Confined-to-Propagating Transition of LO Phonons in $GaAs/Al_xGa_{1-x}As$ Superlattices Observed by Picosecond Raman Scattering

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We report the first observation of a confined-to-propagating transition of LO phonons in $GaAs/Al_xGa_{1-x}As$ superlattices (SL's). Our samples are two series of SL's with constant GaAs layer widths (L_z) and x, while the $Al_xGa_{1-x}As$ layer widths (L_b's) varied. Using picosecond Raman scattering, we have observed a sudden increase in the generation rate of the Raman-active hot phonons as L_b decreased over a narrow range for both series of samples. We demonstrate that this abrupt increase is due to the sharp transition from the so-called confined to the propagating LO phonons. From our results we have determined the GaAs LO phonon penetration depth into the $Al_xGa_{1-x}As$ layers.

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The problem of localization of elementary excitations in solids is of fundamental importance in physics [1]. Recently, there has been considerable interest in the confinement of phonons in $GaAs/Al_xGa_{1-x}As$ superlattices (SL's) and multiple quantum wells (MQW's) [2–9]. This has been particularly so in connection with the ultrafast hot-carrier relaxation in these systems through the Fröhlich electron–LO-phonon interaction [10–16]. The nature of the LO phonon confinement affects the electron–LO-phonon scattering rate which governs much of the ultrafast hot-electron relaxation. It is well known that optical phonons in GaAs/AlAs SL's or MQW's are almost perfectly confined due to the large difference in the frequencies of the optical phonons in these two materials [2,8]. The optical phonons confined within the GaAs layers (i.e., wells) are believed to penetrate only one monolayer into the AlAs layers (barriers) and vice versa [2–5]. When the barrier is an $Al_xGa_{1-x}As$ alloy instead of AlAs, the problem of phonon confinement becomes more complicated [6–8]. For instance, the penetration depth of the GaAs LO phonons into the $Al_xGa_{1-x}As$ barriers is still unknown. Since there exists a partial overlap in energy between the GaAs and GaAs-like LO modes, especially for small x, it is possible that the LO phonon wave functions extend over several periods [7,8]. This leads to the concept of propagating optical phonons much like that of folded acoustic phonons [2,7]. The question of whether the optical phonons are confined or propagating should thus be answered depending on the value of x as well as the layer widths.

In this Letter we discuss the dependence of confined and propagating optical phonons in SL's on the parameters L_b and x. Using a time-resolved Raman-scattering technique [17–23] we have investigated two series of $GaAs/Al_xGa_{1-x}As$ SL's with $x=0.4$ or $x=1$. Each series was grown with a fixed $L_z=100$ Å while the L_b's varied. Our most exciting findings are as follows: (1) LO phonons in these SL's make a transition from the confined to the propagating regime when the barrier width becomes thinner than 20 Å for $x=0.4$ and 11 Å for $x=1$, and (2) the transition is very sharp, occurring within 5 Å of the barrier thickness for both series.

Previously, most of the information concerning the confinement of LO phonons in SL's and MQW's has been obtained from cw Raman scattering [2–9]. In a cw Raman experiment, the confinement is observed in the form of a shift in the LO phonon frequency to lower energy, or multiple peaks with down-shifted frequencies [8]. However, the frequency shift is too small to observe unless the quantum well is much thinner than 100 Å due to the relatively flat LO phonon dispersion curve. It is also very difficult to resolve the question of confined or propagating LO phonons by cw Raman scattering even when the wells (or barriers) are substantially thin such that multiple peaks corresponding to different points in the Brillouin zone are observed. Another technique to probe phonon confinement in SL's and MQW's has been discussed recently by Kim and Yu [11,21–23]. In this technique, referred to as one-beam excite and probe, a nonequilibrium LO phonon distribution is created by hot electrons photoexcited by picosecond or subpicosecond laser pulses.

In bulk materials, the Raman-active wave vector ($q=q_0$) is determined by the energy-momentum conservation of the scattering process. In our backscattering configuration, q_0 is equal to 9×10^5 cm^{-1}. For confined optical phonons, q is primarily determined by the L_z's [2,9]. Away from resonance, the first quantized LO phonon mode is the most Raman active [2]. This corresponds to a $q=\pi/L_z\approx3\times10^6$ cm^{-1} for the GaAs LO mode of our samples. This difference in q between bulk and confined phonons has a significant effect on the phonon generation rates. This is due to the $1/q$ dependence of the Fröhlich electron–LO-phonon interaction Hamiltonian which is responsible for the nonequilibrium phonon distribution. The $1/q$ dependence results in a factor of $[(3\times10^6\ \text{cm}^{-1})/(9\times10^5\ \text{cm}^{-1})]^2\approx10$ reduction in the generation rate of the confined LO phonons compared with that of bulk LO phonons.

Using this technique, Kim and Yu have recently observed a continuous reduction of the hot-phonon generation in a series of GaAs/AlAs samples with decreasing well widths [11,21]. This observation was explained by the increasing Raman wave vector with decreasing L_z's, thus resulting in a small generation rate due to the $1/q$ dependence. Unlike confined phonons, propagating phonons can have Bloch wave vectors equal to those in bulk materials [7,8]. This results in a bulklike generation rate of the Raman-active phonons. Since our picosecond Raman-scattering technique is very sensitive to the magnitude of q, it can be an ideal tool to investigate whether optical phonons are confined or propagating in SL's.

Our experiments were performed at 15 K on a series of $GaAs/Al_xGa_{1-x}As$ SL's grown by molecular beam epitaxy on [100]-oriented GaAs substrates. The nominal sample parameters were ($x=0.4$, $L_z=100$ Å, and $L_b=5$, 10, 15, 20, 25, 30, 40, or 50 Å) and ($x=1$, $L_z=100$ Å, and $L_b=6$, 8, 11, 20, or 100 Å). Hot electrons were excited by picosecond pulses generated by a mode-locked and fiber-compressed Nd-doped yttrium aluminum garnet laser with a repetition rate of 82 MHz. After second-harmonic generation, the photon energy was 2.33 eV with a FWHM autocorrelation trace of ~ 2 ps. The photoexcited electron density at the maximum laser power used was estimated to be $\sim 10^{17}$ cm^{-3} by measuring the incident laser spot size on the samples. The Raman-scattering experiments were conducted in a standard backscattering configuration. A double monochromator together with a charge-coupled-device multichannel detector were used to collect the Raman signal.

Typical hot-phonon Stokes and anti-Stokes Raman spectra are shown in Fig. 1. The 6 cm^{-1} width of the laser pulses used is narrow enough to resolve both the GaAs and GaAs-like LO peaks for the $x=0.4$ series. In this Letter, we will concentrate mainly on the GaAs LO phonons unless otherwise specified. The ratio of the Stokes to anti-Stokes intensities is related to the phonon population N_q [17-23]. The existence of any resonant Raman-scattering effect on the deduced N_q has been carefully checked by measuring the Stokes intensity as a function of the lattice temperature between 15 and 300 K. This is possible since the change in the band-gap energy over this temperature range is much larger than the LO phonon energy. No significant enhancement was found except for the samples with $x=0.4$, $L_b=20$ Å and $x=1$, $L_b=6$ Å. To compensate for the resonance effect on these samples, we have performed our experiments at 170 K and used the thermal phonon population to obtain a correction factor.

In our experiments, we measure the dependence of N_q on the laser power P. This dependence is linear for the power densities we used, thereby allowing for an increase in our data redundancy. Considering the linear dependence of N_q on P, the laser-pulse duration, and the low

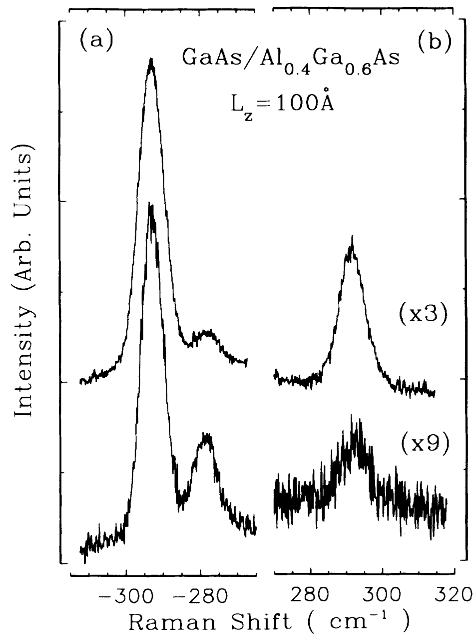

FIG. 1. Typical Raman spectra used to calculate the hot-phonon occupation number N_q. Both the Stokes and anti-Stokes components are shown, respectively, in (a) and (b). Notice the drastic change in (b) as L_b varies from 50 Å (lower spectra) to 5 Å (upper spectra). This is due to the change in the phonon confinement as discussed in the text. The smaller peak in (a) is due to the GaAs-like LO phonon in the barrier.

carrier densities used in our experiments, we can rule out the presence of any phonon relaxation effects. This is due to the fact that the Raman-active LO phonon population in our case is still rising and has not yet reached its maximum (at $\sim$2-3 ps) [17-20]. This is consistent with experiments that showed the LO phonon lifetime to be rather insensitive to alloying or superperiodicity [12,17,19]. Thus, dN_q/dP is proportional to the average generation rate of the Raman-active LO phonon for a fixed laser-pulse width. We directly use dN_q/dP as our experimental data and define it as the hot-phonon generation efficiency [11]. A plot of dN_q/dP normalized to that of bulk GaAs is shown in Fig. 2 as a function of L_b.

For SL's with $x=0.4$ and $L_b \geq 20$ Å, or those with $x=1$ and $L_b \geq 11$ Å, the dN_q/dP's are around 10% that of bulk. The fact that we observe almost the same dN_q/dP over such wide ranges of x and L_b indicates that the hot-phonon generation is rather insensitive to the detailed electronic band structure. This is probably because the momentum-energy conservation of the electron-phonon scattering, which would result in smaller dN_q/dP with increasing electron confinement [11,15,21], is relaxed due to the interface imperfections [9,12,17]. As a result, the hot-phonon generation rate is mainly governed by the number of photoexcited electrons and the magnitude of the Raman-active wave vector, not by the detailed electronic band structure. The confined phonons in both $x=0.4$ and $x=1$ series are also consistent with the off-

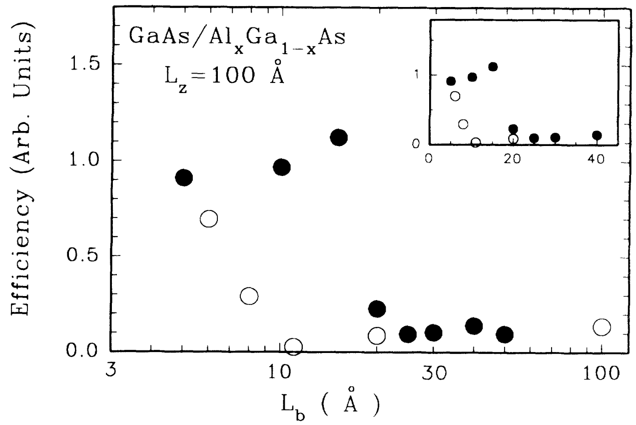

FIG. 2. The hot-phonon generation efficiency relative to bulk GaAs plotted against L_b. The solid circles correspond to the $x=0.4$ series and the open circles to the $x=1$ series. The transition from the confined to the propagating optic-phonon modes occurs between $L_b=15$ and 20 Å for $x=0.4$ and between $L_b=6$ and 11 Å for $x=1$. Inset: The data on a linear scale.

resonance Raman selection rules [2].

We have also found dN_q/dP of the GaAs-like, AlAs-like, and AlAs LO phonons of the barriers to be too small even when the barrier was sufficiently thick to allow for a significant Raman cross section. This is easily understood since the barrier phonons are now faced with the 100-Å-thick GaAs layers acting as "phonon barriers." In addition, most of our samples have barrier layers that are at most half the thickness of the GaAs wells leading to a larger q for these phonons and a much smaller dN_q/dP compared to that of confined GaAs LO phonons. From this observation, we conclude that the GaAs-like, AlAs-like, and AlAs LO phonons of the barriers are all well confined. In addition, it is also important to note that the excited hot carriers will have much lower excess energy in the barrier region leading to very small populations of the barrier phonons.

Between $L_b=15$ and 20 Å of the $x=0.4$ series, and $L_b=6$ and 11 Å of the $x=1$ series, dN_q/dP shows a sudden increase to almost the same value as that of bulk GaAs. The Raman-active nonequilibrium phonon population of bulk GaAs is already close to the peak of the hot-phonon distribution. This excludes effects such as interface imperfection and electron confinement as possible causes of this sudden increase in dN_q/dP. These factors tend to broaden the nonequilibrium distribution and thus decrease the generation efficiency relative to bulk GaAs [11,17,21]. Another possible factor is intervalley scattering which is important in picosecond and subpicosecond cooling of the hot electrons since it only takes about 100 fs or less to remove the high-energy electrons from the Γ valley [20–25]. However, Kim and Yu have recently shown that intervalley scattering rates are essentially the same for bulk GaAs and quantum wells of the order of 100 Å or larger [11]. This can be easily understood since intervalley scattering involves zone-edge phonons. Furthermore, the electrons that undergo intervalley scattering are not very efficient in producing Raman-active hot phonons since they remain mostly in the L or X valley within our time scale [26]. Therefore, the effect of intervalley scattering on our samples is practically constant.

There is the possibility that the property of the interface phonons could be crucial in determining the penetration and the coupling of the optical phonons across the barriers since some of these phonons can penetrate the adjacent layers as much as the confined phonons do [27]. In general, these modes are not Raman active unless near a strong resonance [9]. This is not the case in our experiments; therefore, we do not observe such interface phonons. In addition, Tsen *et al.* have shown in a recent Letter that the hot-electron interaction with the interface phonons in GaAs/AlAs superlattices is weak and decreases with increasing L_z [28]. In our samples with large L_z (100 Å), we expect the contribution of the interface phonons to our measured GaAs LO phonon population to be relatively small.

Photoluminescence excitation experiments were also performed on all our samples to study their electronic properties. Using Schulman and Chang's tight-binding method, we have calculated the electronic band structure of these samples [29]. Our results show no abrupt change in the electronic confinement, and the electronic transitions are rather gradual as L_b varies. Because of this and the sharpness of our observed transitions we can rule out any change in the electronic confinement as a possible explanation of our results.

From all of the above considerations, we conclude that the bulklike nonequilibrium phonon population observed in some of our SL samples is only possible if we have bulklike, extended optic-phonon wave functions. Therefore, the sudden increase in dN_q/dP is a clear indication of a sharp transition from the confined to the propagating optical phonons. The transition occurs when the LO phonon wave functions of adjacent wells begin to have phase coherence by coupling to each other in the barriers. This is the first time such a transition from confined to propagating phonons has been observed. To our knowledge, the transition we observe is thus far the sharpest localized-to-delocalized transition of any elementary excitations in SL's where only the layer widths are varied. The two observed transitions are rather sharp and reflect the unique capability of time-resolved Raman scattering in probing the extent of LO phonon wave functions.

To show that the phonon generation rates of the propagating optic phonons can be indeed bulklike, we consider a wave function of a propagating Raman-active mode that can be written in the following Bloch form: $\psi=\exp(iq_0z)u(z)$, where $u(z)$ has the periodicity of the superlattice and q_0 is equal to that of bulk GaAs [7,8]. This mode is dominated by the bulklike Fourier component $\exp(iq_0z)$ when $u(z)$ corresponds to the first quantized mode (i.e., that half-wavelength mode with its peaks in the center of the GaAs wells). In the limiting case of $L_b=0$, we obtain the bulk wave function since $u=1$ in this case. Therefore, the phonon generation rate of the propagating Raman-active mode is similar to that of bulk, consistent with our results.

The barrier widths at which these two transitions occur are very important experimental points. From these we are able to estimate the LO phonon penetration depth λ into the barriers. We do this by taking half of the barrier widths as our λ's since the LO phonons penetrate the barriers from both sides. This gives us a λ of about 1.5 ML (monolayer) when $x=1$ and about 3.5 ML when $x=0.4$. To our knowledge these are the first direct experimental estimates of λ reported. It is important to note that the larger penetration depth observed for the $x=0.4$ series is consistent with its lower "phonon barrier height" (i.e., the 15 cm^{-1} of energy difference between GaAs and GaAs-like LO phonons). Our result for λ on the $x=1$ series is in good agreement with the theoretically known value of 1 ML [3,5]. The slightly larger value measured can be attributed to interface imperfection which is common in SL's, thus making our measured value an upper estimate for λ. However, for the $x=0.4$ series no such theoretical work is available, probably due to the complex nature of the alloy barrier. Kobayashi and Roy have predicted the existence of bulklike propagating LO phonons using a force-constant calculation on a sample with $L_z=42$ Å, $L_b=8$ Å, and $x=0.3$ [6]. This is consistent with our experimental results. More detailed theoretical studies on the phonon properties in and near the alloy barriers including the interaction of the interface and bulk phonons are much needed.

In conclusion, we have performed a systematic study of the generation of hot LO phonons in $GaAs/Al_{0.4}Ga_{0.6}As$ and GaAs/AlAs SL's as a function of L_b. Using time-resolved Raman scattering we have observed a sharp transition from confined to propagating LO phonons in both series of samples. Estimates of the LO phonon penetration depth were also obtained from the observed transitions. This is the first time such a transition and the penetration depth of LO phonons into $Al_xGa_{1-x}As$ barriers have been reported. We have therefore shown that the question of confined or propagating optic phonons in SL's should be answered carefully depending on the barrier widths and the alloy concentrations.

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