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Initial generation of hot LO phonons by photoexcited hot carriers in GaAs and $Al_xGa_{1-x}As$ alloys studied by picosecond Raman spectroscopy

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The initial generation of hot LO phonons by photoexcited hot carriers is studied with picosecond Raman spectroscopy in GaAs and a series of $Al_xGa_{1-x}As$ samples with $0<x<0.4$. A rapid decrease in the occupation numbers of the GaAs-like and AlAs-like LO-phonon modes is observed as x is increased. This decrease cannot be explained if the electrons excited from the light- and heavy-hole bands were the primary source of generating the hot phonons. It is shown that most Raman-active hot LO phonons are initially generated by the photoexcited electrons originating from the split-off band, when photon energies of 2.33 eV and pulse durations of 1.5 ps are used. We have used a model assuming the instantaneous thermalization of electrons in the Γ valley which are photoexcited from the split-off hole band. Our experimental results are in good agreement with this calculation.

The initial relaxation of hot carriers in polar semiconductors is determined by the interplay between various scattering processes such as carrier-carrier scattering, intravalley carrier–LO-phonon scattering via the Fröhlich interaction, and intervalley scattering. Carrier-carrier scattering is mostly determined by the carrier density whereas intervalley scattering is very sensitive to the excess energy of an electron. Therefore, depending on the density and excess energy of the carriers, either carrier-carrier or intervalley scattering may dominate the initial relaxation process. In the extreme case of very low carrier density ($<10^{16}$ cm^{-3}) and low excess energy, the Fröhlich intravalley scattering may dominate the initial relaxation process. Picosecond (ps) and femtosecond (fs) optical spectroscopies have provided much of the information about these relaxation processes.[1-12] Both intervalley and intravalley scattering rates in several III-V systems have been determined using picosecond and subpicosecond Raman spectroscopy. In most of the time-resolved Raman experiments, the nonequilibrium population of the Raman-active LO-phonon mode is created by photoexcited hot carriers. Those nonequilibrium phonons are probed either as a function of time delay[1-4] or at a fixed time delay varying other parameters such as the laser photon energy or the carrier density.[5,6] Kash, Tsang, and Hvam determined the Fröhlich scattering time in GaAs using a subpicosecond time-resolved Raman technique.[1] Collins and Yu have investigated the intervalley scattering rates in GaAs with picosecond Raman scattering.[5] Kim and Yu have recently determined the intervalley scattering rates in GaAs and $In_{1-y}Ga_yAs$ using subpicosecond Raman scattering.[6,7] In some of these experiments, a simple cascade model was used to analyze the data.[1,5] In this model, the effect of intervalley scattering was sometimes excluded even when the photoexcited electron energy was sufficient enough to enable the intervalley scattering.[1] In most of the analysis of the time-resolved Raman experiments, the generation of the nonequilibrium LO phonons (hot phonons) was assumed to be primarily from the electrons in the Γ valley excited from the light- and heavy-hole bands.[1,5,6] However, in a recent Femtosecond luminescence experiment in GaAs, it was suggested that the majority of electron in the Γ valley, within a few picoseconds after photoexcitation, come from the split-off band when the photon energy is ≥ 2 eV.[8] Most of the photoexcited electrons originating from the light- and heavy-hole bands are quickly transferred to the satellite valleys because of their large excess energy and the ultrafast intervalley scattering times. At a carrier density $\sim 10^{17}$ cm^{-3} these photoexcited carriers will thermalize, by carrier-carrier scattering, to a nonequilibrium Fermi-Dirac distribution. Therefore, it is possible for a small portion of electrons photoexcited from light- and heavy-hole bands to remain in the Γ valley because of thermalization. If we consider thermalized hot electrons excited from the split-off band, most of these will not have enough excess energy to transfer to the satellite valley through the intervalley scattering. Therefore, a significant fraction of the hot phonons initially generated by the intra-Γ-valley Fröhlich scattering should come from the electrons which have been photoexcited from the split-off band.

In this paper, we present a systematic study on the generation of GaAs-like hot LO phonons in $Al_xGa_{1-x}As$ al-

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loys by hot carriers excited with picosecond pulses having an average photon energy of 2.33 eV. We have also studied the AlAs-like modes in several samples and found results analogous to the GaAs-like modes. Therefore, we will only discuss the results of the GaAs-like LO phonons unless otherwise stated. A decrease of more than a factor of 100 in the GaAs-like LO hot-phonon occupation number is observed in the alloys as x changes from 0 to 0.4. We found that this dramatic decrease can be explained only if the electrons from the split-off hole bands were the primary source of hot phonons. Electrons excited from the light- and heavy-hole bands were found to be very inefficient in generating hot LO phonons because intervalley scattering is much faster than the intravalley scattering.

We have measured Raman spectra of the GaAs- and AlAs-like LO phonons in a series of $Al_xGa_{1-x}As$ alloys grown by molecular-beam epitaxy (MBE). The values of x range from 0.0 to 0.39 within the series. These values are determined by photoluminescence measurements at 20 K and using $E_g = E_{GaAs} + 1.45x - 0.25x^2$.[13] The Raman spectra were obtained at 20 K in a standard backscattering geometry. A [110] polarization of the excitation light and nonpolarized scattering light collection were used. A Spectra Physics mode-locked and frequency-doubled YAG laser operating with an 82-MHz repetition rate and a pulse duration of 1.5 ps was used as the source of excitation in the picosecond Raman measurements. A Photometrics liquid-nitrogen-cooled CCD and Spex 1403 spectrometer were used to detect the Raman signal. Our system allows us to detect a hot-phonon population as small as 0.003.

A one-beam–excite-and-probe picosecond Raman-scattering technique has been used to determine the LO-phonon occupation number.[14] The cooling of photoexcited hot carriers generates LO phonons which are probed by the same excitation pulse. The experimental Raman-scattering intensities of the Stokes (I_S) and anti-Stokes (I_{AS}) lines determine the LO-phonon occupations number given by $N_q = 1/(I_S/I_{AS} - 1)$, since there is no appreciable resonant Raman-scattering effects within our samples at an incident photon energy of 2.33 eV. The hot-phonon lifetimes in GaAs, $Al_xGa_{1-x}As$, and $GaAs/Al_xGa_{1-x}As$ QW's are all approximately 5–7 ps,[1,3,15,16] and the time scales that the hot electrons emit LO phonons via the Fröhlich interaction are about 0.15–0.2 ps.[1,17] Therefore, the 1.5-ps duration time of the laser pulses is suitable for pumping and probing the hot-phonon population. In our experiments we measure the dependence of N_q on the incident light power, P. At the power densities used in our experiments N_q vs P is linear, so our measurements increase the data redundancy and reduce the experimental errors. This is consistent with the report that N_q is linear with the electron densities up to 2×10^{17} cm^{-3}.[1] It has also been reported that N_q reaches its maximum value in 2–3 ps.[1,5,6,18] Considering the pulse duration and carrier densities used in our experiments, the linearity of N_q with P implies that the Raman-active LO-phonon occupation number has not reached its maximum so that there are no relaxation effects.[1,6] Thus $\partial N_q/\partial P \propto \langle \partial N_q/\partial t \rangle$ is valid for a constant laser pulse width, and we directly use the experimental $\partial N_q/\partial P$ values, defined as the phonon generation efficiency.[10] Since N_q is linear with P, the ratio of $\partial N_q/\partial P$ between an $Al_xGa_{1-x}As$ alloy and GaAs is certainly equivalent to the same ratio of N_q's. Therefore we can directly compare our experimental values of $\partial N_q/\partial P$ with our calculated values of N_q. The $\partial N_q/\partial P$'s relative to GaAs for the different alloy samples in our series are shown in Fig. 1 for both GaAs- and AlAs-like phonon modes.

A rapid decrease of more than two orders of magnitude in $\partial N_q/\partial P$ of the GaAs-like modes is observed when x is increased from 0 to 0.39. There are several other effects that will reduce the hot-phonon generation efficiency, although the contributions from these effects are too small to explain our results. For instance, the penetration depth of a photon at 2.33 eV will be reduced by merely 40% as x increases from 0 to 0.39.[19] The number of GaAs-like LO-phonon modes will decrease with increasing x since there exist more AlAs-like modes with larger x when the total number of modes is fixed.[18] This effect will at most reduce $\partial N_q/\partial P$ by a factor of 2 between $x=0$ and 0.39. We have also observed a significantly smaller production efficiency of the AlAs-like modes than that of the GaAs-like modes. This is because there are less AlAs-like modes than GaAs-like modes for $x<0.4$, and the minimum carrier excess energy required to emit a Raman-active AlAs-like LO phonon is larger than that for the GaAs-like modes. There is also a possibility that some of the reduction in the $\partial N_q/\partial P$ might come from the localization of LO phonons with increasing alloy concentration.[20] This is due to the fact that localization tends to result in smaller phonon

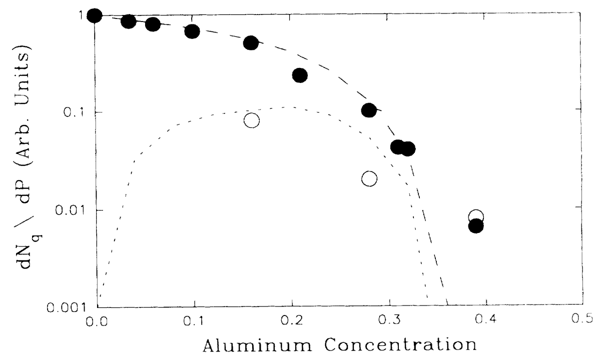

FIG. 1. The phonon generation efficiency for GaAs- and AlAs-like LO phonons plotted as a function of the aluminum concentration x. The maximum phonon occupation number measured in our experiment was less than 0.3. The density of carriers deduced from the spot size and also from the phonon occupation number was approximately 1×10^{17} cm^{-3}. The broken curves are the result of a model calculation assuming all hot phonons are created by electrons excited from the split-off band. An instantaneous thermalization of the photoexcited electrons is assumed throughout the calculation which assumes a sech^2 pulse shape (Ref. 22).

generation efficiency.[10,18] Although more work needs to be done on this subject, recent experimental and theoretical studies indicate that there is a well-defined phonon dispersion in the $Al_xGa_{1-x}As$ alloys.[18,21,22]

To understand the cause of this rapid decrease, we first consider a simple cascade model which includes intervalley scattering but excludes the electrons from the split-off hole bands. This model results in virtually identical $\partial N_q/\partial P$ throughout our samples. This is because one can normally expect a production efficiency of $\approx \tau_{\text{inter}}/(\tau_{\text{inter}}+\tau_{\text{intra}})$ LO phonons per electron for electrons which are excited from the light- and heavy-hole bands. τ_{inter} and τ_{intra} are the intervalley and intravalley scattering times, respectively, at the point of photoexcitation. Since the excess energy of these electrons above the L- and X-valley minima do not change appreciably as x varies from 0 to 0.39, the intervalley scattering rates are expected to be a slowly varying function of x. Combined with the almost constant Fröhlich scattering rate, this results in a virtually unchanging production efficiency of LO phonons as a function of x. Clearly, the two orders of magnitude decrease in the phonon generation efficiency cannot be explained with this model.

We conclude that the rapid decrease in the dN_q/dP is due to the decrease in the number of electrons in the Γ valley that do not participate in the intervalley scattering and to the reduction of excess energies with increasing x. Electrons which scatter to the L or X valleys will return to the Γ valley with a time constant of ~ 2.5 ps at room temperature.[9] This scattering time becomes appreciably longer, at least by a factor of 2, when the lattice temperature is lowered to cryogenic temperatures.[23] This is because the zone-edge phonon populations vanish at low temperature. Therefore, within our pulse width, the sources of these Γ-valley electrons are the electrons excited from the split-off hole band and the electrons from the light- and heavy-hole bands that remain in the Γ valley due to electron-electron or electron-hole scattering to lower-energy points. There is no simple way to estimate the relative contribution of these two sources. However, the number the electrons excited from the split-off hole band is expected to be larger than that due to carrier-carrier collisions at the carrier densities we use in GaAs.[24]

A production efficiency of much less than one LO phonon per electron is expected from the electrons which may emit a phonon before scattering into satellite valleys. This is because of the relatively fast intervalley scattering rates compared to that of the intravalley Fröhlich interaction at a photon energy >2 eV.[6,8,9,11] On the other hand, the electrons photoexcited from the split-off hole band can emit as many as eight LO phonons inside the Γ valley of GaAs. Actually, only about five of these LO phonons can be Raman active since the q of the phonons is much larger when emitted by the cooling of electrons near the Γ-valley minimum. Therefore, most of the reduction in the $\partial N_q/\partial P$ between GaAs and $Al_{0.39}Ga_{0.61}As$ can be accounted for from simply considering the electrons which are photoexcited from the split-off hole band. This can be easily visualized from the schematic diagrams in Figs. 2(a) and 2(b). Figure 2(a) depicts the situation when GaAs is photoexcited by 2.33-eV photons, whereas Fig. 2(b) is the same situation for the alloy with $x=0.39$. In both cases, electrons with sufficient energy to undergo the intervalley scattering emit at most one LO phonon. On average, they emit much less than one LO phonon per electron. The electrons from the split-off hole band can emit many LO phonons as described in Fig. 2(a), since they do not have enough energy to participate in the intervalley scattering process. The experimentally observed rapid decrease in the phonon generation efficiency of more than a factor of 10 between $x=0.28$ and 0.39 may also be explained once we realize that the split-off hole band is the main source of the elec-

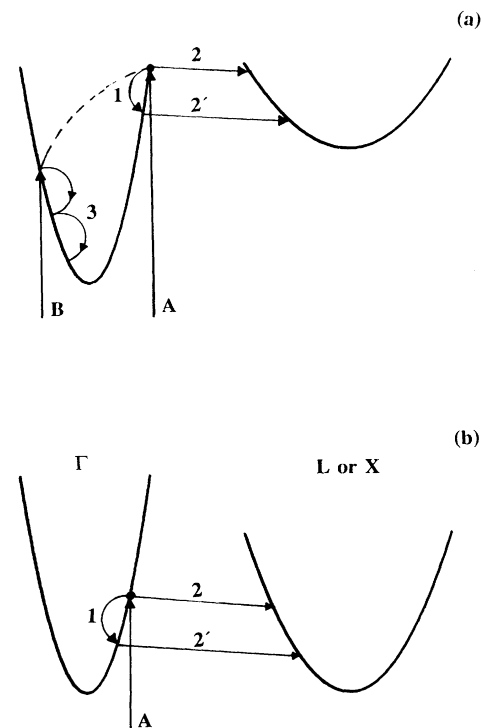

FIG. 2. (a) Schematic diagram representing the hot-phonon generation by electrons excited from various hole bands in GaAs. Arrow A denotes the photoexcitation from the heavy- and light-hole bands, whereas arrow B denotes the photoexcitation from the split-off hole band. Arrow 1 represents hot-phonon generation by electrons from the heavy- and light-hole bands. Arrows 2 and 2′ represent the intervalley scattering of these electrons, whereas the broken arrow depicts the scattering of these electrons by carrier-carrier collisions to the lower-energy Γ points. The hot-phonon generation by the electrons from the split-off hole band is represented by arrow 3. Since the electrons from the split-off hole band do not participate in the intervalley scattering because of lower excess energy, they remain in the Γ valley to generate most of the Raman-active hot LO phonons. (b) Hot-phonon generation in $Al_{0.39}Ga_{0.61}As$ where the minima of Γ, L, and X valleys are almost degenerate. Because of the larger band gap (2.02 eV), no electrons from the split-off hole band are excited. All photoexcited electrons can undergo the intervalley scattering. As a result the phonon occupation numbers observed at this alloy concentration are only 1% of the bulk GaAs value. The arrows have the same labeling as in (a).

trons that generate Raman-active hot phonons. At $x=0.28$, the excess energy of photoexcited electrons is approximately 110 meV, which is near the minimum energy required to generate the Raman-active hot LO phonons. Thus, below $x=0.28$ only those electrons that have larger energy than 110 meV as a result of thermalization can generate Raman-active hot phonons. The cascade model using only electrons from the split-off hole band is not in good agreement with our experimental results. The calculated N_q's from this model decrease faster with increasing x. Since the cascade model assumes a monoenergetic distribution of electrons, N_q will rapidly decrease with lower excess energy. On the other hand, instantaneous thermalization gives rise to a spread in the hot-electron distribution which makes it less sensitive to changing excess energies. This is especially so when the average of the hot-electron distributions are near 110 meV.

To test the validity of our conclusions, we fit our experimental data with a simple analytical model that assumes an instantaneous thermalization of electrons from the split-off band. This assumption is valid since electrons in the Γ valley thermalize in a time scale of 100 fs, which is shorter than our pulse width.[8] We consider the nonparabolicity of the Γ-valley conduction and valence bands in our model. For GaAs, the Γ-valley conduction band can be written as $\Gamma(E)=E+\alpha E^2+\beta E^3$, where $\alpha=0.577$ eV^{-1} and $\beta=0.047$ eV^{-2}.[5,25] The non-parabolic valence bands may be determined using the Kane model and are found in Ref. 5. Since we conclude that almost all of the photoexcited electrons originating from the heavy- and light-hole bands rapidly transfer to the satellite valleys before emitting any LO phonons, we consider only the split-off hole band in our calculation of N_q. From the joint density of states in GaAs at photon energies of 2.33 eV, we have determined that approximately 23% of the total photoexcited electron density ($\sim 10^{17}$ cm^{-3}) comes from the split-off hole band. Because of this relatively small electron concentration we can assume a Boltzmann distribution of hot electrons. We have used Eqs. (26) and (27) of Ref. 5 to determine the hot-phonon occupation number. Since split-off hole band electrons generate nearly all of the Γ-valley hot phonons, we only consider the effects of emission and absorption of LO phonons on the hot-electron distribution. The effects of intervalley scattering of these electrons are negligible. In our calculation we assume an optical pulse with a sech^2 profile having a full width at half maximum of 1.5 ps. We have also included the changing effective masses, energy band gaps, the relative contribution form the GaAs- and AlAs-like LO phonons, and penetration depths with increasing x.[18,19,26–28] The fit denoted as the broken curves in Fig. 1 is remarkably good, in view of the simplicity of our model. We have obtained this fit without having to include localization of LO phonons due to alloy potential fluctuations. Since localization tends to decrease the phonon generation efficiency, our results might suggest that no significant localization of LO phonons due to alloying exist. This is consistent with the experimental work of Ref. 18, however, that work only considered alloy concentrations up to $x=0.24$.

Since we have excluded the contributions to N_q from the other hole bands, our model yields $N_q=0$ at $x=0.39$ for both GaAs- and AlAs-like phonon modes. At this concentration no electrons can be excited into the conduction band from the split-off band at a photon energy of 2.33 eV. We have observed, however, a combined $\partial N_q/\partial P$ of 2% that of GaAs for the GaAs-like plus AlAs-like LO phonons. Since nearly all of the photoexcited electrons are scattered into the satellite valleys, we interpret this small efficiency as due to the electrons that emit one LO phonon before transferring to the satellite valleys. If we consider such effects as the changing band structure, and the penetration depth on x, then only 6% of the total $\partial N_q/\partial P$ in GaAs will be due to those electrons which emit one LO phonon before undergoing intravalley scattering. This small contribution from the light- and heavy-hole bands on $\partial N_q/\partial P$ suggests that the intervalley scattering to the L or X valley is much faster than the intravalley Fröhlich scattering. This is in good agreement with the recently determined ultrafast intervalley scattering time using 6-fs pulses.[12] In Ref. 12, the intervalley scattering rate in GaAs was determined to be approximately 20 fs. This was measured with lower photon energies (~ 2 eV) but at higher temperature (300 K) than ours. These two effects tend to cancel each other due to the decrease in the band gap at higher temperatures. If we use 200 fs as our intravalley scattering time, then we can expect the generation of $\approx 20/(200+20)$ LO phonons per electron from the electrons which are excited from the light- and heavy-hole bands. Our results might not directly support the ultrafast intervalley scattering rates of Ref. 12. This is because the phonon production efficiency using 200 fs of electron–LO-phonon interaction time might not be valid in this low-density regime when the spatial coherence of the photoexcited electron-hole pair is most likely broken by the intervalley scattering. This is because the Fröhlich scattering time of 200 fs is for the electron, not for the electron-hole pair. The LO-phonon emission by the electron-hole pair before they lose their coherence is not well known.

Our results demonstrate that under certain experimental conditions (photon energy, carrier density, optical pulse duration) it is necessary to include photoexcited electrons from the split-off hole bands together with intervalley scattering in the analysis of time-resolved Raman spectra. Since previous experiments have often excluded these effects, those results may need reconsideration. For instance, in the analysis of the data in Ref. 1, both intervalley scattering and the split-off hole band contributions have been ignored, but the electron-phonon interaction time in this experiment is in good agreement with the theoretically predicted value. However, the 2-ps risetime of the anti-Stokes Raman signal might reflect the time constant for the L-Γ scattering of the electrons at room temperature.[9] In a picosecond Raman-scattering experiment used to determine the intervalley scattering rates in GaAs, the analysis excluded the electrons from the split-off hole bands.[7] The intervalley scattering rates were mainly determined by the reduction in the hot-LO-phonon production efficiency as the photon energy in-

creased. This is because of the increasing intervalley scattering rates with larger carrier excess energies and populations. The pulse width in this experiment was about 4 ps, so the electrons returning to the Γ valley from the L and X valleys are an important source in generating the Γ- valley Raman-active hot phonons. Since the number and excess energy of electrons excited from the split-off hole band increases with increasing photon energy, the deformation potentials estimated in Ref. 7 are expected to be less than the real values. Since electrons in the Γ valley thermalize very quickly (within 100 fs with carrier densities as low as 10^{17} cm^{-3}), the instantaneous thermalization model is more appropriate than the cascade model for almost all excitation densities used in time-resolved Raman experiments.[8,9] This is because in most of these experiments the risetime is typically 1–2 ps when optical pulses near 1 ps are used.

In conclusion, we have investigated the hot-LO-phonon generation in GaAs and $Al_xGa_{1-x}As$ alloys using picosecond one-beam–excite-and-probe Raman scattering. Using 1.5-ps optical pulses at an average photon energy of 2.33 eV our results indicate that the initial generation of Raman-active hot LO phonons in GaAs and $Al_xGa_{1-x}As$ comes from photoexcited electrons which originated from the split-off hole band. Because of the ultrafast intervalley scattering times in these materials, the initial generation of hot LO phonons by electrons originating from the light- and heavy-hole bands are shown to be very inefficient. Therefore, to accurately determine the intravalley electron–LO-phonon scattering times, it is necessary to consider the electrons originating from the split-off hole band. This is especially so when the photon energy is sufficiently high such that photoexcited electrons are able to undergo intervalley scattering. We have obtained a good agreement between our experimental results and a model based upon the instantaneous thermalization of electrons excited from the split-off hole band. Our experimental results are in good agreement with recent results using femtosecond lasers.[8,12]

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