Pergamon

Solid State Communications, Vol. 91, No. 7, pp. 545-549, 1994
Elsevier Science Ltd
Printed in Great Britain
0038-1098/94 $7.00+.00

0038-1098(94)00406-4

RESONANCE RAMAN SCATTERING BY INTERFACE PHONONS IN THE MAGNETIC FIELD IN GaAs/AlAs SUPERLATTICES.

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(Received 14 March 1994 by M.Cardona)

The dependence of the intensity of resonant Raman scattering by interface (IF) phonons in GaAs/AlAs superlattices on magnetic field, temperature, and pumping power have been investigated. IF phonon spectra, forbidden according to the momentum conservation condition, were observed in a magnetic field. Oscillations of the IF phonon intensity vs. magnetic field, corresponding to the resonance of the exciting light energy with the levels of magneto-excitons which are the intermediate states in the scattering process, have been observed. The IF phonon intensity decreases with increasing temperature. We consider such behaviour to be connected with the temperature-induced delocalization of magneto-excitons bound near static defects which are responsible for the relaxation of the momentum conservation condition. The delocalization energy is observed to increase with magnetic field.

Introduction.

It is well known that surface optical vibrations arise at the boundary between two media provided at least one of them is a polar ("active") medium. The frequencies of these "surface phonons" are located inside the TO-LO splitting range where the dielectric constant of the active medium is negative. The amplitude of the surface vibration decreases with increasing distance from the interface. Such surface (interface) phonons have been observed and investigated with the methods of infrared [1] and Raman [2] spectroscopy.

The intensity of Raman scattering (RS) by interface phonons is quite small when observed in the back-scattering configuration commonly used for opaque media. The cross-section in the back-scattering configuration is small due to the fact that the normal component of the wave vector of the surface wave q_z is imaginary. As a result, the momentum conservation condition is not rigorously obeyed, and the scattering cross-section is reduced in comparison with the case of "forward-scattering" [3,4] used with the transparent media.

However, even in the back-scattering configuration Raman scattering by surface modes can be detected. Recently the dispersion relations $\omega(q_\parallel)$ for the heterostructure GaAs/AlAs have been obtained using this technique with the tilted light beam direction. The $\omega(q_\parallel)$ dependence has been measured in the range of wave vectors $q_\parallel$ where retardation effects are essential, i.e. $q_\parallel \sim \omega_{TO}/c$, $q_\parallel$ being the component of the wave vector in the interface plane [5].

In multilayer structures with small layer thicknesses d, where $q_\parallel d < 1$, (for example, in semiconductor superlattices (SL) and quantum well (QW) structures) the surface waves localized on the different interfaces overlap. This leads to the appearance of so-called interface modes ($q_\parallel \neq 0$), propagating also along the z-axis (i.e. to the appearance of a real $q_z \neq 0$).

A theory of these collective excitations for the SL has been developed [6,7]. The condition $\varepsilon_1(\omega) = -\varepsilon_2(\omega)$, corresponding to the existence of surface waves on the interfaces between two semi-infinite media, can be obtained in the particular case $q_\parallel d_1 \gg 1$ and $q_\parallel d_2 \gg 1$, where $d_{1,2}$ are the thicknesses of the SL layers (formula (6) from Ref. [6]). In the general case the interface phonon frequencies can be presented in the form of two branches $\omega^{\pm}(q_\parallel, d_2)$ for every $\omega_{LO} - \omega_{TO}$ splitting range of the two

media. Here the signs (ω^+ and ω^-) for the branches of interface (IF) phonons according with [8] are used. The specific shape of the function $\omega(q_\parallel)$ depends on $q_z(d_1+d_2)$ as a parameter [7,8].

Raman spectra of IF phonons in GaAs/AlAs SL's were observed under resonant conditions by Sood et.al. [8]. These spectra were further investigated in a number of subsequent works [9-13]. However, it is worth mentioning that although the peaks found in these studies belong to the $\omega_{LO} - \omega_{TO}$ splitting ranges of GaAs and AlAs, their frequencies do not fulfill both the momentum conservation condition for $q_\parallel$ and the dispersion $\omega(q_\parallel)$ simultaneously.

For excitation far from exciton resonances an enhancement of the Raman intensity of the IF phonons was observed by Gammon et.al. [9,10] upon application of a strong magnetic field B up to 7 T directed along the z axis. At the same time an enhancement of the confined LO phonons took place. An increase of the pumping power density P resulted in a decrease of the intensity of the IF phonons. At $P \approx 4 \cdot 10^3$ W/cm^2 the IF phonon scattering induced by the magnetic field practically disappeared. A similar but considerably smaller decrease in Raman intensity was observed in Ref.[11] for the scattering by the confined LO phonons.

The nature of the structural static defects responsible for relaxation of the momentum conservation condition in the process of RS by IF phonons has been discussed in [9-13]. There are two general explanations of this phenomenon. The first is connected with the localization of the magneto-excitons (which are intermediate states in the scattering process) at the islands of the QW width fluctuations. The second one is the influence of the charged impurities on the scattering process. The nature of the defects remains open. On one hand, the increase of the pumping power density could lead to the neutralization of charged impurities by photoexcited carriers. As a result, the influence of the charged impurities on the RS intensity would decrease. However, the role of the magnetic field in this case would not be clear. On the other hand, the influence of the islands of the QW width fluctuations on the RS intensity, which could increase in a magnetic field, is hardly compensated by the photoexcited carriers [9-11].

In order to clarify the situation we have investigated the dependence of the Raman intensity by interface phonons on magnetic field, temperature and pumping power.

Experimental details and results.

The superlattice sample studied was grown on a (001) GaAs substrate by molecular beam epitaxy. It consisted of 50 periods of 65Å (d_1) GaAs / 22Å (d_2) AlAs which were not intentionally doped. The characterization of the SL was made by x-ray diffraction.

The energies of confined excitons associated with n=1 and n=2 heavy-hole (HH) and light-hole (LH) states as measured with photoluminescence and photoluminescence excitation spectroscopy were found to be: E_{HH1}= 1.614 eV, E_{LH1}= 1.625 eV, E_{HH2}= 1.92 eV, E_{LH2} = 2.01 eV.

Raman spectra were observed using Kr^+ laser energies $\hbar\omega_{exc}$ = 1.833 eV (far from exciton resonances) and 1.916 eV (in the vicinity of the HH2 exciton incoming resonance). These two cases of excitation will be described separately. Raman spectra were recorded in the $z(x,x)\bar{z}$, $z(x,y)\bar{z}$ and $z(x+iy,x\pm iy)\bar{z}$ back-scattering configurations; z is normal to the layers and x,y are along [100] and [010] directions. The sample was held inside a superconducting magnet in a He-cryostat. A magnetic field up to 7.5 T was directed perpendicularly to the SL plane. In the measurements of the Raman intensity vs. temperature the pumping power density, P, was kept well below 10^2 W/cm^2 to decrease additional heating of the sample.

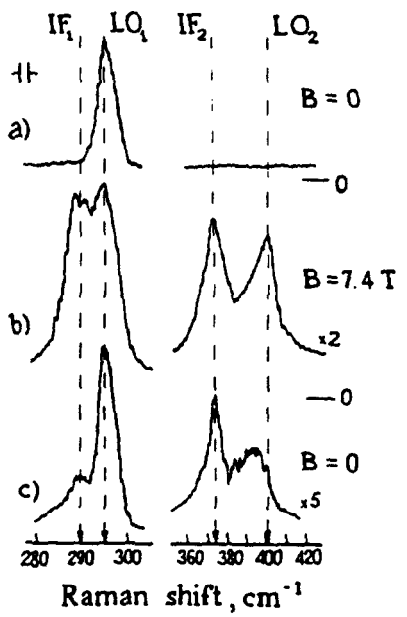

Fig.1. Raman spectra of a GaAs/AlAs (65Å/22Å) SL. The configuration is $z(x,x)\bar{z}$, T = 2 K, power density P = 10^2 W/cm^2. The labels $IF_{1(2)}$ and $LO_{1(2)}$ denote, respectively, interface and confined LO modes in GaAs (AlAs).

Excitation energy $\hbar\omega_{exc}$ = 1.833 eV.

a) B = 0.

b) B = 7.4 T

Excitation energy $\hbar\omega_{exc}$ = 1.916 eV.

c) B = 0.

Raman spectra at $\hbar\omega_{exc}$ = 1.833 eV are shown in Fig.1(a,b). At B=0 (Fig.1a) only the peak corresponding to the LO phonon of GaAs (labeled LO_1) was observed. The intensities of this peak for two scattering configurations $z(x,x)\bar{z}$ and $z(x,y)\bar{z}$ are practically equal. The confinement of LO_1 phonon was not observed because of the large value of d_1.

In a magnetic field (Fig.1b) new structures appeared in the Raman spectra with the Stokes shifts ~402 cm^{-1} (near and below the LO frequency of AlAs) and peaks ~ 290 cm^{-1} and ~ 374 cm^{-1} within the TO-LO splitting range of GaAs and AlAs, respectively. We conjecture that the structures at ~290 cm^{-1} (labeled IF_1) and ~374 cm^{-1} (labeled IF_2) may be attributed to the interface phonons: ω^+ GaAs-like and ω^+ AlAs-like branches, respectively. The maximum of the peak at ~402 cm^{-1} coincides with the LO AlAs confined peak measured for $\hbar\omega_{exc}$= 2.41eV excitation. (The Raman spectrum shown in Fig.1b is similar to that of sample C in Ref.[8]). The intensities of both interface ($IF_{1,2}$) and optical ($LO_{1,2}$) phonons first increase and then oscillate with increasing magnetic field (Fig.2a).

The intensities of the IF_2 phonon as a function of the inverse temperature, T^{-1} , are shown in Fig.3 for several magnetic fields. At T > 25 K the plots of $ln(IF_2)$ vs. T^{-1} yield straight lines with slopes increasing with B. The strong enhancement of the LO_1 phonon intensity disappears gradually as the temperature rises concurrently with a decrease of the $IF_{1,2}$ phonon intensities. At T > 45 K the LO_1 intensity measured at B=7.4 T tends to the value measured in the absence of a magnetic field at the same temperature.

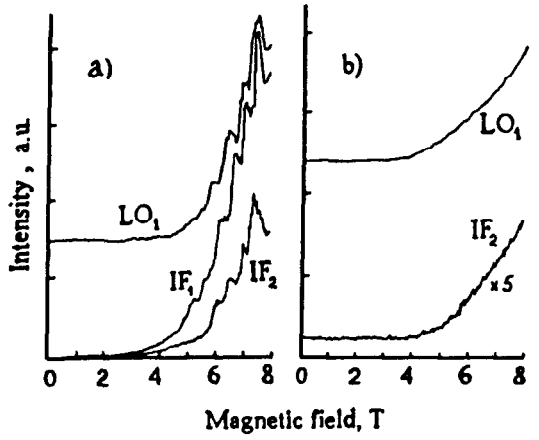

Fig.2. Intensity of interface IF_1 and IF_2 modes and confined LO_1 modes as a function of the magnetic field. T= 2 K, P=10^2 W/cm^2 . The configuration is $z(x+iy,x+iy)\bar{z}$.

a) $\hbar\omega_{exc}$= 1.833 eV, b) $\hbar\omega_{exc}$ = 1.916 eV.

The dependence of the IF_1 phonon intensity upon pumping power was investigated in the range P < 10^3 W/cm^2 at B= 7.4 T. For this purpose two sample holding techniques were used. A sublinear dependence of the IF_1 and LO_1 phonon intensities vs. P was seen when the sample was held in a liquid helium bath at T=2K (Fig.4a). In this case the ratio IF_1/LO_1 did not depend significantly on pumping power.

In the second case the sample was attached to a thermally insulated holder in liquid helium vapor (Fig.4b). The raising portion of the curve IF_1 vs. P at P < 10^2 W/cm^2 gives way to a decreasing portion leading to the vanishing of the IF_1 phonons at P < 10^3 W/cm^2. The IF_1/LO_1 relation drastically decreases with P [14].

Taking into account both the noticeable difference between cases a) and b) (Fig.4) for the dependence of IF_1 and LO_1 on P and the temperature dependence shown in Fig.3 we conclude that the decrease in the IF phonon intensity with increasing exciting power (case b)) may be explained by local heating of the sample with the exciting light. The local temperature of the sample in this case at P= 10^3 W/cm^2 was estimated to be about 40K.

So far we have presented the results obtained under excitation away from excitonic resonances. Under resonant excitation with the HH2 exciton transition the IF phonon peaks in the Raman spectra were observed even in the absence of the magnetic field (Fig.1c). The $IF_{1,2}$ phonons were polarized in the $z(x,x)\bar{z}$ configuration. The intensity of LO_1 phonons in the $z(x,x)\bar{z}$ configuration was more than that in the $z(x,y)\bar{z}$ configuration by a factor of 3. An increase of IF and LO phonon intensities with increasing magnetic field was also observed (see Fig.2.b).

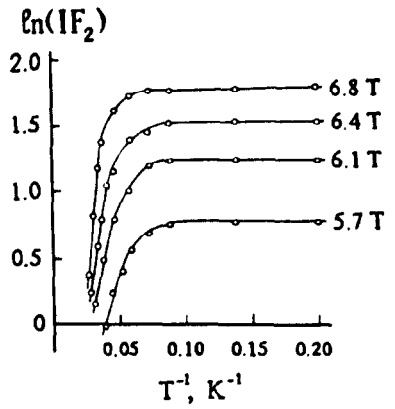

Fig.3. Intensity of interface IF_2 modes as a function of inverse temperature between 2 K and 60 K at B = 5.7 T, 6.1 T, 6.4 T and 6.8 T. $\hbar\omega_{exc}$ = 1.833 eV, P = 10^2 W/cm^2.

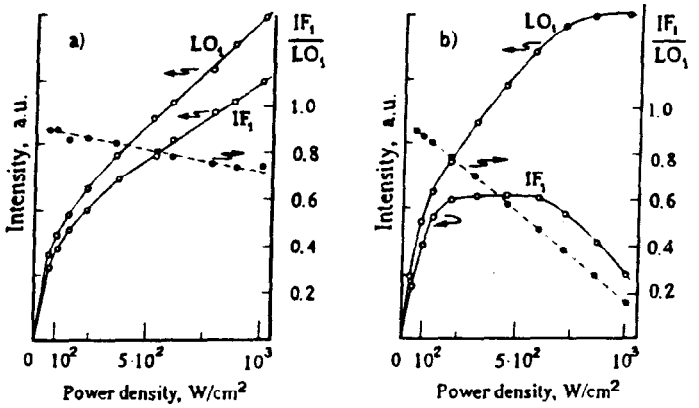

Fig.4. Intensity of interface IF_1 and confined LO_1 modes as a function of power density P at B = 7.4 T (solid curves). $\hbar\omega_{exc}$ = 1.833 eV. Dashed curves - ratio between the IF_1 and LO_1 mode intensities.
a) The sample is held in a liquid helium bath at T = 2 K.
b) The sample is on the thermally insulated holder in liquid helium vapor.

Discussion.

We discuss first the case of excitation in the first confined subband, i.e. $\hbar\omega_{exc} < E_{HH2}$.

The oscillations of the interface and optical phonon intensities observed for magnetic fields higher than 5T (Fig.2a) correspond to resonances of the incident photons with the levels of magneto-excitons (or correlated electron-hole pairs) which are intermediate states in the scattering process. The appearance of the "forbidden" scattering by IF phonons may be explained by localization of those intermediate states . Our experimental results suggest the model of magneto-exciton localization at islands of the QW width fluctuations [5]. In this model the enhancement of the IF and LO phonon intensities due to the magnetic field corresponds to an increase in the density of localized states. Provided the magneto-exciton is restricted in the plane of the quantum well, it can become trapped by islands of comparable sizes.

An increase in temperature leads to the decrease of the exciton trapping time which reveals itself in the disappearance of both the "forbidden" interface phonons and additional LO phonon intensity induced by the magnetic field. This additional intensity is connected with the contribution of the localized states to the RS process. Indeed, at T > 45 K, when the $IF_{1,2}$ phonon intensity vanishes, the LO_1 phonon intensity does not depend on the magnetic field. It should be mentioned that such behavior of the LO phonon intensity vs temperature in the case of resonance excitation was described earlier by Zucker et.al. [15,16]. These authors discussed the low temperature effects on exciton localization revealed by resonant Raman scattering.

The dependence of IF_2 phonon intensities vs. T obtained in this work make it possible to measure the delocalization energy of the magneto-excitons, E_b . The values of E_b are observed to increase from 2 meV to 5 meV with magnetic field increasing from 5.7 T to 7.4 T.

Let us compare the above mentioned dependence of IF and LO phonon intensities on magnetic field and temperature with those for the case of resonant excitation, i.e., $\hbar\omega_{exc} \approx E_{HH2}$ ($\hbar\omega_{exc}$= 1.916 eV). The observation of IF phonons at B=0 (Fig.1c) points out the fact that the intermediate state in this case is the localized HH2 exciton. As a magnetic field is applied, the $IF_{1,2}$ and $LO_{1,2}$ phonon intensities undergo changes, though rather weak in comparison with the effects found for $\hbar\omega_{exc}$=1.833eV. The absence of intensity oscillations suggests that the contribution of magneto-exciton states derived from the first subband is small. The observed decrease of the Raman intensity with increasing temperature is also consistent with the idea of exciton delocalization. The measured value of the delocalization energy E_b = 5.3 meV does not depend on the magnetic field at least for B < 7.5 T.

Conclusion.

We have observed oscillations of the IF and LO phonon intensities in the SL in magnetic fields corresponding to resonances of the incident

photons with the energies of magneto-excitons which are the intermediate states in the scattering processes. The IF phonon intensity decreases with increasing temperature. Such behavior is connected with the temperature induced delocalization of the magneto-excitons bound near static defects which are responsible for the relaxation of the momentum conservation condition in the RS process. The results can be explained in the framework of a model of magneto-exciton localization at the islands of QW width fluctuations. It is shown that the previously observed decrease of the IF phonon intensity with increasing pumping power is mainly caused by local heating of the sample by the exciting light.

Acknowledgments - We are thankful to M.Cardona and R.Merlin for useful discussions, to N.N.Ledentsov for excitation-luminescence spectra measurements and to N.N.Faleev for x-ray measurements. This research has been supported by the Volkswagen Foundation and the Russian Foundation of Fundamental Investigations.

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