende materials, the Raman tensors for Fröhlich and deformation potential mechanisms can be written as:[22]

$$R_F = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & a \end{pmatrix} \qquad R_{DP} = \sqrt{\frac{1}{3}} \begin{pmatrix} 0 & d & d \\ d & 0 & d \\ d & d & 0 \end{pmatrix}$$

Therefore, for circularly polarized incident and scattered light the deformation-potential mechanism is relevant for orthogonal polarizations ($\sigma^+\sigma^-$ and $\sigma^-\sigma^+$), while in the polarized scattering configurations ($\sigma^+\sigma^+$ and $\sigma^-\sigma^-$) the Fröhlich mechanism is the appropriate one. As can be seen from Fig. 5, for σ^+ incident light both resonances exhibit a maximum at the same energy, and the one corresponding to the Fröhlich mechanism ($\sigma^+\sigma^+$) is an order of magnitude larger than the one related to the deformation-potential. The behavior is quite different for σ^- incident light, as can be seen on the right hand side of Fig. 5. A double structure in the outgoing channel is observed for $\sigma^-\sigma^+$, while only a broad peak is seen for $\sigma^-\sigma^-$. The appearance of two local maximum in the resonance Raman profiles for σ^- incident light has also been reported for $Cd_{1-x}Mn_xTe$ in the presence of magnetic field.[16] A comparison of the resonance Raman experiments and the excitation spectra allow us to identify the origin of this doublet. The lower energy peak corresponds to the outgoing resonance with the ground state exciton $|3/2,->$ whereas the higher energy one is associated with the ground state $|3/2,+>$ exciton. The incoming channel is only resonantly enhanced for the latter exciton. The fact that, for deformation-potential scattering exciting with σ^- light, the Raman efficiency resonates with the $|3/2,->$ exciton points to an additional mixing between the holes with spin-up and spin-down which build the excitonic wave function. This mixing is also observed in the PLE spectra when the emitted light is analyzed into its circularly polarized components.[8] Let us also remark that the doublet in the resonance Raman profiles cannot be resolved if the scattered light is not analyzed, due to the larger strength in the $\sigma^-\sigma^-$ configuration close to resonance. A similar doublet should be also expected for $\sigma^+\sigma^-$, however, the lower oscillator strength of the ground state exciton corresponding to σ^- excitation (as can be inferred from a comparison of the $\sigma^+\sigma^+$ and $\sigma^-\sigma^-$ resonance profiles) hinders its resolution.

4. Conclusions

We have investigated resonance Raman scattering in CdTe/$Cd_{1-x}Mn_xTe$ superlattices in the presence of an external magnetic field. With increasing magnetic field the resonance profiles sharpen and shift to lower and higher energies for σ^+ and σ^- incident light, respectively, in agreement with the splittings observed in photoluminescence excitation spectra. The outgoing channel of the resonance dominates over the incoming one and their intensity ratio is independent of magnetic field. An analysis of the polarization of the scattered light proves that the Fröhlich mechanism is more important than the deformation-potential one in resonance. This analysis also reveals that the excitonic wave functions arise from a complicated mixing of valence band states.

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