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SUBPICOSECOND RAMAN STUDY OF HOT ELECTRONS AND HOT PHONONS

IN GaAs

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INTRODUCTION

There has been much interest in the study of fast relaxations of hot electrons and hot phonons in GaAs and related family of semiconductors because of their importance to the operation of high speed devices. Optical techniques based on picosecond (ps) and femtosecond (fs) lasers have played an important role in these studies.[1-4] In particular optical techniques such as time-resolved photo-induced absorption[1,3] and luminescence[2] have been used to study the relaxation and cooling of the hot electrons. The disadvantages of these optical techniques are that they do not provide information on the phonons which are excited by the cooling electrons. For example, Leheny et al.[5] have found that the cooling rate of photoexcited hot electrons in bulk GaAs decreased when the electron density was increased to above 10^{17} cm^{-3}. Leheny et al.[5] suggested that this effect can be explained by the screening of the Frohlich electron-longitudinal optical (LO) phonon interaction. Subsequent work has shown that the observed carrier density was too low for screening effects to be significant. Instead, an alternate explanation known as the "hot phonon effect", has been proposed.[6-8] In this model the hot electrons excite a large non-equilibrium population of LO phonons during relaxation. When the hot electrons and the hot LO phonons reach thermal equilibrium, the LO phonons are not effective in cooling the electrons. Raman scattering has the advantage of being able to determine both the electron and the phonon populations. Using picosecond laser pulses, Huang and Yu[9] have determined the temperature of photoexcited hot electrons and hot phonons in GaAs using Raman scattering. Their results indicate that the electrons and LO phonons are in thermal equilibrium within a time shorter than their pulse length of about 4.5 ps. Recently, we have used Raman scattering with subpicosecond laser pulses to measure the electron and phonon temperatures in GaAs as a function of electron density.[10] For densities above 10^{18} cm^{-3} we found that the phonon temperature actually overshot the electron temperature. Our results have been explained by including scattering of electrons from the zone-center valley to the higher conduction band valleys at the L and X points of the Brillouin zone. In this paper we will describe in detail the use of Raman scattering with subpicosecond laser pulses for investigating both the Frohlich interactions between

electrons and LO phonons and the intervalley electron-phonon interactions.

EXPERIMENTAL DETAILS

 The source of subpicosecond laser pulses used in our experiment is a colliding-pulse modelocked (CPM) dye laser pumped by a cw Ar ion laser. The construction of the CPM laser has been described by Valdmanis et al.[11] Without any tuning element inside the cavity this laser produces Fourier-transform-limited pulses of typically less than 100 fs duration. Unfortunately the corresponding spectral width of the laser is too large for observing the LO phonon in GaAs. We used a one-plate birefringent filter inside the cavity to narrow down the laser linewidth. To resolve the transverse optical (TO) and LO phonons in GaAs the linewidth of the laser was narrowed to 25 cm^{-1} while the full-width-at-half-maximum (FWHM) of the laser pulse length is increased to 600 fs. Using birefringent plates of different thickness the FWHM of the CPM laser can be changed discretely between 200 fs and 600 fs. The photon energy of the CPM laser is about 2.0 eV and the energy per pulse is typically 0.3 nJ. With a repetition rate of 120 MHz the average power of the laser is over 20 mW. The dye laser is passed through a set of Brewster angle prisms to reduce the dye fluorescence before being focussed on the sample.

 The sample studied was a 0.3 μ thick intrinsic GaAs layer grown by molecular beam epitaxy on a [001] oriented GaAs substrate. The GaAs layer was sandwiched between two AlAs layers. The AlAs layers act as barriers to prevent the photoexcited electrons from diffusing away from the GaAs layer. The top AlAs layer was protected by an 80 A GaAs cap layer. The sample was maintained either at room temperature or around liquid nitrogen temperature during the experiment. The density of the photoexcited electron was varied by changing the size of the laser focus on the sample from about 100 μ to about 10 μ. The photoexcited carrier density was
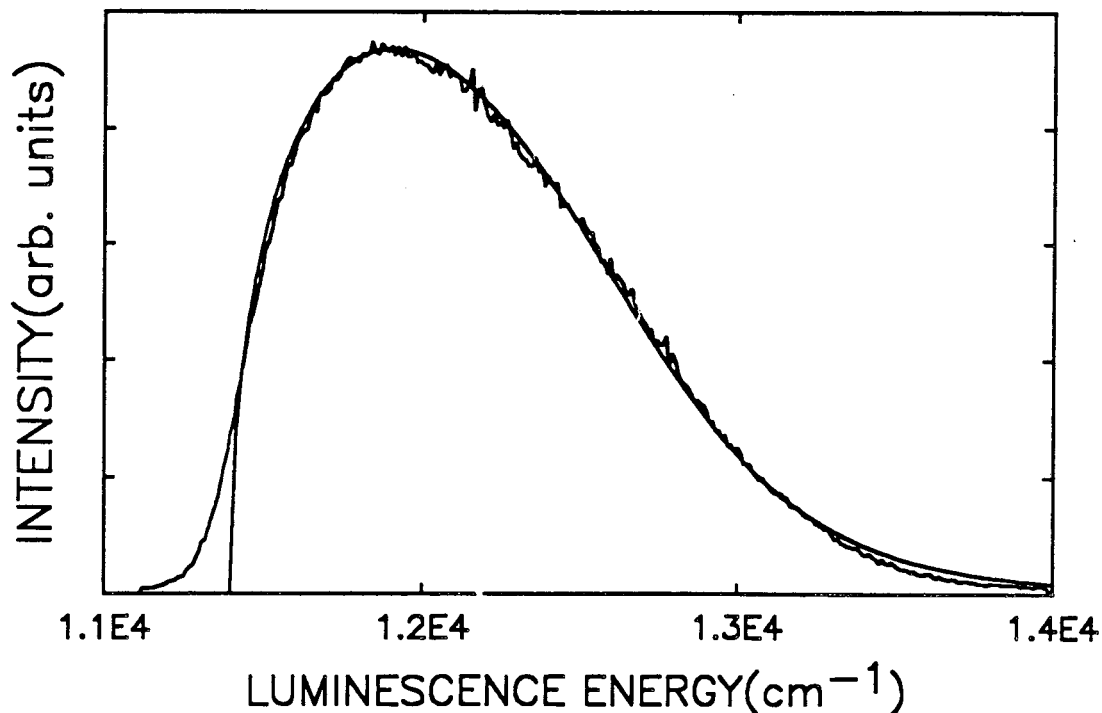

Fig. 1 The time-integrated photoluminescence spectrum of GaAs excited by laser pulses of 600 fs. The smooth curve is a theoretical luminescence curve for a degenerate electron gas with a quasi-Fermi energy of 155 meV or electron density of 5×10^{18} cm^{-3} and electron temperature of 400 K.

determined by analyzing the time-integrated photoluminescence spectra.
Figure 1 shows a typical time-integrated luminescence spectra. Laser
heating of the sample was determined by measuring the luminescence and
Raman spectra with a cw dye laser of identical time-averaged power density
and photon energy.[12] It was found that laser heating of the sample was
minimal. The light scattering experiment was performed in a
backscattering geometry. Several combinations of polarization geometries
with the incident and scattered radiations polarized along both the [1I0]
and [100] directions have been used. In all cases the observed scattering
spectra are consistent with the spectra reported in the literature using
cw lasers. The results presented in this paper were obtained with both
the incident and scattered light polarized along the [1I0] directions.
In this configuration scattering by the LO phonon, the plasmon and by
single particle excitations (SPE) of the photoexcited electron-hole
plasmas are all allowed.

EXPERIMENTAL RESULTS

        A typical scattered spectrum obtained on a sample at around 77 K is
shown in Fig. 2. This scattered spectra can be decomposed into three
parts: (a) a broad background due to luminescence extending all the way to
the bandgap of GaAs around 1.5 eV; (b) relatively sharp peaks caused by
scattering from the coupled LO-plasmon modes on both side of the laser
line; and (c) a broader peak centered on the laser line caused by
scattering from SPE. All three features change in their characteristic
ways when the electron density is increased by decreasing the laser focus.
At low electron density the photoluminescence is mainly centered around
the band gap and its intensity is negligible near the laser line. The
temperature of the photoexcited electron can be determined from its
lineshape. At higher densities the electron gas becomes degenerate and
hotter so the luminescence spectra starts to extend to higher photon
energies. Both the electron quasi-Fermi energy and temperature can be
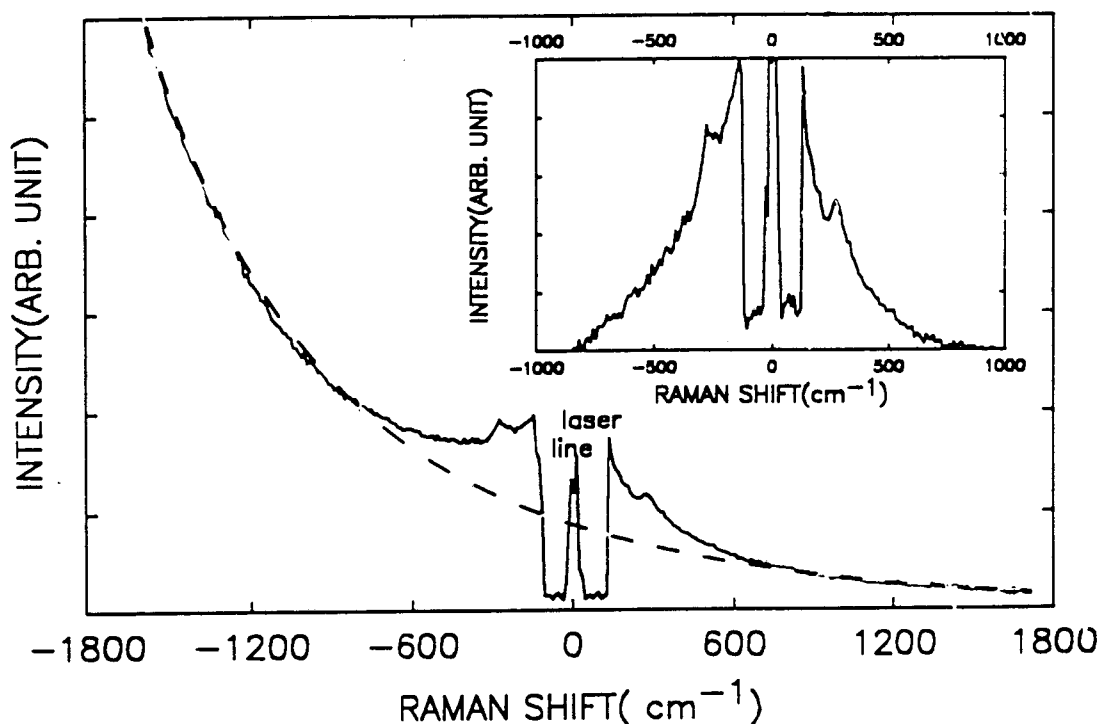

Fig. 2 Typical scattering spectrum of GaAs superimposed on the hot
luminescence background (broken line). The inset shows the Raman spectra
after subtraction of the hot luminescence background. In both spectra note
the change in scale in the vicinity of the laser line.

deduced from the luminescence spectra as shown in Fig. 1. At low density
the LO phonon peaks dominate the Raman spectra. As the electron density
increases, the phonon peak position shifts towards lower frequencies as
shown in Fig. 3. The SPE spectrum is absent in the Raman spectra at low
excitation intensities. It grows quadratically with laser intensity since
it is produced by scattering of light from the photoexcited electrons. At
high intensities the SPE spectra dominate over phonon scattering.

The red shift of the LO phonon frequency in Fig. 3 can be explained
by the coupling between the plasmon and the LO phonon.[13] Normally this
coupling produces two modes. The lower frequency mode approaches the TO
phonon frequency as the electron density is increased. The higher
frequency mode starts with the LO frequency at low electron density and
approaches the plasmon frequency at high densities. The photoexcited
electrons, however, have a spatial density profile determined by the laser
beam profile. As shown by Collins and Yu,[14] if the electrons have a
Gaussian profile, the coupled modes will show up as two peaks in the Raman
spectra; one at the LO phonon frequency and the other at the TO phonon
frequency. The peak at the LO phonon frequency results from scattering
from unscreened LO phonons in the low density ($<10^{17}$ cm^{-3}) region. The
peak at the TO phonon frequency results from the LO phonon screened by the
free electrons in the high density region ($>10^{18}$ cm^{-3}). As the light
intensity is increased, the high density region grows relative to the low
density region so the relative intensity of the two peaks can be used to
determine the peak electron density. Because of the larger laser width
associated with our subpicosecond pulses, we cannot resolve the two peaks
at the TO and LO frequencies. Instead we observe a continuous shift of
the phonon peak as shown in Fig. 3. Thus the position of the phonon peak
is another way to estimate the photoexcited electron density in the high
density region. The densities obtained from both the luminescence and
Raman spectra usually agree well with the densities calculated from the
number of photons absorbed by the sample. The dependence of the coupled
mode frequency on density also allows us to determine the phonon
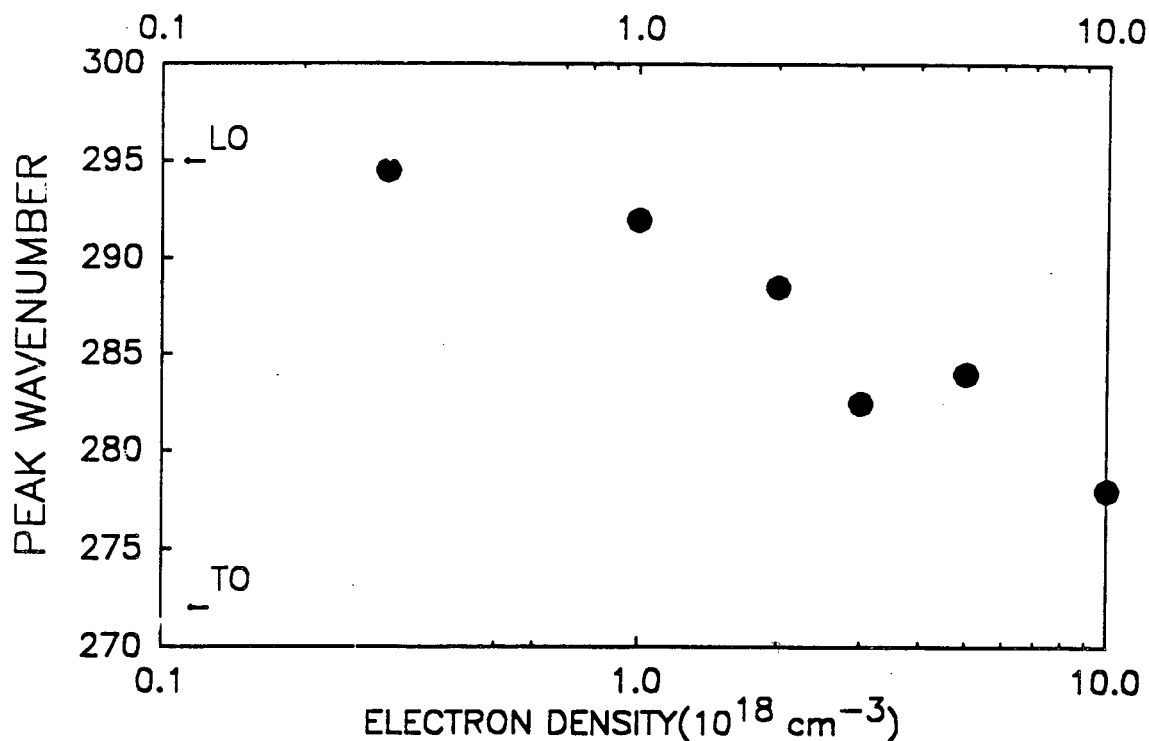

Fig. 3 Dependence of the coupled-LO-phonon-plasmon mode on photoexcited
electron densities. The laser pulse FWHM is 600 fs.

temperature in both the low and high density regions.

We have deduced both the phonon occupation number N_q and electron temperature T_e from the ratios of the anti-Stokes to Stokes Raman intensities.[16] Care was taken to correct for the dispersion of the scattering efficiencies by measuring the dispersion with a cw tunable dye laser. For convenience, we define an "effective phonon temperature" T_q by $N_q = (\exp(\hbar\omega/k_b T_q) - 1)^{-1}$ where ω is the LO phonon frequency and k_b is the Boltzmann's constant. In Fig. 4 we plot both T_e (open circles) and T_q (closed circles) deduced from the Raman spectra of GaAs at 77 K as a function of electron density excited by laser pulses of 600 fs FWHM. For densities less than 5×10^{17} cm^{-3} the SPE spectra are too weak for T_e to be determined reliably. Since the photoexcited hot electrons and hot phonons and the Raman spectra from these excitations were obtained from the same laser pulse, there is a question as to what these measured temperatures mean with respect to the cooling curves of the hot electrons and phonons. To answer this question, we divide the laser pulse into two halves. We assume that the hot electrons and hot phonons are excited by the first half of the laser pulse. These hot electrons and phonons then scatter the second half of the laser pulse to produce the Raman signals. Therefore, the measured temperatures represented the temperature of the hot electrons and phonons after cooling for a duration approximately less than half of the laser pulse width. A more quantitative calculation based on a sech^2(t) pulse profile showed that the measured temperature should be equal to the electron temperature after cooling for a time equal to 0.4 times the FWHM of the laser pulse.[15]

It should be noted that Raman scattering measures LO phonon with a specific wave vector determined by the incident and scattered photon wave vectors. For the backscattering geometry and photon energy we use, the LO phonons have wave vectors Q=7.5×10^5 cm^{-1}. Fortunately the distribution of
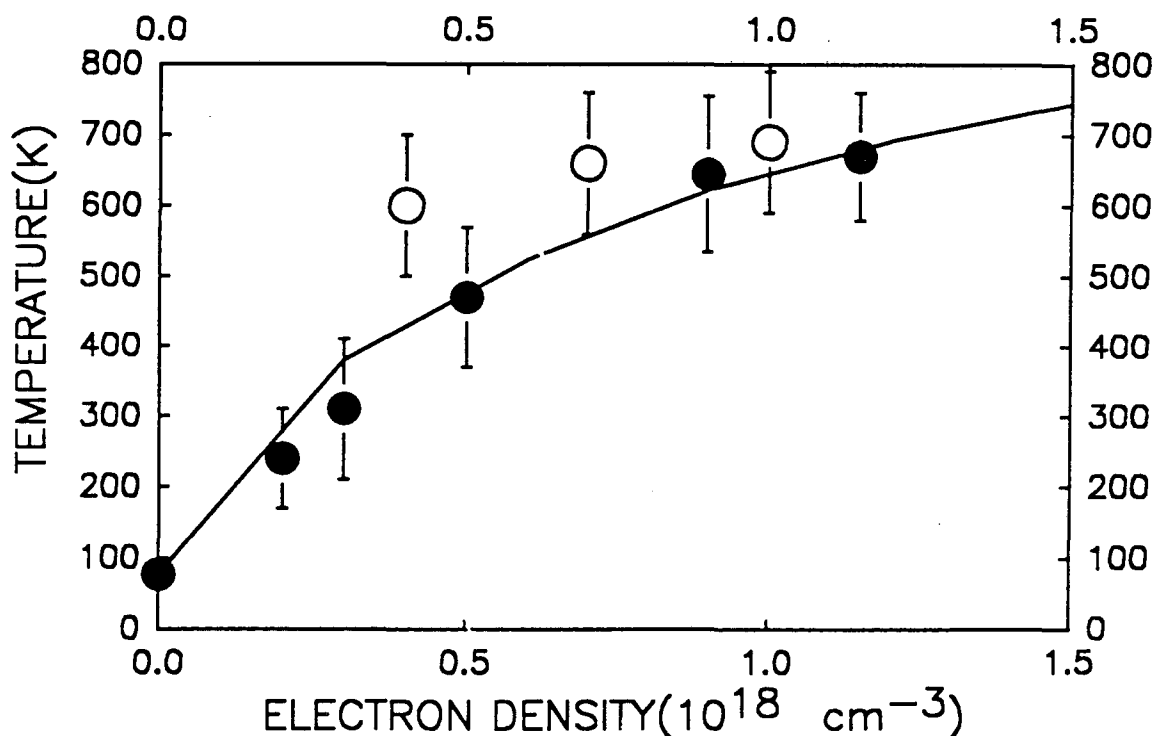

Fig. 4 Temperatures of hot electrons (open circles) and LO phonons (closed circles) in GaAs at 77 K excited by laser pulses of 600 fs FWHM. The curve drawn through the experimental points has been calculated from the multivalley model discussed in the text.

LO phonons generated by the relaxation of hot electrons in GaAs has a maximum very close to this wave vector.[16] Thus Raman scattering is a fairly sensitive probe of the nonequilibrium LO phonons generated by the relaxation of hot electrons in bulk GaAs.

The striking features of the curves shown in Fig. 4 are the strong increase in the phonon temperature for electron densities below 10^{18} cm^{-3} and the relatively low and constant value of the electron temperature around 700 K in the same range of densities. Since these hot electrons are excited with excess energy of over 400 meV, we cannot explain this low electron temperature as caused by cooling via emission of LO phonons only. With the short pulse width of 600 fs, the electrons have time to emitted only one or two LO phonons and in the process lost less than 80 meV of energy. Thus our results suggest that the electrons are cooled very efficiently by another process, not associated with the LO phonons. To confirm the existence of this additional cooling mechanism we have determined the cooling curve of the hot electrons at times shorter than 600 fs. By using different birefringent filters we have decreased the FWHM of the CPM laser pulses to 400 fs and 200 fs respectively. The spectral width of these pulses were too broad for measuring the phonon peaks but they were adequate for observing the much broader SPE scattering. The electron temperatures T_e measured in GaAs for three different laser pulse widths are shown in Fig. 5. We notice that T_e increases appreciably as the laser pulse length is decreased. This reflects the fact that when the laser pulse is shortened, the electrons have less time to cool before they are probed by light scattering. For the shortest laser pulse of 200 fs FWHM the electrons will have time to emit at most one LO phonon so the low electron temperature must be attributed to another cooling mechanism.
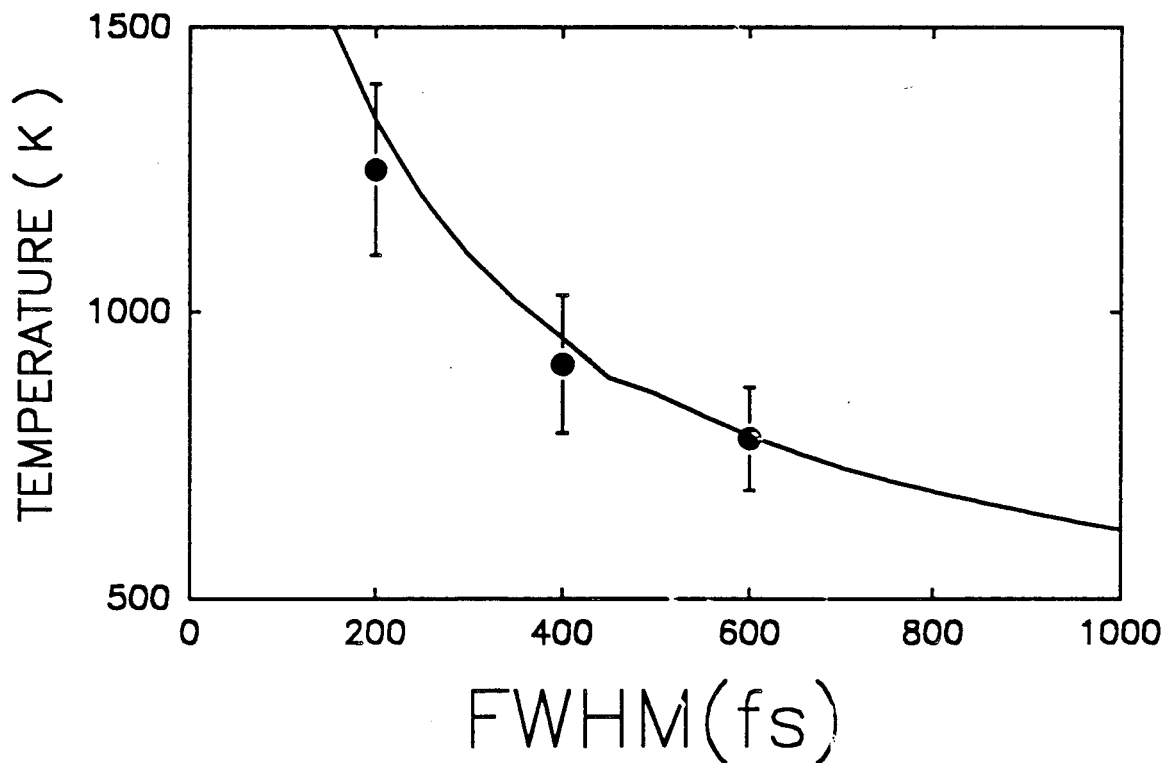

Fig. 5 Cooling curve of photoexcited hot electrons in GaAs obtained by measuring the electron temperatures as a function of the FWHM of laser pulses. The solid circles are experimental points while the curve drawn through the circles are calculated from the multivalley model discussed in the text.

MODEL CALCULATIONS

     To model the cooling curve of the hot electrons and the increase in hot phonon temperature with electron density, we have considered two possible models.

Model 1: Single Conduction Band Valley at Zone Center.

     In the first model we have neglected intervalley scattering between the conduction band minimum at Γ and higher conduction band minima at the X and L points of the Brillouin zone. The incident photons excite electron-hole pairs in the conduction and valence bands at zone center. The electrons and holes relax by emission of LO phonons via the Frohlich interaction. Within the subpicosecond time regime, other relaxation processes such as electron-TO phonon interaction, electron-acoustic phonon interaction and expansion of the electron-hole plasma are all negligible.[17] For electron densities below 10^{18} cm^{-3} it has been shown theoretically that electron-hole scattering is not as important as electron-LO scattering.[18] To simplify the calculation we further assume that the electrons thermalize to a Fermi-Dirac distribution instantaneously. This assumption is justified at electron densities above 10^{18} cm^{-3} based on both theoretical considerations and experimental evidence.[19] Although this assumption may not be valid at lower densities; it does not affect the rate at which LO phonons are produced. The cooling of the electron and LO phonon temperatures calculated within this single conduction band valley model are shown as continuous curves in Fig. 6. The electron density was assumed to be 10^{18} cm^{-3}. The electron-LO phonon scattering time was fixed at 200 fs. The behavior of T_e and T_Q shown in Fig. 6 are easily
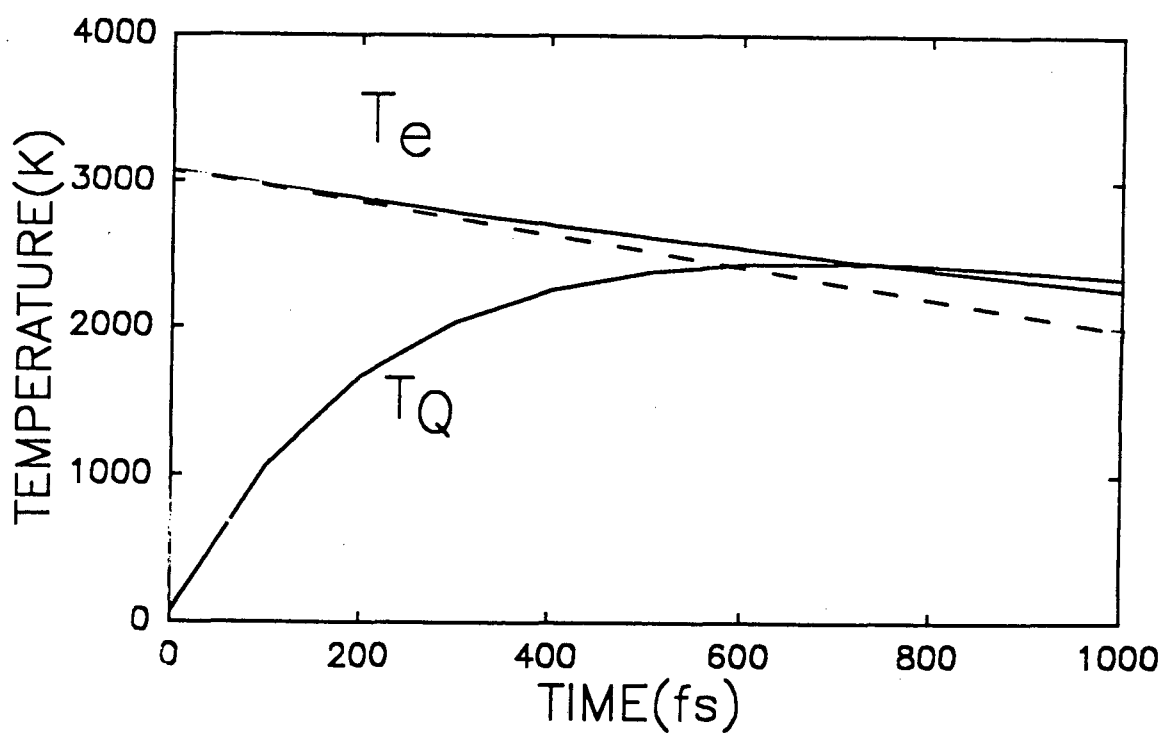

Fig. 6 Theoretical cooling curves (continuous lines) of hot electrons and phonons excited by laser pulses of 0 fs FWHM within the single conduction band model where electrons interact with LO phonons only. The broken curve is the calculated cooling curve of electrons when there are no hot phonons.

understood. The electrons cool by emission of LO phonons. As the LO phonon temperature increases, the cooling rate of the electrons slows down until at long enough time the two systems reach thermal equilibrium. The broken line in Fig. 6 shows the cooling curve of the electrons if the phonon temperature remains constant at 77 K. Thus Fig. 6 contains the essence of the "hot phonon" effect. The predictions of Fig. 6 , however, are not consistent with our experimental results. Both the electron and LO phonon temperatures predicted by this model are far too high compared with the observed temperatures. We note that our T_e are comparable to those observed by Shah et al.[2] using subpicosecond time-resolved photoluminescence. At higher electron densities we have found that T_Q overshot T_e but the highest value of T_Q achieved was much lower than that predicted by Fig. 6.[10] Other energy loss mechanisms, such as electron-hole scattering, can increase the cooling rate and lower T_e but will also predict a strong dependence of T_e on electron density. Experimentally we found that T_e is not very sensitive to electron density. At densities near 10^{19} cm^{-3} T_e actually increases with densities while electron-hole scattering will predict a decrease in T_e.[10]

Model 2: Multiple Conduction Band Valleys.

In this model we include the scattering of electrons from the Γ valley to the higher conduction band minima at X and L. It is known from study of hot electron transport in GaAs, such as the Gunn effect, that these scattering processes are much faster than intravalley scattering by LO phonons.[17] For example, in GaAs the deformation potential $D_{Γ-X}$ for the Γ to X intervalley electron-phonon scattering is about 10×10^8 eV/cm.[12] Because of the large density of states at the X valleys, electrons with high enough energy will transfer to the X valley in about 10-50 fs depending on the electron energy. Thus the intervalley processes are more efficient than the Frohlich interaction in cooling the hot electrons in the Γ valley because they remove the high energy electrons in times shorter than 100 fs. Although eventually these electrons in the higher valleys will return to the Γ valley, the return processes take more than 1 ps because the Γ valley has smaller density-of-states.[2] Thus at times shorter than 1 ps intervalley scattering can account for the very fast cooling of the hot Γ valley electrons we observed.

In calculating the cooling curves of the hot electrons, we found that the L valleys also played a significant role. If only the X valleys are included in the model, the electron temperature will drop very fast within the first 100 fs. But once the electron temperature is below 1500 K, the cooling rate decreases rapidly because there are now very few electrons left in the high energy tail of the distribution capable of transferring to the X valley. As a result the electron temperature in Fig. 4 will be about 1200 K rather than 700 K as observed. The theoretical cooling curve (continuous line) in Fig. 5 is obtained by including both the X and L valleys in the model and by setting $D_{Γ-L}$ to 7×10^8 eV/cm. Once the intervalley scattering cooling rates are known, the dependence of the LO phonon temperature on electron density can be calculated. The theoretical curve (solid line) in Fig. 4 has been obtained with a electron-LO phonon scattering time of 200 fs.

DISCUSSIONS

From the subpicosecond Raman experiments we conclude that intervalley scattering can play a dominant role in the cooling of the photoexcited hot electrons in GaAs within the first ps after excitation. The electron cooling rate depends strongly on time, density and temperature. The so-called hot phonon effect, arising from electron-LO

phonon interaction, is important only for times longer than 1 ps or for electrons with initial temperature lower than about 700 K. For higher electron temperatures the electron cooling rate within the first ps is dominated by intervalley scattering. The reason is because of the ultrafast rate of transfer of electrons from the Γ valley to the higher conduction band minima in GaAs. The separation between the Γ and X valleys in GaAs is about 500 meV. With a scattering time of about 50 fs or less, the electron cooling rate due to Γ to X intervalley scattering is about 10 meV/fs and is about two orders of magnitude larger than electron-LO phonon scattering. The Γ to L intervalley scattering plays a similar, albeit, smaller role. Both the deformation potential $D_{\Gamma\text{-}L}$ and density-of-states for the L valleys are smaller than the corresponding quantities for the X valleys. The Γ to L scattering time is about 100-200 fs. The separation between the Γ and L valley is only about 300 meV. As a result of these two factors, when T_e is >>1000 K the cooling rate due to Γ to L scattering is much smaller than that due to Γ to X scattering; although it is still larger than the electron-LO phonon cooling rate. As the electrons cool to about 1000 K or lower the Γ to L scattering becomes more important since there are very few electrons energetic enough to scatter to the X valleys. The observed T_e of about 700 K measured with laser pulse of 600 FWHM in Fig. 4 is determined mainly by the Γ to L scattering. At this electron temperature the Γ to L scattering rate is comparable to the electron-LO phonon scattering rate. Thus electrons and LO phonons can reach thermal equilibrium only after the electron temperature is lower than about 700 K.

CONCLUSIONS

In conclusion, we have studied the cooling of hot electrons and heating of LO phonons in GaAs excited by subpicosecond laser pulses as a function of photoexcited electron densities. For electron densities above 2×10^{17} cm^{-3} we found that the LO phonon temperature rose rapidly with electron densities. On the other hand the electron cooling rate was found to be too fast to be explained by the electron-LO interaction. Our results have been explained quantitatively by scattering of electrons from the zone-center conduction band valley to the higher conduction valleys at the X and L points of the Brillouin zone. By fitting our results to model calculations, we find that the electron-LO scattering time is about 200 fs and the Γ to L intervalley deformation potential is 7×10^8 eV/cm.

ACKNOWLEDGEMENTS

REFERENCES

1. W. Z. Lin, L. G. Fujimoto, E. P. Ippen, and R. A. Logan, Appl. Phys. Lett. 50, 124 (1937).
2. J. Shah, B. Deveausd, T. C. Damen, W. T. Tsang, and P. Lugli, Phys. Rev. Lett. 59, 2222 (1987).
3. C. L. Tang, F. W. Wise, and I. A. Walmsley, Solid State Electron. 31, 439 (1988).
4. J. A. Kash, J. M. Hvam, and J. C. Tsang, Phys. Rev. Lett. 54, 2151 (1985).
5. R. F. Leheny, J. Shah, R. L. Fork, C. V. Shank and A. Migus, Solid State Commun. 31, 809 (1979).

6. W. Potz and P. Kocevar, Phys. Rev. B $\underline{28}$, 7040 (1983).

7. W. Cai, M. C. Marchetti, and M. Lax, Phys. Rev. B $\underline{35}$, 1369 (1987).

8. W. W. Ruhle, K. Leo and E. Bauser, Phys. Rev. B $\underline{40}$, 1756 (1989).

9. Y. H. Huang and P. Y. Yu, Solid State Commun. $\underline{63}$, 109 (1987).

10. D. S. Kim and P. Y. Yu, Phys. Rev. Lett. $\underline{64}$, 946 (1990).

11. A. Valdmanis, R. L. Fork and J. P. Gordon, Opt. Lett. $\underline{10}$, 131 (1985).

12. C. L. Collins and P. Y. Yu, Phys. Rev. B $\underline{30}$, 4501 (1984).

13. M. V. Klein in <u>Light Scattering in Solids, Topics in Applied Physics,</u> Vol. 8, ed. by M. Cardona (Springer-Verlag, New York, 1975) p.148.

14. C. L. Collins and P. Y. Yu, Solid State Commun. $\underline{51}$, 123 (1984).

15. D. S. Kim and P. Y. Yu (unpublished).

16. C.L. Collins, Ph. D. Thesis (University of California, Berkeley, 1983, unpublished).

17. E. M. Conwell and E. O. Vassel, IEEE Trans. Electron Devices $\underline{13}$, 22 (1966).

18. M. A. Osman and D. K. Ferry, Phys. Rev. B $\underline{36}$,6018 (1987).

19. W. H. Knox, C. Hirlimann, D. A. Miller, J. Shah, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. $\underline{56}$, 1191 (1986); D. Hulin, A. Migus, A. Antonetti, and J. L. Oudar, in <u>Proceedings of the 18th International Conference on the Physics of Semiconductors</u>, ed. by O. Engstrom (World Scientific, Singapore, 1987) p.1279; and J. A. Kash and J. C. Tsang, <u>ibid</u>, p.1287.

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