Resonance Raman scattering induced by interface roughness in a short-period GaAs/$Ga_{1-x}Al_xAs$ superlattice

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We have studied by means of resonant Raman scattering, photoluminescence, and photoexcitation the interface (IF) modes, as well as the electronic system and the coupling mechanism between electrons and phonons in a short-period superlattice. We observe the resonance of Raman scattering by the IF modes with the $n=2$ electronic transition. From explicit calculations of the Raman cross section we conclude that the coupling mechanism is the intraband Fröhlich interaction induced by the roughness at the interfaces.

INTRODUCTION

The interface (IF) mode in superlattices (SL's) has been the subject of many studies. The IF-mode frequencies and their dispersion curves were calculated long ago,[1] and were experimentally observed for the first time by Merlin *et al.*,[2] and more recently by other groups in GaAs/AlAs,[3] GaAs/$Ga_{1-x}Al_xAs$,[4] CdTe/$Cd_{1-x}Mn_xTe$,[5] and GaSb/AlSb (Ref. 6) SL's.

The experimental technique which is preferentially used is resonance Raman scattering (RRS). This technique involves the coupling of light to normal vibrational modes through the electronic system. Therefore, a lot of information is included in the RRS results, such as the nature of the different excitations, or the electron-phonon–coupling mechanisms. The IF modes are usually forbidden in the backscattering configuration because of the momentum conservation. However, this momentum selection rule will be considerably relaxed if the scattering process is induced by defects at the interface. Under this condition phonons with all **k** vectors become allowed. The dominant interaction process under these circumstances under resonance conditions is the defect-induced Fröhlich interaction.[3] It is not clear whether the Fröhlich interaction is induced by impurities distributed homogeneously in the SL, or by the interface roughness.[7]

In this paper we measured the RRS by the IF modes with the $n=2$ transition in a short-period SL. In this type of SL the $n=2$ electronic wave function is unconfined; therefore the properties of this excitation are sensitive to the IF roughness. We used two different calculational approaches to fit the experimental data. Both methods of calculation indicate that the scattering mechanism is an intraband Fröhlich interaction induced by the roughness at the interface.

EXPERIMENT

Our measurements were performed at 80 K on a [001]-oriented SL grown by molecular-beam epitaxy (MBE), defined by $d_1(GaAs)=42.1$ Å and $d_2(Ga_{1-x}Al_xAs)=8.4$ Å with $x=0.35$. The as-grown SL is typically p type ($p\leq 10^{14}$ cm^{-3} for GaAs and $p\leq 10^{15}$ cm^{-3} for $Ga_{1-x}Al_xAs$). The total thickness consists of 100 periods. Raman scattering in the range of the acoustic modes show three sets of folded modes which reflect the good superstructure quality. In order to check the influence of the interface broadening on the IF vibrational modes, several pieces of the sample were annealed. The thermal annealing was carried out at a temperature of 850 °C for 10-, 30-, and 60-min durations. Before thermal treatment the samples were rinsed in trichloroethene, acetone, methanol, and deionized water, in that order, and then air dried. The surface of the sample during annealing was protected from arsenic loss by close contact with a GaAs wafer. In order to prevent oxidation, the annealing was done under a vacuum of 10^{-6} torr in a quartz tube. Raman spectra excited by both a cw Ar^+ laser and a Rhodamine-6 G dye laser were recorded with a Spex Industries double monochromator. The laser intensity was kept low, 10 W/cm^2, to prevent heating effects. We used the backscattering configuration at the (001) face.

RESULTS

Figure 1 shows several Raman spectra obtained for different laser energies far and close to resonance for a virgin sample (solid line) and for a sample annealed for 30 min at 850 °C (dashed line). The spectra off resonance were taken in the depolarized $Z(X,Y)\bar{Z}$ configuration. The LO mode is related to the GaAs well, and the LO_1 and LO_2 modes are the vibrations in the $Ga_{1-x}Al_xAs$ barrier. The spectra near resonance were taken in the polarized $Z(X,X)\bar{Z}$ configuration, in which case each spectrum is composed of two peaks S_1 and S_2, the frequencies of which are laser energy dependent. After annealing, far from resonance the Raman peaks labeled LO′,LO_1',LO_2' are shifted towards lower frequencies, the shift being

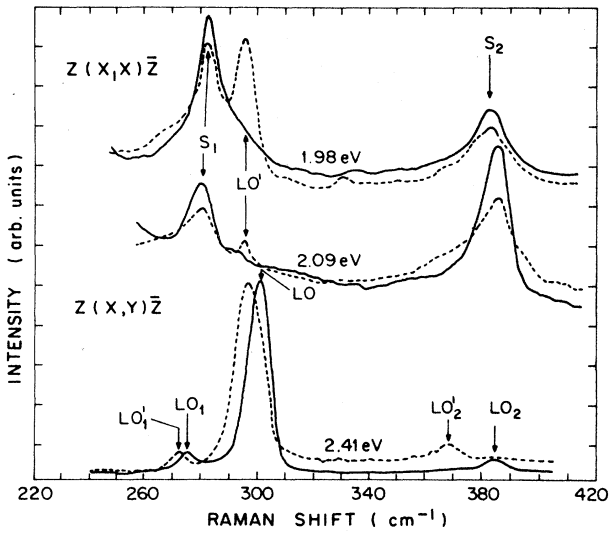

FIG. 1. The solid lines represent the Raman spectra of the sample before annealing; the dashed lines represent the Raman spectra of the annealed sample at 850 °C for a duration of 30 min. The upper two spectra were taken in the $Z(X,X)\bar{Z}$ configuration with laser energy equal to 1.98–2.090 eV, under resonance conditions. The lowest spectrum taken in the $Z(X,Y)\bar{Z}$ configuration was done so with a 2.41-eV energy laser.

more pronounced for the LO_2' peak. These post-annealing results described so far are similar to the ones reported and explained in Ref 8, and therefore will not be discussed further. Under resonant conditions, the Raman spectrum is composed of three peaks—LO', S_1, and S_2. The spectra taken in the polarized and depolarized configurations were essentially the same. The phonon frequency dependence on the laser energy is summarized in Fig. 2. The open circles represent the Raman shift of the virgin sample and the solid circles represent that of the sample annealed for 30 min at 850 °C.

The peaks S_1 and S_2 seen under resonant conditions are interpreted as the IF modes for two reasons. First, because they exhibit dispersive behavior as a function of the laser energy.[4] This is a characteristic feature of the IF modes only: the IF-mode dispersion curve is scaled with the mini-Brillouin-zone. Therefore, the IF-mode frequencies are sensitive to the small changes of the incident-light wave vectors, especially when the refractive index is dramatically altered, which happens near sharp electronic transitions. Secondly, the IF-mode frequencies at a given laser energy are not affected by the broadening of the interface, caused by a 30-min thermal annealing. The nonsensitivity of the IF mode to the broadening was attributed to the field-penetration properties of the electrostatic IF modes. This, in turn, implies that the IF modes' frequencies are not sensitive to the interface width.[8] For an annealing time of 60 min the vibrational mode character is basically that of an alloy, and the IF modes disappear.

In our case we calculate the dispersion of the IF mode using the electrostatic continuum model.[3] The results of our calculation do not fit the experimental results. This may be due to the very narrow width of the barrier which does not allow the proper use of the macroscopic continuum model.

In order to find the energies and the character of the actual electronic transitions, we measured the photoluminescence and the excitation spectra of the sample. In Fig. 3(a) we present the photoluminescence spectrum at 77 K when the sample is excited by the 2.41-eV energy laser. The strong luminescence peak labeled W_1 is the radiative transition associated with the first excitonic state. This line is accompanied by a weaker band, which is believed to be a bound exciton (BE) associated with W_1. The second weak luminescence peak W_2 is the radiative transition associated with the second excitonic state, and it is also accompanied by a BE band. The excitation spectrum was measured at the W_2 peak. A strong absorption band starting at 2 eV is seen, which corresponds to the second band. The RRS intensity is also peaked at 2 eV. In Fig. 3(b) we present the photoluminescence measurement of the annealed sample; the W_1 line is shifted slightly to higher energy and the W_2 line disappears.

These experimental data can be interpreted in the framework of the Krönig-Penney model, which should be valid in our case [d(GaAs) > 30 Å].[9] This calculation is

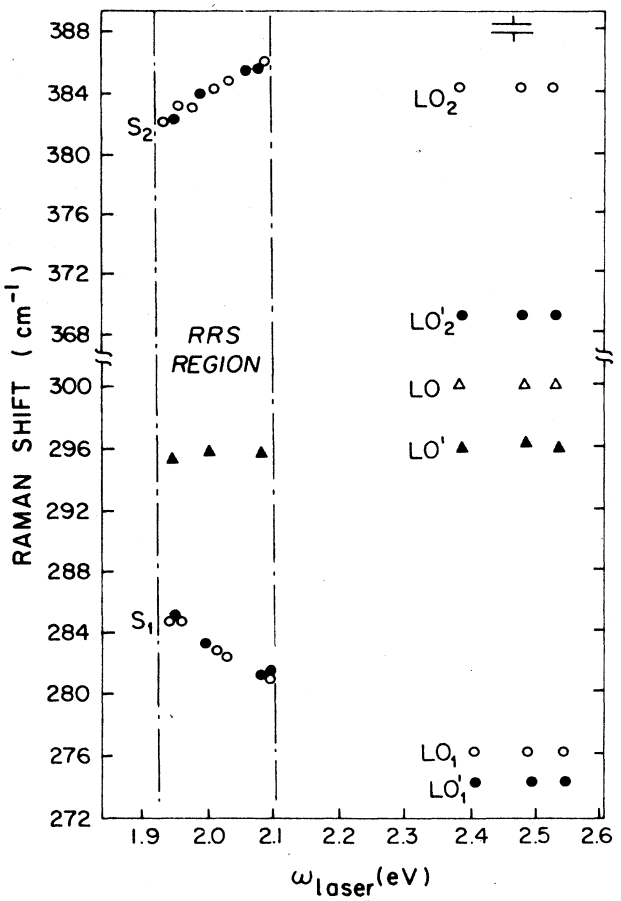

FIG. 2. Raman shift as a function of the laser energy. The open circles represent the Raman shift of the virgin sample and the solid circles represent that of the sample annealed for 30 min at 850 °C.

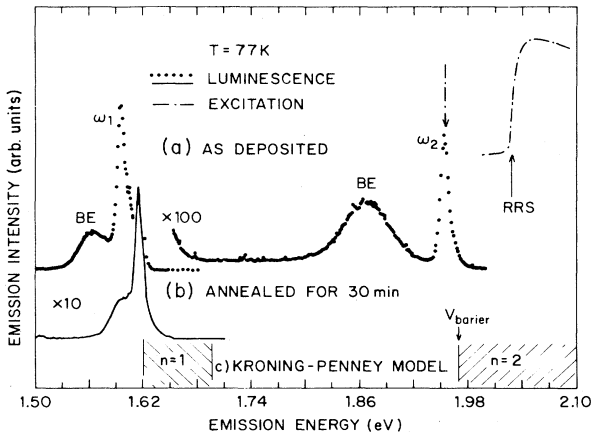

FIG. 3. (a) Luminescence and excitation spectra of the sample before annealing. The excitation spectra were monitored at W_2 energy. (b) Luminescence spectra of the annealed sample. (c) The two bands were calculated using the Kronig-Penney model (Ref. 10).

shown in Fig. 3(c) and fits the experimental results well.

The resonant profiles of IF modes S_1 and S_2 (when the laser energy equals the $n=2$ transition) are plotted in Fig. 4(b). Only the outgoing channels could be seen, which indicates that the coupling mechanism is closely related to the impurity- or disorder-induced Fröhlich interaction.[10] This mechanism can also explain the mixing between the $Z(X,X)\bar{Z}$ and $Z(X,Y)\bar{Z}$ configurations under resonance conditions. The additional elastic scattering of the free carriers by the defects can induce the depolarized scattering.[11]

DISCUSSION

There several features associated with the coupling between the IF modes and the electronic wave functions that will be discussed.

As can be observed when we examine the width of the S_1 and S_2 peaks in Fig. 1, the lines are not as broad as the IF modes previously observed,[2-4] especially the S_2 line. This can be related to the high selectivity introduced by the enhanced exciton-phonon coupling. This effect can take place if the IF mode and the exciton share the same spatial extent. In order to justify this argument we have calculated the spatial variation of the electronic envelope function associated with the minimum of the $n=2$ electronic band, by using a new formalism of the Kronig-Penney model derived by Cho and Pruenal.[12] We also calculated the spatial amplitude of the IF-mode wave function in the direction normal to the interface.[13] These results are plotted in Fig. 5; as one can see, the spatial probability distribution of the two excitations overlap and, therefore, enhanced exciton-phonon coupling is expected. This may explain why the S_1 and S_2 modes are narrow.

The main feature we wish to discuss is related to the RRS profile as seen in Fig. 4 where only the outgoing resonance is observed. In order to specify the physical quantities responsible for the absence and existence of the incoming and outgoing resonances, respectively, we cal-

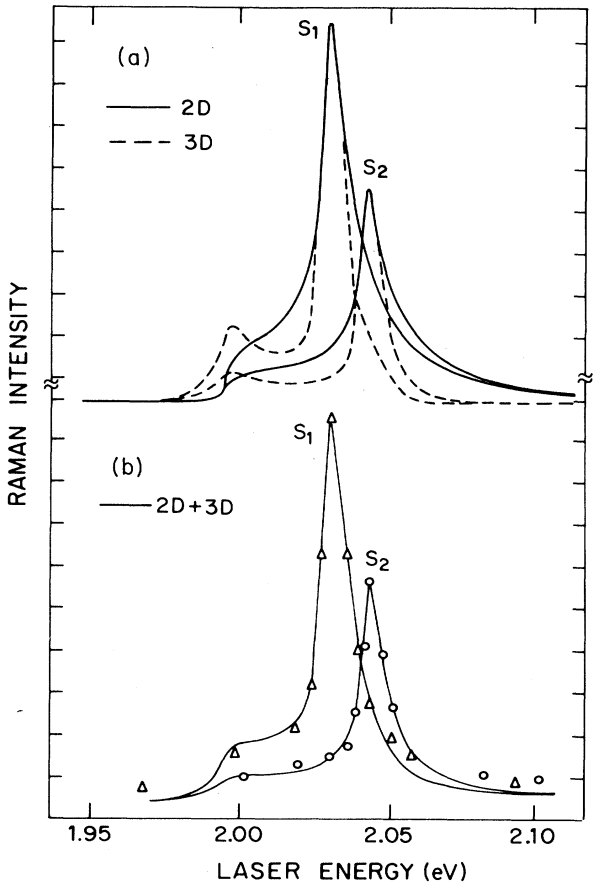

FIG. 4. (a) The solid-line profile represents the calculated Raman cross section using the impurity-induced Fröhlich-interaction theory for a 2D system. The dashed line is the calculation using the same theory for a 3D system. (b) The experimental results of S_1 and S_2. The solid line is the average of the two theories, 2D+3D.

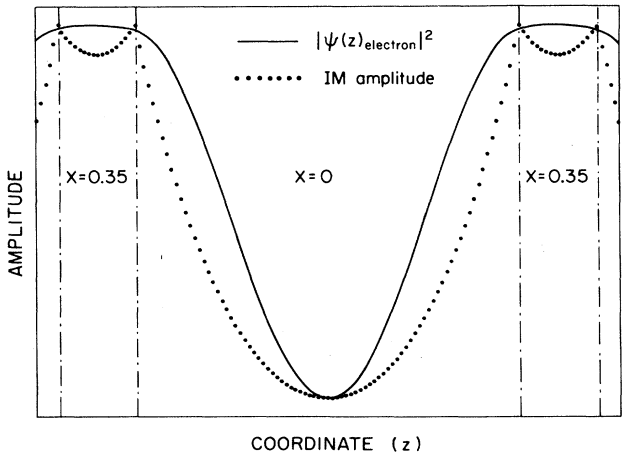

FIG. 5. The electron-envelope-function probability in the well ($X=0$) and in the barrier ($X=0.35$) (solid line). The dashed line represents the interface-mode amplitude.

culate the Raman cross section and compare it to the experimental data.

There are two different ways to calculate the Raman cross section using the impurity-induced Fröhlich-interaction theory. First, the original theory was derived by Menendez and Cardona[10] and applied to three-dimensional (3D) systems. Secondly, a modified theory applied to 2D systems (i.e, multiple quantum wells) was derived by Kauschke *et al.*[14]

It is well known that in short-period SL's (5–50 Å) the electronic system exhibits a continuous transition from 3D to 2D character.[15] A detailed calculation using either method of calculation shows that for a large Fermi-screening-potential parameter ($q_f \simeq 0.04$ Å^{-1}) the ratio between the outgoing and the incoming resonance is ~ 6, which is consistent with the experimental data. The calculated profile is shown in Fig. 4(a). In Fig. 4(b) we present the best fit to experimental data using the mean profile between the 2D and 3D cases.

The parameter q_f has a very important physical meaning. It is related to the effective number of screeners (n_s) in the system which can be either impurities or defects associated with interface roughness. From the fitting procedure we obtain $q_f \simeq 0.04$ Å^{-1} and we find that

$$n_s = \frac{3}{32\pi} q_f^3 \cong 10^{18} \text{ cm}^{-3} . \quad (1)$$

This is about 3 orders of magnitude above the expected impurity concentration. Therefore we rule out impurities as a major source of screeners. On the other hand, the roughness of the interfaces can provide the unconfined electron-hole pair with any momentum q required for the double-resonance condition, especially when the spatial variation of the excitonic envelope function has a maximum at the interfaces as in our case (see Fig. 5). Another argument to support our interpretation is the appearance of the LO′ Raman line after 30 min of thermal annealing (see Fig. 1). This effect may be related to the broadening of the interface which extends into the GaAs well and renders Raman active the LO′ mode.

In conclusion, we have shown that in short-period SL's the coupling mechanism between the IF mode and the electronic wave function is closely related to the overlap between the electronic- and vibrational-amplitude wave functions. In addition, it has been shown that the most preferential factor in inducing the breakdown of the selection rules on q is the interface roughness. Finally, we want to remark that this feature of the SL's may be used as a way to estimate the IF roughness in SL's.

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