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TEMPERATURE DEPENDENCE OF RAMAN SCATTERING BY ACOUSTIC PHONONS IN A SUPERLATTICE GaAs/AlGaAs IN STRONG MAGNETIC FIELDS

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We observed simultaneously two types of Raman spectra in a GaAs/AlGaAs superlattice under high magnetic field: a folded longitudinal acoustic phonon, I_{FA}, and a structureless background, I_B. The temperature dependence of the intensities in both spectra was studied in the range 2-100K and it was established that the background spectrum is due to the first-order Raman scattering from acoustic phonons with a wide range of wave vectors.

We obtained the expression for the intensities ratio I_B/I_{FA}. It depends on the relationship between the homogeneous and the inhomogeneous broadening of the intermediate state of the photoexcited e-h pair. A rapid decrease of I_B observed at T>30K is thought to be due to the increase of the homogeneous width with temperature.

1. Introduction

Secondary emission with a structureless spectrum close to the excitation line has been reported previously in MQW structures under strong magnetic fields[1]. Its intensity increased with the magnetic field, B, and oscillations of the intensity vs. B have been observed under some conditions of the excitation. This emission was interpreted as geminate recombination of photoexcited electron-hole pairs and the width of the spectrum was attributed to the scattering of the pairs by acoustic phonons.

A gigantic enhancement of Raman scattering from the longitudinal folded acoustic (FA) phonons has been observed in a superlattice (SL) GaAs/AlGaAs 46Å/42Å under a strong magnetic field[2].

The already mentioned structureless spectrum of geminate recombination was also present as a background along with the narrow Raman lines of the first folded acoustic doublet (17.5-20.8)cm^{-1}. At helium temperatures the intensity of the background at the Raman shift of the FA mode was twice as much as the latter's peak intensity.

The oscillations of the intensities of the FA-phonon and the background spectrum vs. magnetic field had maxima at the same values of the field, this being an evidence that both spectra of Raman scattering have the same intermediate state. Let us note that the background spectrum can be treated as geminate recombination or, if we adopt another language, as a spectrum of Raman scattering from acoustic phonons with the geminate pairs (or magnetic excitons) being intermediate states in the Raman process.

In order to further investigate the nature of the background spectrum we studied its temperature dependence and compared it with that of the FA-phonon in the same sample. It was found that at T<30K both spectra have the same temperature dependence which is determined by the Bose factor n+1, were $n=(\exp(\hbar\Omega/kT-1)^{-1}$, Ω is the phonon frequency. It means that the background spectrum is also governed by the first order Raman scattering process, as the spectrum of the FA-phonon does. This result agrees with the conclusion made in Ref.3 from the comparison of the experimental and calculated Raman spectra.

Note, that a broad structureless

Raman spectrum in which different phonons of the acoustic branch are participating is natural for a single quantum well, because in this case the wave vector of acoustic phonons, q_z, is conserved to an accuracy of π/L_W, where L_W is the quantum well width.

However, in the case of a SL such spectrum is changed into that of the folded phonons due to the interference of the contributions from different wells.

Simultaneous presence of both spectra in a SL was explained in ref.3 by the imperfections of the SL structure, namely minor differences in the well widths and consequently in their energy levels. Under the condition of highly resonant excitation this boils down to the selective probing of individual wells leading to the appearance of the broad spectrum characteristic to a single well.

In the present work it was found that at T>30K the intensity of the background spectrum begins to decrease rapidly, whereas the intensity of the FA-phonon keeps growing. It was shown theoretically that the ratio of the intensities of both spectra is governed by a relationship between homogeneous and inhomogeneous broadening of the intermediate state in the Raman process. It follows from the experimental data that the homogeneous broadening grows with temperature.

2.Experimental Results

The temperature dependence of the Raman spectra was measured in the temperature range between 2 and 100K from a SL GaAs/$Al_xGa_{1-x}As$, 46Å/42Å, x=0.3 grown by MBE method. It was the same structure, which has been used in Ref.2.Raman spectra were measured with double-grating spectrometer DFS-24 in backscattering Faraday geometry in magnetic fields up to 7.5T. The spectral slit width was set at 1.5cm^{-1}. The sample was excited by the 7525Å (1.647eV) line of a Kr^+ laser. The incident power density did not exceed 5W/cm^2. Raman spectra were recorded in the circular ($\sigma_-\sigma_-$) polarization.

The peak of the exciton luminescence at T=2K was at 1.615eV and its halfwidth under the lowest power density was 1.7meV. The energy of the exciting light was a little higher than the energy of the electron-heavy hole transition (1e-1hh). The initial energy of the photoexcited electron was about 17-20meV at T=2K and B=0. i.e. it was smaller than the longitudinal optical phonon energy (36.6meV).

Raman spectra at T=2K and T=96K are shown in Fig.1.

As in Ref.2, the intensity oscillations of the background spectrum as well as of the FA-phonon vs. magnetic field were observed ,Fig.2. The oscillations are due to the resonances with the transitions between the electron and heavy hole Landau levels. Each resonance consisted of one peak. The resonance at the highest field was connected with the n=1 Landau levels. The halfwidth of this resonance at T=30K

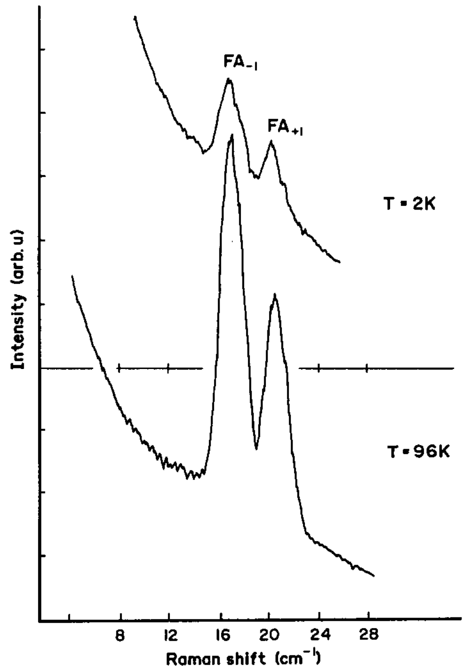

Fig.1.Raman spectra at T=2K (B=5.2T) and T=96K (B=7.5T).
Note the decrease of the background intensity with temperature.

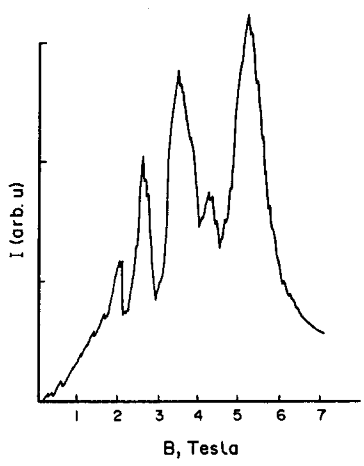

Fig.2.Magnetic oscillations of the total intensity at the Raman shift of the FA_{-1} mode at T=30K.

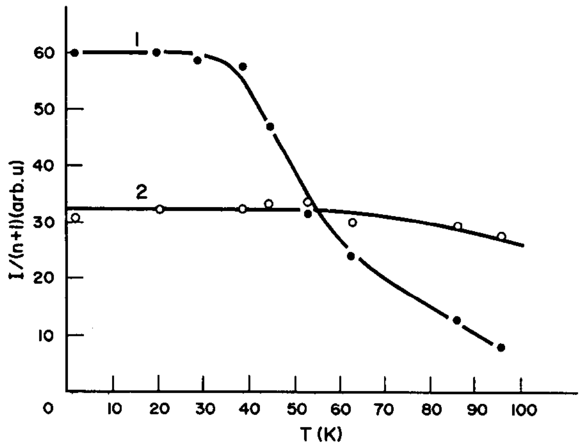

Fig.3. Temperature dependence of the reduced intensity (I divided by the Bose factor) of the background (1) and the FA-phonon (2) at the Raman shift of the FA_{-1} mode.

was ΔB=1.0Tesla, which corresponds to 3meV.

When the temperature increases the magnetic field positions of the oscillation maxima are shifting due to the temperature dependence of the band gap energy. Therefore at each temperature the Raman spectra were recorded under corresponding resonance magnetic fields, i.e. the fields which corresponded to the maximal intensity at the FA-phonon frequency.

Note, that in Fig.1 the background intensity at T=2K is twice that of the FA-phonon peak, whereas at T=96K the background intensity is one third of the phonon peak. The intensities were compared at the same Raman shift.

The temperature dependence of the reduced intensity (the intensity divided by the Bose factor, n+1) is shown in Fig.3 for the background spectrum (curve 1) and the FA-phonon (curve 2). At T<30K the reduced intensities do not depend on temperature. This signifies that the background spectrum is also caused by a first-order Raman scattering process, as the spectrum of the FA-phonon does. In the region T>30K the reduced intensity of the background starts decreasing rapidly, whereas that of the FA-phonon decreases only slightly.

3. Theory

Let us calculate the Raman intensity assuming that both the peaks related to the FA-phonons and the background spectrum are due to the first-order scattering by acoustic phonons with wave vectors parallel to the SL growth axis. The background (spectrum of "geminate recombination") arises due to the spread in energies of the excited electron-hole pairs in different wells of the SL structure. Tunneling between different wells is neglected here.

The transition matrix element for Raman scattering is given by the sum over the intermediate states of the electron-hole pair. If the energy of the exciting photon, $\hbar\omega_{exc}$, is fixed, the resonances in the magnetic field dependence of the scattering intensity exist, which correspond to each of the intermediate state, where the electron and the hole occupy the given Landau levels. The expression for the term $\mathcal{M}$, which corresponds to a particular resonance (in the Stokes region) has the form

$$\mathcal{M}=\frac{F(\Omega_q)}{(NL)^{1/2}}\frac{1}{2\pi\lambda^2}\sum M(\varepsilon_m)\exp[imL(q-2k) \tag{1}$$

where

$$M(\varepsilon_m)=[\varepsilon_m+i\Gamma/2]^{-1}[\varepsilon_m-\hbar\Omega_q+i\Gamma/2]^{-1}$$

In (1) summation is performed over individual wells of the MQW structure, which are labeled by the numbers m, q is the phonon wave number (the wave vector is directed along the growth axis of the structure), k is the wave vector of light, Ω_q is the phonon frequency,

$\lambda=(c\hbar/eB)^{1/2}$ is the magnetic length, L is the period of the structure, the factor $F(\Omega_q)/(NL)^{1/2}$ depends on the phonon frequency and includes the matrix element of the electron-phonon interaction as well as the matrix elements of interaction with the fields of the exciting and the scattered waves for a single quantum well, N is the number of wells.

Equation (1) corresponds to the back-scattering Faraday configuration. The exponential factor arises due to the phase difference between the matrix elements for light scattering from different wells. In Eq.(1)

$$\varepsilon_m=\hbar\omega_{exc}-(E_m+\beta B), \tag{2}$$

where $E_m+\beta B$ is the energy of the e-h pair in the intermediate state corresponding to the given resonance in a well m , βB is the magnetic field dependent part of this energy. At the resonance $\varepsilon_m=0$, so one may write $\varepsilon_m=\beta(B_m-B)$, where B_m is the resonance field for the m-th well. In this way we account for the difference between the wells only through the difference between the energies E_m. Just this difference is the cause of the background spectrum. If the homogeneous width Γ is small, even a small shift of the resonance will lead to a large change of the $M(\varepsilon_m)$ value in Eq.(1) and consequently destroy the coherence of the waves scattered from different wells.

The scattered intensity over a unit frequency interval is proportional to an expression

$$\Im \propto \sum_q |\mathcal{M}|^2 \, \delta(\Omega-\Omega_q)[1+n(\Omega)] \qquad (3)$$

where Ω is the Stokes shift.

If N (the number of wells in the structure) is sufficiently great and the energies E_m are randomly distributed between the wells with a probability $W(E_m-\bar{E})$, it can be shown that

$$\Im=\Im_B+\Im_{FA}$$

where $\Im_B$, the intensity of the background spectrum, is given by

$$\frac{\Im_B}{n(\Omega)+1} \propto A(\langle|M|^2\rangle-|\langle M\rangle|^2)\,\frac{1}{\bar{\Omega}} \qquad (4)$$

and $\Im_{FA}$ is the sum of narrow peaks, which correspond to the scattering from the folded acoustic phonons with the wave vectors $q_n=(2\pi n/L)+2k$, $n=0,\pm1,\pm2,\ldots$ and is given by

$$\frac{\Im_{FA}}{n(\Omega)+1} \propto \sum_n A|\langle M\rangle|^2\delta(\Omega-\Omega_{q_n}) \qquad (5)$$

In the Eq.(4) and Eq.(5) the following designations are made

$$A=\frac{|F(\Omega)|^2N}{4\pi^2\lambda^4 L}\,;\quad \bar{\Omega}=\frac{2\pi}{L}S,$$

where S is the sound velocity, and < > means the averaging over the energies E with a probability $W(E_m-\bar{E})$.

Let us note, that $E_m-\bar{E}=\bar{\varepsilon}-\varepsilon_m$ according to Eq.(2), $\bar{\varepsilon}=\beta(\bar{B}-B)$, $\bar{B}$ is the field, which corresponds to the averaged position of the incoming resonance.

If the monochromator is set at the frequency of the FA-phonon and the width of the peak is defined by the spectral slit width (i.e.the whole intensity of the peak is detected) the measured intensity will be given by:

$$I=I_B+I_{FA}$$

$$\frac{I_B}{n(\Omega)+1} \propto A(\langle|M|^2\rangle-|\langle M\rangle|^2)\,\frac{\Delta\Omega}{\bar{\Omega}}\,; \qquad (6)$$

$$\frac{I_{FA}}{n(\Omega)+1} \propto A|\langle M\rangle|^2$$

where $\Delta\Omega$ is the spectral slit width of the monochromator.

The analysis of Eq.(6) shows that there is a strong dependence of the intensity, I_B, on the relationship between the homogeneous and inhomogeneous broadening, as follows also from physical considerations. If the homogeneous width is much smaller than the inhomogeneous one, $I_B>I_{FA}$. When the homogeneous width increases the value of the ratio I_B/I_{FA} changes for an inverse one.

Fig.4 presents results of numerical calculation of the ratio I_B/I_{FA} vs. Γ/γ, where γ is the inhomogeneous width. We

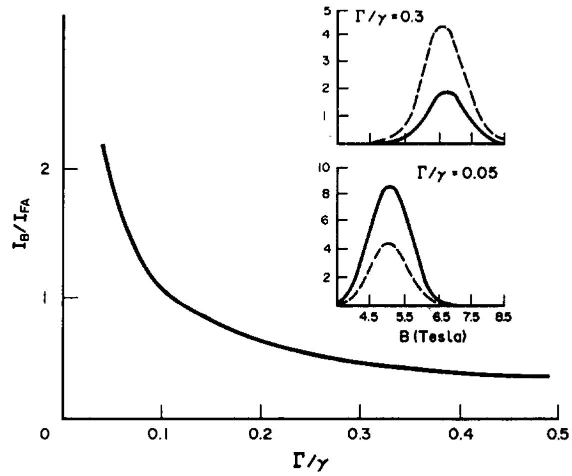

Fig.4.Intensities ratio I_B/I_F vs. Γ/γ calculated according to Eqs.(6) with the parameters: γ=3.2meV, $\bar{\Omega}$=2.1meV, $\Delta\Omega/\bar{\Omega}$=0.09, β=3meV/T. The inset shows magnetic field dependence of I_B (solid curve) and I_{FA} (dashed curve) for Γ/γ=0.05 and 0.3.

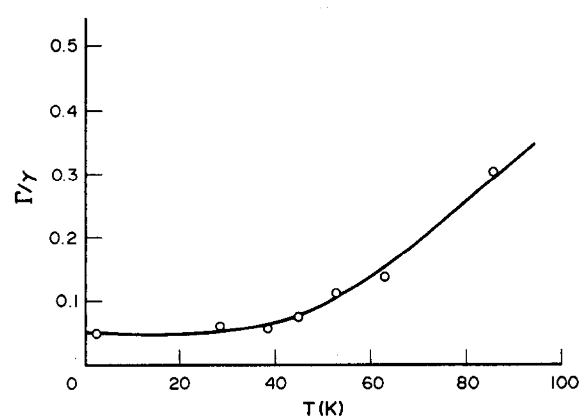

Fig.5. Temperature dependence of Γ/γ for γ=3.2meV and $\bar{\Omega}$=2.1meV.

have assumed a Gaussian profile for $W(\bar{\varepsilon}-\varepsilon)$ with a halfwidth equal to γ.

The calculation was performed with the following parameters, which correspond to the experimental situation: γ=3.2meV, $\bar{\Omega}$=2.1meV, β=3meV/T, and $\Delta\Omega/\bar{\Omega}$=0.09.

In the inset to Fig.4 the magnetic field dependencies of I_B and I_{FA} are shown, which have been calculated for two values of Γ/γ. At Γ/γ=0.05 the ratio I_B/I_{FA} corresponds to the one measured at T=2K and at Γ/γ=0.3 to T=85K.

Let us note that the incoming and outgoing resonances were not resolved in the calculated I vs.magnetic field dependence for γ=3.2meV, as can be seen from the inset to Fig.4. Only one resonant peak was observed in the experiment too.

Comparing the calculated dependence I_B/I_{FA} vs. Γ/γ with the measured one vs. temperature we could evaluate the temperature dependence of the Γ/γ. The data obtained in this way are shown in Fig.5. The solid curve in Fig.5 is the least square fit to the equation

$$\Gamma/\gamma=A+C\exp(-\Delta/kT)$$

where A=0.05, C=2.4 and Δ=17meV.

One can see that the decrease of the background intensity with respect to temperature may be explained by the increase of the homogeneous width, Γ, of the intermediate level of the electron-hole pair.

Elucidation of the process responsible for this broadening of the intermediate state with temperature needs further investigation.

Let us note that the inhomogeneous width, which causes the appearance of the background spectrum, may be due not only to the interwell width fluctuations but also to the width fluctuations within each individual well (intrawell fluctuations). In the latter case acoustic phonons with the nonzero q_x, q_y components can participate in the scattering process.

4.Conclusion

In summary, we have observed two types of Raman scattering spectra in a superlattice GaAs/AlGaAs under high magnetic fields: the spectrum due to longitudinal folded acoustic phonons and a structureless background and studied the temperature dependence of the intensity of both spectra.

From the temperature dependencies a conclusion was drawn that the background spectrum, I_B, is due to the first-order Raman scattering from acoustic phonons.

A strong decrease of intensity of the background spectrum vs. temperature was observed for T>30K and explained as due to the increase of the homogeneous width with temperature.

A theoretical expression for I_B/I_{FA} as a function of the relationship between the homogeneous and the inhomogeneous broadening of the intermediate state of the excited electron-hole pair was obtained. Along with the measured temperature dependence of I_B/I_{FA} it allowed us to elucidate the temperature dependence of the homogeneous width.

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