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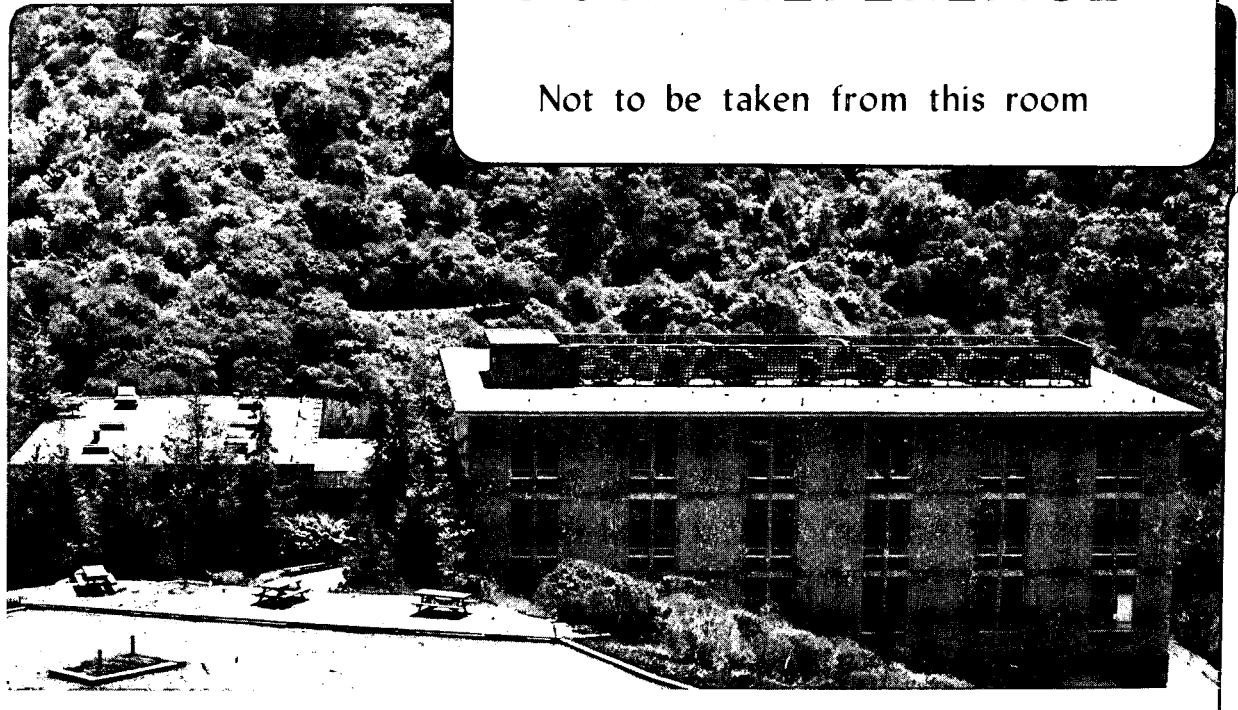

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Generation of Nonequilibrium Optical Phonons in Bulk and Microstructure Fabricated from GaAs

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ABSTRACT

Back in the 1970's Leite and coworkers demonstrated that non-thermal equilibrium distributions of optical phonons (or hot phonons) can be generated optically and studied easily by Raman scattering. Since then this technique has become an important method for studying the properties of phonons and of electron-phonon interactions. Some of these results in bulk and quantum wells of GaAs are reviewed.

1. INTRODUCTION

In 1973 a paper appeared in Solid State Communication entitled: "Photoexcited Hot LO Phonons in GaAs". The authors were J. Shah, R.C.C. Leite and J.F. Scott.[1] This paper demonstrated for the first time that a non-thermal equilibrium distribution of longitudinal optical (LO) phonons can be generated in GaAs by focussing a cw Ar ion laser onto a bulk GaAs sample. The occupation numbers of both the transverse optical (TO) and LO phonons were measured from the ratio of the Anti-Stokes (AS) to Stokes (S) Raman intensities. As the laser power density on the sample was increased, the TO phonon occupation number was found to increase only slightly while the LO phonon occupation number increased by a factor of 3! The authors explained this difference in behavior between the LO and TO phonons by the strong interaction between electrons and LO phonons via the Fröhlich interaction. The processes via which the laser

excites the non-thermal LO phonons (or abbreviated as hot phonons by these three authors) are as follows. The incident photons excite electrons from the valence band into the conduction band. Since the photon energy is much larger than the band gap of GaAs the photo-excited electrons have kinetic energies much larger than k_bT (where k_b is the Boltzmann's constant and T is the lattice temperature). These energetic electrons (also known as hot electrons) relax by emission of LO phonons. The time to emit one LO phonons is about 0.2 picosecond (ps).[2] If the LO phonon lifetime were much longer than 0.2 ps, a non-thermal population of LO phonon can be excited even by a cw laser. Knowing the phonon generation rate, the LO phonon lifetime can be determined from the hot phonon population. The LO phonon lifetime of GaAs at room temperature was determined by Shah, Leite and Scott[1] to be 5 ps.

In subsequent papers, Leite and coworkers[3,4] demonstrated that non-thermal distribution of hot phonons could be excited optically in many other semiconductors. In general, the more ionic semiconductors have stronger Fröhlich interaction and hence larger hot phonon populations. This pioneering work of Leite and coworkers has demonstrated a new method for studying the properties of phonons and of interaction between electrons and phonons. The beauty of this technique is its simplicity. The same laser used to excite the hot phonons is used to determine the hot phonon population by Raman scattering. The technique can be easily extended to become a time-resolved experiment by using two short pulses. For example, phonon lifetimes can be determined directly by exciting a non-thermal population with a pump pulse and probing the decay of the hot phonon population with a suitably delayed probe pulse.[5] The fact that Raman scattering is an instantaneous process implies that the time resolution of this technique is limited only by the length of the laser pulses. In this article we will discuss some of the applications of photo-excited hot LO phonons in GaAs to study electron-phonon interaction. We will also discuss the limitations of this technique in studying electron-phonon interaction in

semiconductor quantum wells and propose methods to overcome these limitations.

2. NON-EQUILIBRIUM PHONON SPECTROSCOPY

In 1983 Collins and Yu[6] proposed to use the efficiency of photons in exciting hot phonons as a means to probe electronic band structure and intervalley scattering in semiconductors. Their idea was simply that the efficiency of a photon in producing hot phonons depended on the photon energy and the band structure. The amount by which the photon energy exceeds the band gap of the semiconductor determines the kinetic energy of the hot electrons. The number of hot phonons which will be emitted by a hot electron is given by its kinetic energy divided by the phonon energy E_{LO}. For hot electrons with a given kinetic energy, the distribution in momentum space of hot phonons emitted by these hot electrons depends on the band structure. Typically this phonon distribution has a minimum and a maximum value determined by the band dispersion.[7] On the other hand, the range of wave vectors of hot phonons probed by Raman scattering in a particular scattering geometry is well defined. Thus the variation of the hot phonon population with the excitation photon energy can be used to probe the band dispersion. In semiconductors like GaAs, besides the conduction minimum at the Brillouin zone center (Γ) there are higher conduction band minima at the L and X points of the Brillouin zone. When a hot electron has kinetic energy higher than these conduction minima, it can relax into these higher minima by *inter*valley scattering (IVS). If these IVS rates are faster than the *intra*valley electron-LO phonon scattering rate, the electrons are removed from the Γ valley to the zone-edge valleys. Since the zone-center hot electrons are responsible for generating hot LO phonons we expect a drop in the hot phonon generation efficiency when the photon energy becomes larger than the onset energies for IVS. Thus the location of the higher conduction minima and the IVS rates can be deduced from such decreases in the hot LO

phonon generation efficiency in GaAs. Since this technique involves measuring the hot phonon generation efficiency as a function of photon energy, Collins and Yu[6] has labeled this technique: hot phonon spectroscopy.

To apply hot phonon spectroscopy to GaAs, Collins and Yu[6] used tunable picosecond laser pulses from a synchronously pumped dye laser to excite a piece of bulk GaAs. The laser pulse length was 4 ps and comparable to the hot LO phonon lifetime while the separation between the laser pulses was about 12 ns. Compared to the continuous wave Ar ion laser used by Shah et al. higher population of hot LO phonons could be generated without heating the sample. The hot LO phonon occupation number measured by Collins and Yu[7] at a constant electron density of 4.8×10^{18} cm^{-3} are shown in Fig. 1 as the solid circles. The experimental results are compared with two different

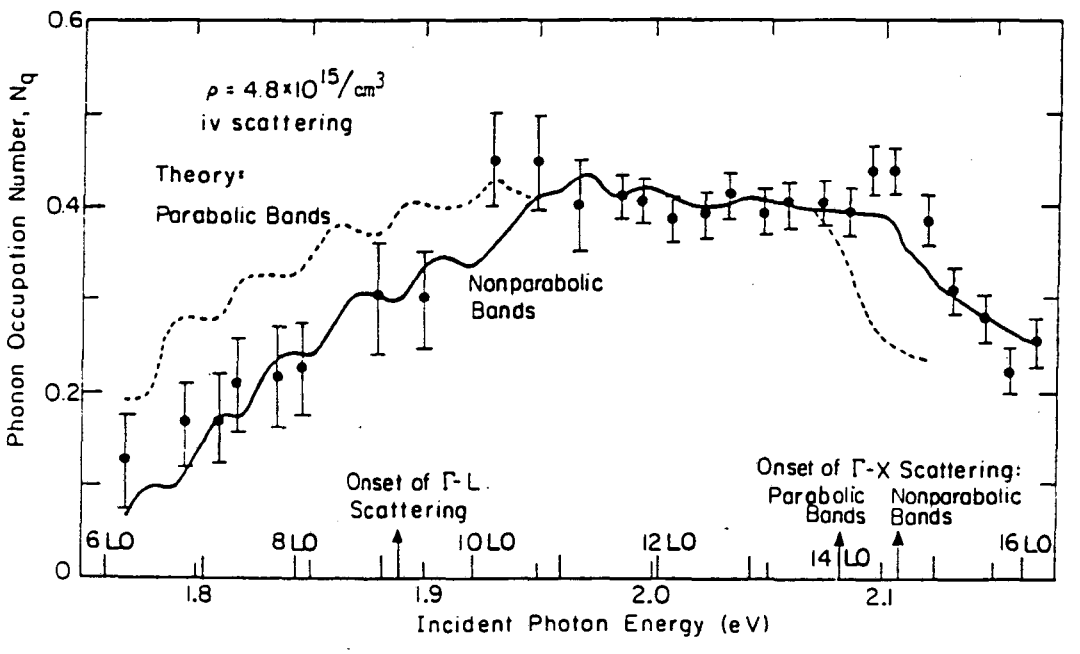

Fig. 1 Hot LO phonon occupation number excited in bulk GaAs excited by picosecond laser pulses. The solid circles are experimental points. The solid and dashed curves are calculated from two different band models. Reproduced from Ref. [7].

model calculations. In one model the conduction band of GaAs was assumed to be parabolic (dashed curve in Fig. 1). In the second model (shown as the solid curve in Fig. 1) the conduction band was assumed to be nonparabolic with dispersion given by:

$$\hbar^2k^2/2m=E+\alpha E^2+\beta E^3, \quad (1)$$

where k and E were the electron wave vector and energy respectively. Obviously the second model agrees much better with the experimental results. In particular the energy for the onset of IVS from Γ to X is quite sensitive to the conduction band nonparabolicity. From the magnitude of the decrease in the LO phonon population due to IVS to the X conduction minima, Collins and Yu[7] deduced the Γ-X IVS deformation potential ($D_{\Gamma-X}$) to be 10^9 eV/cm. Collins and Yu did not observe any significant decrease in the hot phonon population at the onset of Γ-L IVS. This led them to conclude that the Γ-L IVS deformation potential ($D_{\Gamma-L}$) is less than 1.5×10^{18} eV/cm. More recently, $D_{\Gamma-L}$ has been determined by several independent methods[8-10] and the measured values are all $>4\times10^8$ eV/cm. This discrepancy between the value of $D_{\Gamma-L}$ estimated by Collins and Yu[7] and more recent results can be explained by the fact that there are two possible interpretations of the absence of a drop in LO phonon population at the onset of Γ-L IVS. One explanation is that the deformation potential is small so IVS does not remove a significant number of hot electrons from the Γ valley and hence the LO phonon generation rate is not changed. The other possibility is that $D_{\Gamma-L}$ is large so that many electrons are removed in less than 1 ps from the Γ valley by IVS. However, the electrons in the L valley have no where to relax other than to return to the Γ valley. If these electrons return to the Γ valley within the duration of the laser pulse of 4 ps, the number of hot LO phonons emitted by the Γ electrons is unchanged by IVS to the L valley. Recently it has been shown that this return of hot electrons from the L valley to the Γ valley occurs in about 7 ps[8] and therefore explains the absence of

a decrease in the hot LO phonon generation rate at the onset of the Γ-L IVS. The situation is different for IVS to the X valleys. In this case hot electrons scattered into the X valleys do not return to the Γ valley directly. Instead they are scattered into the L valley. The X-L scattering rate is larger than the X-Γ scattering rate because the L valley density-of-states is higher. As a result, a decrease in the hot phonon generation rate is observed at the onset of Γ-X IVS. The relaxation paths for electrons in the X and L conduction valleys are shown schematically in Fig. 2.

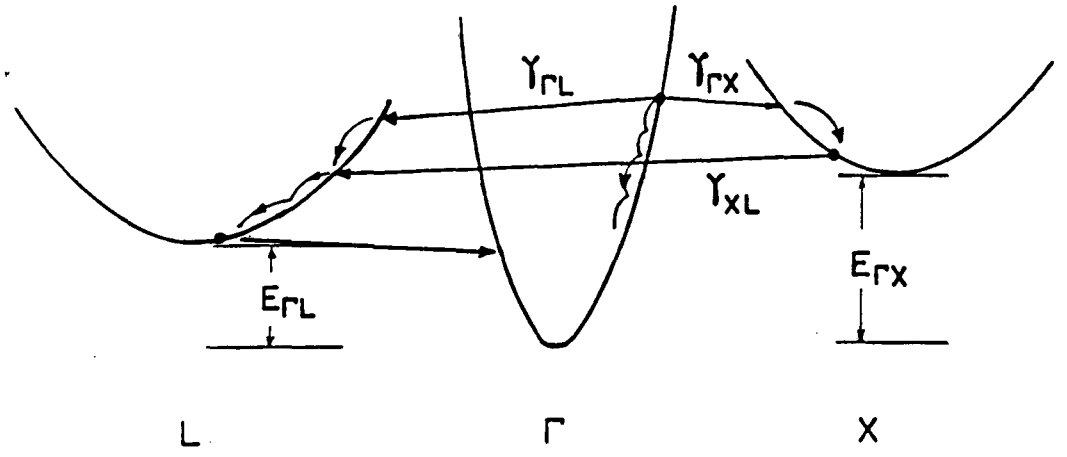

Fig. 2 Interband and Intraband scattering of hot electrons by phonons.

3. THE 'HOT PHONON EFFECT' AND PHONON TEMPERATURE OVERSHOOT

It has assumed that a photoexcited hot electron gas in GaAs cools predominantly by emission of LO phonons provided the electron temperature is high enough to emit LO phonons. Measurement of hot electron cooling curves in GaAs using picosecond laser pulses were first reported in 1979.[11,12] However, the experimental cooling curves agree well with theory only for low excitation densities. For photo-excited electron density higher than 10^{17} cm^{-3} it was found

that the cooling rate was significantly reduced.[13] Two explanations have been proposed to explain this density dependence of hot electron cooling rate. One explanation is based on screening of the Fröhlich interaction by the high density of electron-hole pairs.[13] The second explanation is based on the generation of a high non-thermal population of LO phonons (this is now known as the 'hot phonon effect').[14] Much theoretical and experimental work has since been devoted to understand the effects of screening and of hot phonons on the cooling of hot electrons in GaAs at high electron densities.[15] It is now generally believed that screening is not important at densities below 10^{19} cm^{-3}. However, the evidence in support of the hot phonon explanation has mostly been indirect.

We have applied Raman scattering to investigate the hot phonon effect in reducing the hot electron cooling rate in bulk GaAs under high excitation conditions. The idea is an extension of the experiment of Shah, Leite and Scott.[1] Using a single beam of picosecond or subpicosecond laser pulses, one can excite hot electrons and hot phonons. The photo-excited hot electron will scatter light inelastically via single-particle excitations (SPE).[16] The electron temperature can be determined directly from the ratio of AS to S Raman intensities of the SPE spectra:

$$I_{AS}/I_S = \exp(\hbar\omega/k_b T_e) \qquad (2)$$

where I_{AS} and I_S are , respectively, the AS and S Raman intensities of the SPE spectra at frequency ω, and T_e is the electron temperature. In discussing the 'hot phonon effect', it is convenient to define a 'phonon temperature' T_{LO} in terms of the phonon population N_{LO} via the equation:

$$N_{LO} = [\exp(E_{LO}/k_b T_{LO}) - 1]^{-1} \qquad (3)$$

The advantage of the Raman technique is that both the electron and phonon temperatures can be determined in the same experiment under

identical conditions. If the 'hot phonon effect' explanation were correct, we would expect that at low excitation intensities, no hot LO phonon would be observed. As the excitation density is increased we expect that the phonon temperature will increase while the hot electron temperature will also be increased due to the reduced cooling rate. At sufficiently high excitation level, the electrons and phonons should reach thermal equilibrium and have the same temperature.

In the experiment performed by Huang and Yu[17] on GaAs using laser pulses of 4 ps duration, it was found that at photoexcited electron densities of 10^{18} cm^{-3} the electron and phonon temperatures were both equal to about 800 K. This suggested that large amount of hot phonons were indeed generated by the relaxation of the dense, hot electron gas. Furthermore, the hot electrons and hot phonons reached thermal equilibrium within 4 ps, the duration of the laser pulses. In a more recent experiment, Kim and Yu[18] used subpicosecond laser pulses generated by a colliding-pulse modelocked (CPM) dye laser to excite and probe hot electrons and hot phonons in GaAs. In this case they found that the LO population in GaAs increased with photo-excited electron density as expected. However, at electron densities above 10^{18} cm^{-3} they found that the phonon temperature became <u>higher</u> than the electron temperature as shown in Fig. 3.

Both the low electron temperature and the "overshoot" of the electron temperature by the phonon temperature observed with subpicosecond laser pulses cannot be explained by a simple model in which hot electrons cool by emission of LO phonons only. The results calculated from this simple model are shown as the broke curves in Fig. 3. The reason for the failure of this model in explaining the experimental results is that, within the subpicosecond duration of the laser pulses, the hot electrons have time to emit one or two LO phonons only. Thus LO phonon emission cannot explain the rapid cooling of the hot electrons from the 2500 K temperature at excitation to the observed temperature of about 600 K in fraction of a ps. On the other hand, the low electron temperature can be

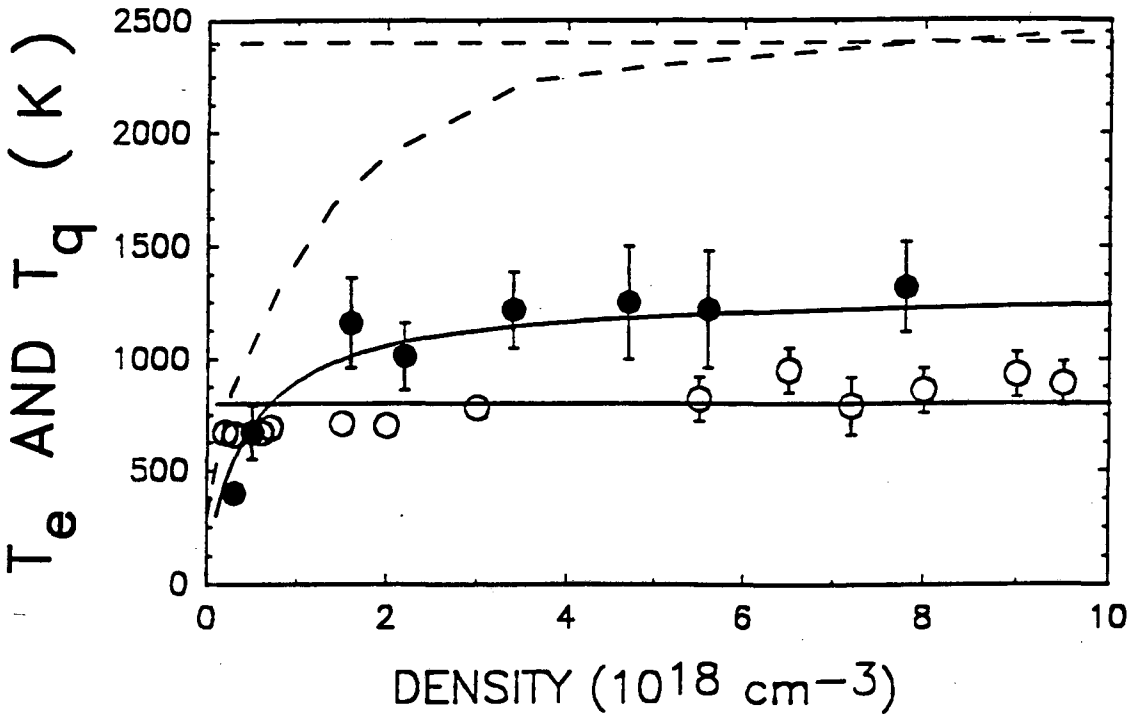

Fig. 3 Hot electrons (open circles) and hot phonon temperatures (closed circles) in GaAs excited by subpicosecond laser pulses of 0.6 ps duration plotted as a function of the photoexcited electron densities. The broken curves are the electron and phonon temperatures calculated with a conduction band model neglecting IVS. The solid curves are calculated with a model including IVS. The deformation potential $D_{\Gamma\text{-}L}$ was adjusted to 7×10^8 eV/cm in order to fit the data points. Reproduced from Ref. [18].

explained by IVS which removes the high energy electrons in about 0.1 ps. By including IVS in the model the experimental results could be explained quantitatively as shown by the solid curves in Fig. 3. Thus our result shows that: *for high electron densities and for time scales shorter than a ps hot electrons in GaAs cool predominantly by scattering into the higher conduction band valleys at X and L.* The high LO phonon temperature is explained by the small phonon

density-of-states near zone-center. Thus the so-called 'hot-phonon effect' is effective in reducing the hot electron cooling rate only for times longer than a picosecond and involves the larger wave vector LO phonons. The value of the deformation potential $D_{\Gamma-L}$ in GaAs we deduced from fitting the experimental points in Fig. 3 was 7×10^8 eV/cm. This value of $D_{\Gamma-L}$ is in good agreement with the values determined by other techniques.[8-10]

4. NONEQUILIBRIUM PHONON GENERATION IN QUANTUM WELLS

There has been much interest in the properties of quasi-two-dimensional microstructures fabricated from GaAs and AlAs. These structures are usually referred to as quantum wells (QW) or multiple quantum wells (MQW) because electrons and holes are confined inside the GaAs layers (whose band gap is smaller than that of AlAs). What is less well known is that the optical phonons are also confined in these QW.[19,20] The phonons are confined because the optical phonon frequencies in GaAs and in AlAs do not overlap. As a result optical phonons cannot propagate from one layer to the next. The confinement of electrons and phonons in QW has significant influence on the electron-phonon interaction and hence on hot electron relaxation in QW.

Figure 4 shows schematically a symmetric MWQ where both GaAs and AlAs layer thicknesses are given by L. The z-axis is taken to be perpendicular to the layers. Let q denote the GaAs LO phonon wave vector in the QW. Due to confinement of the LO phonon inside the GaAs layer, the q_z component is

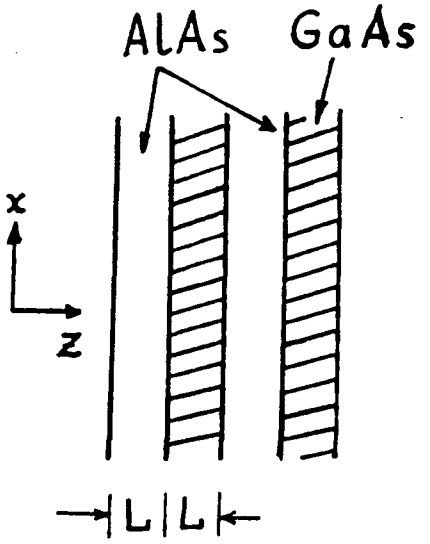

Fig. 4 Schematic diagram of a symmetric GaAs/AlAs MQW.

quantized into: $q_z = m\pi/L$ where m is an integer. The Fröhlich interaction matrix element $|F|^2$ is proportional to $1/q^2$ so the immediate effect of quantization of the LO phonon is to make the Fröhlich interaction dependent on the QW thicknesses:[21]

$$|F|^2 \approx [q_x^2 + q_y^2 + (m\pi/L)^2]^{-1} \quad (4)$$

As we have mentioned earlier, the LO phonons emitted during hot electron relaxation has a well defined range of wave vectors determined by the electron dispersion. In case of QW, only the components of the wave vector <u>parallel</u> to the layers (q_x and q_y) are conserved in electron-phonon scattering. For GaAs, q_x and q_y typically have values between 3×10^5 to 2×10^6 cm^{-1}. Thus for QW layer thicknesses of 10 nm or less, the q_z component becomes much larger than the component parallel to the layers. As a result $|F|^2$ decreases with L as $(m\pi/L)^{-2}$. In addition to its dependence on L^2, $|F|^2$ also depends on the symmetries of the confined electron and phonon envelope wave functions. For example the wave function of the electron subbands are either symmetric or anti-symmetric with respect to reflection about the center of the layer. Similarly the confined LO phonon modes produce electric potentials which have definite parity under this reflection. These symmetries determine which quantized phonon modes can mediate an *inter*subband scattering and which modes can mediate an *intra*subband scattering.

We have studied the dependence of $|F|^2$ on quantum well thicknesses by measuring the efficiency of hot phonon generation by photoexcited hot electrons in a series of GaAs/AlAs MQW with varying thicknesses.[21] Our results, shown in Fig. 5, confirmed the prediction that, as L decreases, the photo-excited hot electrons become less efficient in emitting hot LO phonons. We have also found an additional effect which decreased the amount of hot phonons observable by Raman scattering in QW. Due to confinement, wave vector conservation in Raman scattering holds only for the component of wave vectors parallel to the layers. In a typical backscattering geometry where the incident and scattered radiations are both propagating almost perpendicular to the layers, the component of wave

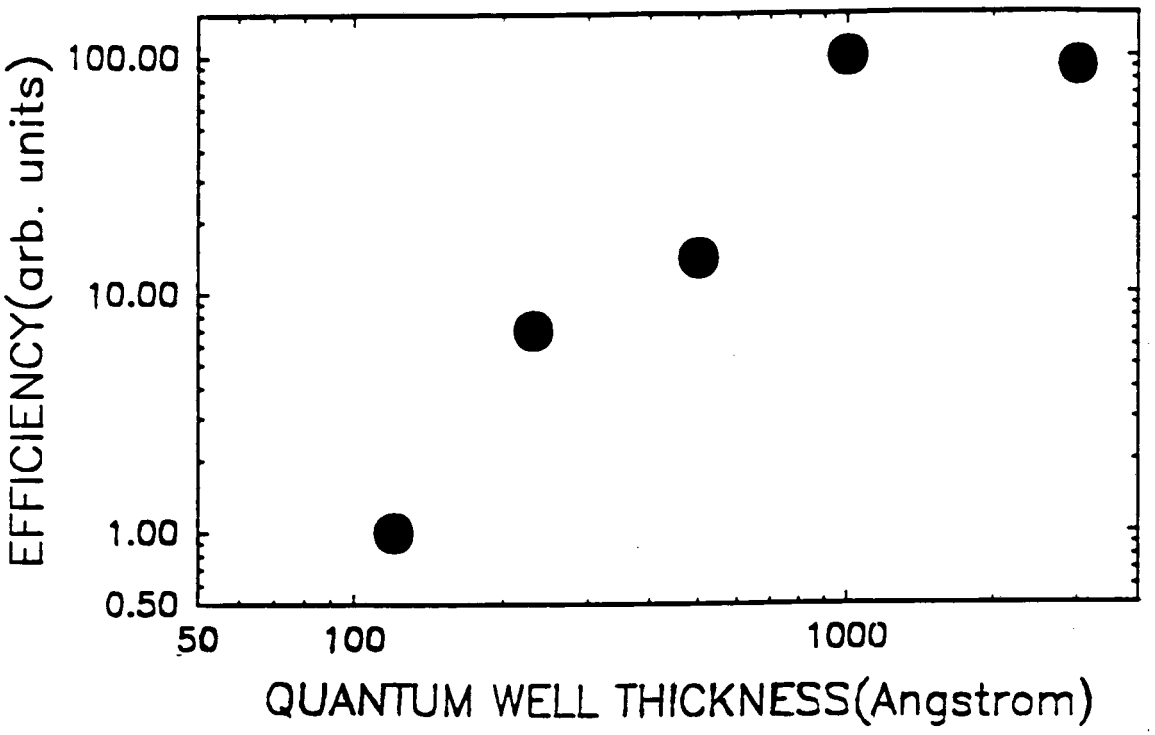

Fig.5 Efficiency of hot electrons excited by subpicosecond laser pulses in generating hot phonons in GaAs/AlAs MQW as a function of well thickness. The electron densities have been kept below 10^{18} cm^{-3} in the measurements to avoid saturation effects.

vector in the xy-plane of the Raman phonon is very small (around $0.5x10^5$ cm^{-1} in our experiment). On the other hand the hot phonons emitted by inter- and intra-subband hot electron relaxation have typically wave vectors larger than $3x10^5$ cm^{-1} in the xy-plane. These considerations imply that hot phonons are more difficult to observed in QW and hence for studying electron-phonon interaction in QW.

To overcome these limitations we suggest the following solutions. The distribution of phonon wave vectors generated by *inter*subband relaxation of hot electrons depends very much on the separation (Δ) between the two subbands involved. This situation is clear from examination of Fig. 6. When the two subband separation is close to the phonon energy $\hbar\omega$, the phonons emitted during

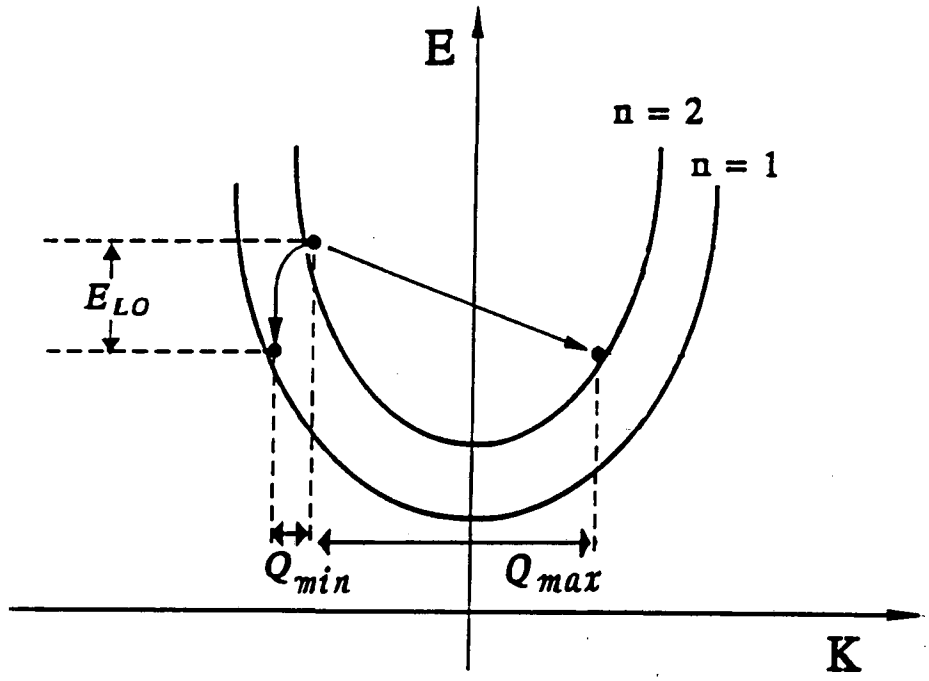

Fig. 6 A schematic diagram showing how energy and momentum conservation in the xy-plane during intersubband electron-phonon scattering determines the minimum and maximum wave vectors of phonon emitted.

intersubband scattering have very small wave vectors. Thus one expects a 'resonance' in the hot phonon generation efficiency when the subband separation is 'tuned' to be equal to the phonon energy. One way to achieve this resonance is to change the QW layer thickness. To estimate the QW layer thickness for this resonance to occur, we have calculated the electronic subband energies in GaAs/AlAs QW using an effective-mass model. The electrons are assumed to have effective mass of 0.67 times the free electron mass while the potential well has a depth of 1.1 eV. Within this simple model, the separation between the two lowest subbands becomes equal to the LO phonon energy (36 meV) when the well thickness L is around 20 nm. To estimate the width of this resonance, we assume that hot electrons obey Boltzmann statistics and are in quasi-equilibrium with a

temperature of 1500 K. The phonon observable in the experimental Raman scattering has a wave vector of 0.5×10^5 cm^{-1} in the xy-plane. The calculated full-width-at-half-maxima of this 'resonance" is only about 1.4 nm.[22] In order to see this resonance the QW thickness has to be controlled to within one monolayer. This is achievable nowadays with growth techniques such as Molecular Beam Epitaxy.

Another way to overcome the limitation of Raman scattering in observing hot phonon in QW is to grow the MQW on a grating fabricated on the substrate. If the grating period is D, wave vector conservation in the xy-plane during electron-phonon scattering will be relaxed by $2\pi/D$. By varying D it should be possible to determine the distribution of hot phonons emitted by hot electrons in QW.

5. CONCLUSIONS

After Leite and coworkers have demonstrated that non-thermal distribution of optical phonons can be generated and detected readily in bulk semiconductors, much progress has been made in applying these hot phonons to study electron-phonon interactions and hot electron relaxation in semiconductors such as GaAs. While it is desirable to extend the technique to study hot phonons in QW, there are difficulties which need to be overcome. We have proposed two ways to overcome these difficulties and we plan to test these ideas experimentally. If these ideas turn out to be feasible, we expect that hot phonons will play an important role in the study of electron-phonon interactions in QW also.

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REFERENCES

[1] Shah, J., Leite, R.C.C. and Scott, J.F., Solid State Commun. 8, 1089 (1970).

[2] Conwell, E.M., and Vassell, M.O., IEEE Trans. on Elect. Devices, ED-13, 22 (1966).

[3] Mattos, J.C.V., and Leite, R.C.C., Solid State Commun. 12, 465 (1973).

[4] Lluesma, E.G., Mendes, G.,Arguello, C.A., and Leite, R.C.C., Solid State Commun. 14, 1195 (1974).

[5] Von der Linde, D., Kuhl, J. and Klingenberg, H., Phys. Rev. Lett. 44, 1505 (1980).

[6] Collins, C. and Yu, P.Y., Phys. Rev. B27, 2602 (1983).

[7] Collins, C. and Yu, P.Y., Phys. Rev. B30, 4501 (1984).

[8] Shah, J., Pinczuk, A, Gossard, A.C., and Wiegmann, W., Phys. Rev. Lett. 54, 2045 (1985).

[9] Ulbrich, R.G., Kash, J.A., and Tsang, J.C., Phys. Rev. Lett. 62, 949 (1989).

[10] Fasol, G., Hackenberg, W., Hughes, H.P., Ploog, K., Bauser, E., and Kano, H., Phys. Rev. B41, 1461 (1990).

[11] Shank, C.V., Fork, R.L., Leheny, R.F., and Shah J., Phys. Rev. Lett. 42, 112 (1979).

[12] Von der Linde, D., and Lambrich, R., Phys. Rev. Lett., 42, 1090 (1979).

[13] Leheny, R.F., Shah, J., Fork, R.L., Shank, C.V., and Migus, A., Solid State Commun. 31, 809 (1979).

[14] Van Driel, H.M., Phys. Rev. B19, 5928 (1979).

[15] Shah, J., Pinczuk, A., Gossard, A.C., and Wiegmann, W., Phys. Rev. Lett. 54, 2045 (1985); Pötz, W., and Kocevar, P., Phys. Rev. B28, 7040 (1983); Ruhle, W.W., Leo, K., and Bauser, E., Phys. Rev. B40, 1756 (1989).

[16] Klein, M.V., in Light Scattering in Solids ,Topics in Applied Physics, Vol. 8, ed. by M. Cardona (Springer-Verlag,New York, 1975) p.148.

[17] Huang, Y.H. and Yu, P.Y., Solid State Commun. 63, 109 (1987).

[18] Kim, D.S. and Yu, P.Y., Phys. Rev. Lett. 64, 946 (1990); Phys. Rev. B43 (in press).

[19] Klein, M.V., IEEE Quantum Electron. QE-22, 1760 (1990).

[20] Sood, A.K., Menendez, J., Cardona, C., and Ploog, K., Phys. Rev. Lett. 54, 2111 (1985).

[21] Kim, D.S. and Yu, P.Y., in **Ultrafast Laser Probe Phenomena in Bulk and Microstructure Semiconductors III**, ed. by Robert R. Alfano (SPIE Proceeding Series, Vol. 1282, 1990) p.39.

[22] Details of these calculations will be published elsewhere.

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