LBL-23345
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April 1987

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LBL-23345 c.2

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

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To Appear in Solid State Communications LBL-23345

Femtosecond Collision Times of Hot Electrons in GaAs Determined by Picosecond Time-Resolved Light Scattering

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ABSTRACT

Picosecond laser pulses have been used to excite dense electron-hole plasmas in GaAs and to probe the dynamics of the plasmas by time-resolved Raman scattering from single particle excitations. The Raman lineshapes are explained by a theory which includes the electron collision time. For a plasma with density of 5×10^{18} cm^{-3} the collision time is found to be about 20 femtoseconds.

PACS: 78.30 Gt
72.20 Jv

It is well known that the inelastic light scattering spectrum can be used to probe properties of solid state plasmas[1]. Much theoretical and experimental work has been devoted to the use of light scattering techniques to study the plasmas, screening of longitudinal optical (LO) phonons by plasmons and single particle excitations in doped semiconductor samples. There have been relatively fewer attempts to apply this technique to study photoexcited plasmas.[2-4] Recently there has been much interest in the relaxation processes of hot electron-hole plasmas excited in semiconductors by picosecond and femtosecond laser pulses.[5] Considering the fact that light scattering spectroscopy is capable of studying both electrons and phonons in the same experiment, it appears to be an attractive technique for studying fast relaxation processes in hot plasmas on picosecond (ps) and femtosecond (fs) time scales. Recently Kash and coworkers[6] have reported subpicosecond time-resolved studies of Raman scattering by LO phonon and coupled LO phonon-plasmon modes in GaAs. In this Communication we report a picosecond time-resolved Raman study of single particle excitations in dense and hot photoexcited plasma in GaAs. By analyzing the scattering lineshape we have obtained the electron collision time and temperature as a function of time.

Our experiment was performed on a 4 micron thick, high purity epitaxial layer of GaAs grown on a [100] oriented substrate. The sample, mounted on the cold finger of a liquid nitrogen dewar, was excited by the output of a mode-locked Rhodamine 6G dye laser synchronously pumped by an actively mode-locked Ar^+ laser. The laser photon energy was typically

around 2.13 eV to avoid any resonance effect. The laser pulse duration was about 4.5 ps as determined by an autocorrelator and the pulses were separated by 12 nanoseconds from each other. The energy per pulse is typically about 1 nJ. The laser beam was passed through 4 heavy flint prisms to remove the dye fluorescence and then focused to a spot size of about 10 micron on the sample. The scattered radiation was analyzed by a Spex double monochromator and detected by a cooled photomultiplier tube using photon counting techniques. To obtain the time-resolved light scattering spectra the pulse train was divided into pump and probe beams by using a beam splitter. The pump beam, containing about 90% of the total beam energy was polarized perpendicularly to the probe beam. By using shutters the Raman signals were collected sequentially with both beams present and with only one of the two beams present. By subtracting the single beam signals from the two-beam signal with a computer, we obtained the Raman signal resulting from scattering of the probe beam from the plasma excited by the pump beam. By delaying the probe beam with respect to the pump beam we can study the time evolution of the plasma excited by the pump beam. By crossing the polarizations of the pump beam and probe beams we minimize the effect of the coherent artifact,[7] which results when the two beams overlap temporally. The signals obtained by the subtraction process were typically quite noisy and about ten spectra were averaged to achieve a reasonable signal to noise ratio.

Light can be scattered inelastically both by the collective

excitations (plasmons) and single particle excitations (SPE) in a plasma.[1] The scattering mechanisms for these processes are classified as charge density fluctuation (CDF), spin density fluctuation (SDF) and energy density fluctuation (EDF). We have selected to study SPE in our photoexcited plasma by SDF, since the resultant spectra are easier to interpret. This is achieved by polarizing the probe beam along the [011] direction and analyzing the backward scattered radiation along the [01$\bar{1}$] direction. Only the SDF mechanism is allowed for this perpendicular polarization between the incident and scattered radiation. Since the LO phonon is also forbidden for this scattering geometry, there is no background due to the coupled modes. The only disadvantage of this scattering geometry is the presence of a strong elastic peak due to the pump beam which is polarized parallel to the scattered radiation and is not completely removed by the subtraction process.

Fig. 1 shows a typical spectrum obtained before the subtraction process. Besides the SPE spectrum centered around the laser peak, a strong background due to hot luminescence is also present. Using the fact that this hot luminescence has an exponential dependence on the photon energy, it can be subtracted off to give the time-resolved SPE Raman spectra shown in Fig. 2. To interpret our spectra we utilize the SDF scattering cross section given by Hamilton and McWhorter :[8]

$$\frac{d^2\sigma}{dw d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 [1+n(w)] \frac{\hbar}{\pi} \mathrm{Im}[K_2(q,w)] \qquad (1)$$

where $n(w) = [\exp(\hbar w/K_B T_e)-1]^{-1}$, K_B is the Boltzmann constant, T_e is the carrier temperature and $K_2(q,w)$ is a response function which is essentially proportional to the dielectric function $\varepsilon(q,w)$. Equation (1) has been found by Abramsohn et al.[9] to explain SPE lineshapes adequately only for low density plasmas. However these authors also found that for plasma densities $N > 2 \times 10^{16}$ cm^{-3} the experimental SPE lineshape in GaAs was Lorentzian and narrower than predicted by Eq.(1). Collins and Yu[10] have also found that Eq.(1) did not fit the SPE lineshape of dense hot plasmas in GaAs excited by picosecond laser pulses. Tsen and Bray[11] showed that for $N > 7 \times 10^{16}$ cm^{-3} the SPE lineshape could be explained by Eq.(1) provided collision effects were included in $\varepsilon(q,w)$ by using the Lindhard-Mermin expression.[12]

The broken curves in Fig. 2 are plots of Eq.(1) where $K_2(q,w)$ is now replaced by the Lindhard-Mermin dielectric function $\varepsilon_{LM}(q,w,\tau)$ where the collision time τ and the carrier temperature T_e are treated as adjustable parameters. In fitting the experimental curves, it is also necessary to know the carrier concentration. We have estimated the carrier density excited by the pump beam both from the incident laser power and from the coupled-mode Raman spectra[10] obtained in the scattering geometry: incident and scattered radiation parallel to each other. The maximum density deduced from the coupled mode was $\sim 5 \times 10^{18}$ cm^{-3} at 0 ps delay and was comparable to the

calculated density assuming a Gaussian profile equal to that of the laser beam. Once the initial carrier concentration is known, the carrier densities at subsequent delays can be determined from the integrated intensity of the SPE spectra. The value of N, τ and T_e deduced for the three delay times shown in Fig. 2 are 5×10^{18} cm^{-3}, 20 fs, 800 K for 0 ps delay; 2.4×10^{18} cm^{-3}, 35 fs and 500 K for 10 ps delay; and 0.8×10^{18} cm^{-3}, 70 fs and 400 K for 20 ps delays. We should point out that the carrier temperature T_e is essentially determined by the ratio of anti-Stokes to Stokes intensity while τ is determined by the linewidth. The lineshape of the SPE is not very sensitive to the carrier density for the small τ values we obtained here. If we assume that the carrier density profile is Gaussian like the incident laser intensity, the net effect is to increase τ by less than 5 fs. The estimated uncertainty in the values of τ and T_e we obtained from the SPE spectra are about 5 fs and 100 K respectively.

The hot carrier collision times we obtain from our Raman results are much shorter than those reported in previous Raman experiments in GaAs by Romanek et al.[2] and by Pinczuk et al.[3] because of the high density and carrier temperature in our experiment. More recently, using a fs laser, Oudar et al.[13] have determined a carrier-carrier dephasing time of ~ 300 fs at a density of 3×10^{17} cm^{-3}. In another measurement with fs laser, Rosker et al. [14] determined the scattering time to be 32 fs in GaAs for carrier for density of 3.0×10^{18} cm^{-3}. Studies of hot electron transport in thin (< 600 A) GaAs devices have

suggested that the electron collision time is > 50 fs for carrier densities of the order of 1×10^{18} cm^{-3}. [15, 16] Thus the recent experimental results are consistent with the collision times we obtain. In addition the decrease in the measured collision time with delay can be explained by decrease in the plasma density and temperature. It is tempting to deduce the plasma cooling rate from the measured T_e versus delay. Unfortunately, because our pulse length is relatively long compared to the cooling time of the plasma, our value will at best represent some time-averaged cooling rate.

In conclusion, we have used picosecond laser pulses to excite hot and dense plasma in GaAs. Using time-resolved Raman scattering due to SPE in the plasma, we have determined the electron temperature and collision time as a function of time. The collision time for a plasma of density equal to 5×10^{18} cm^{-3} and a temperature of 800 K is found to be only 20 fs.

ACKNOWLEDGEMENTS

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Science, Materials Sciences Division of the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

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FIGURE CAPTIONS

Fig. 1 A typical spectrum obtained at zero delay showing the Raman scattering from the photoexcited carriers superimposed on a hot luminescence background.

Fig. 2 Raman spectra due to single particle scattering by the photoexcited plasma for three different time delays. The solid curves are the experimental curves obtained by averaging typically ten spectra similar to those shown in Fig. 1 after subtracting out the hot luminescence background. The broken curves are theoretical fits to the experimental curves as discussed in the text.

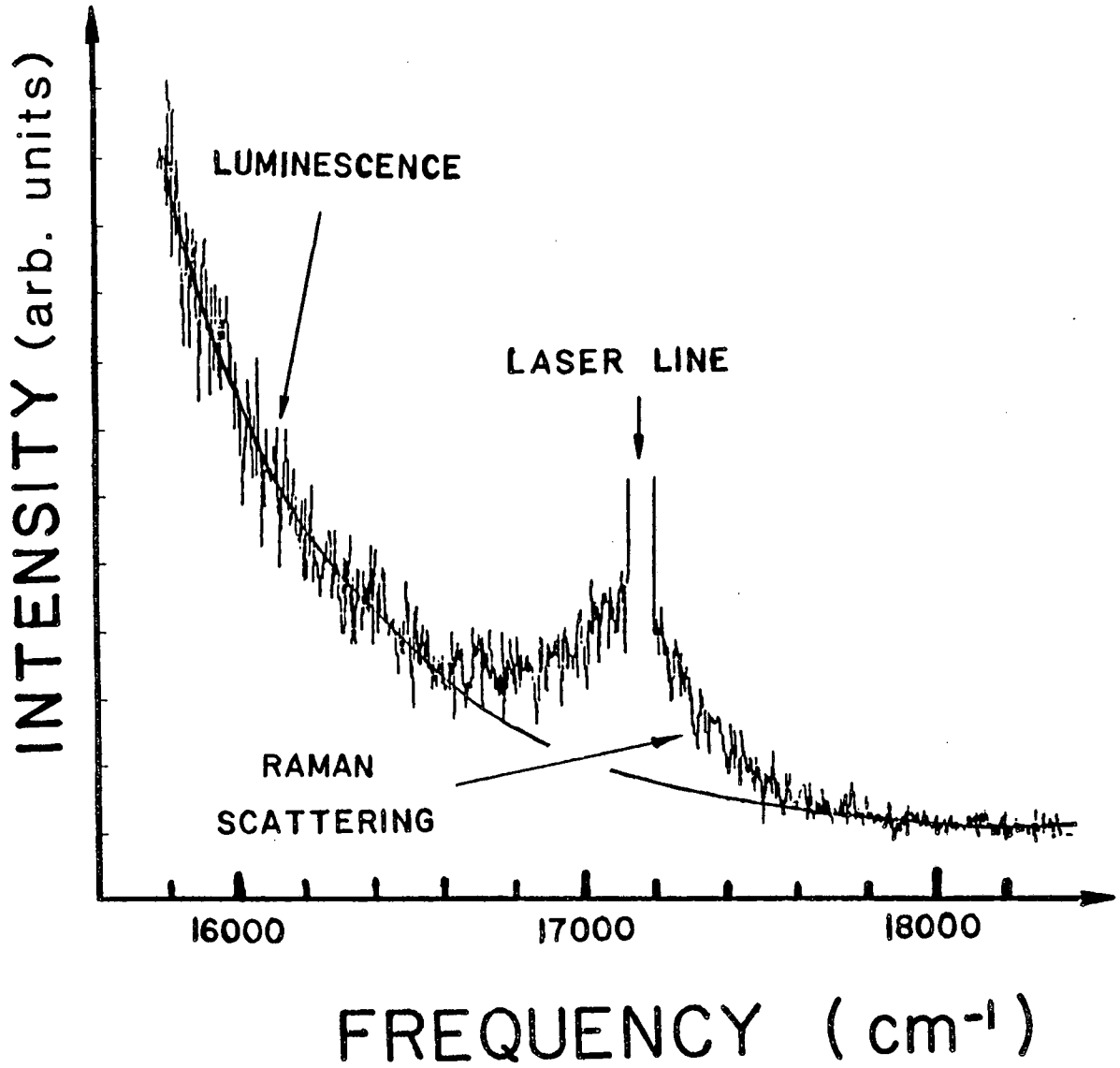
INTENSITY (arb. units)
LUMINESCENCE
LASER LINE
RAMAN
SCATTERING
16000
17000
18000
FREQUENCY (cm⁻¹)

Fig. 1

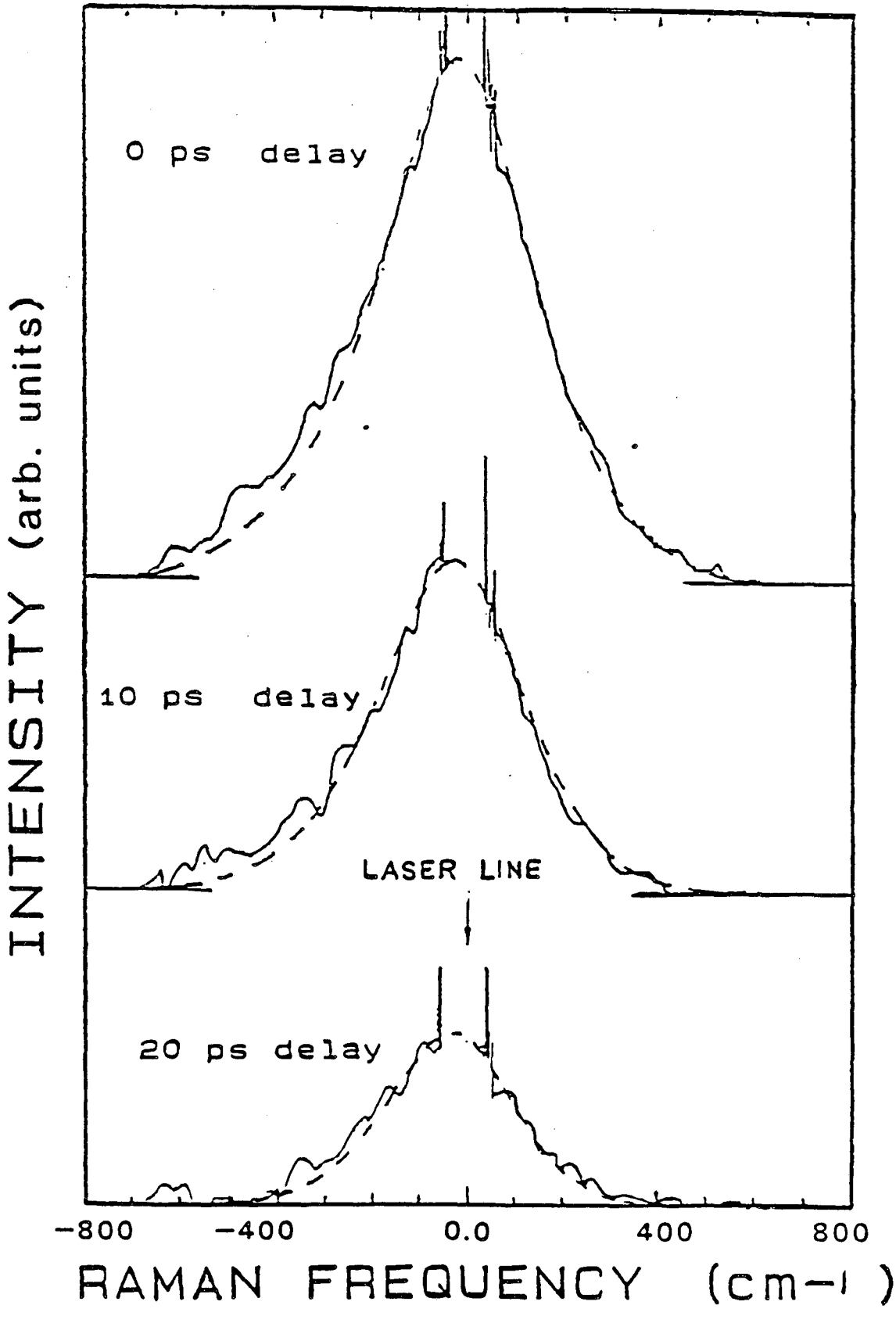
0 ps delay
10 ps delay
LASER LINE
20 ps delay
INTENSITY (arb. units)
-800
-400
0.0
400
800
RAMAN FREQUENCY (cm-1)

Fig. 2

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