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C.L. Collins and P.Y. Yu

March 1984

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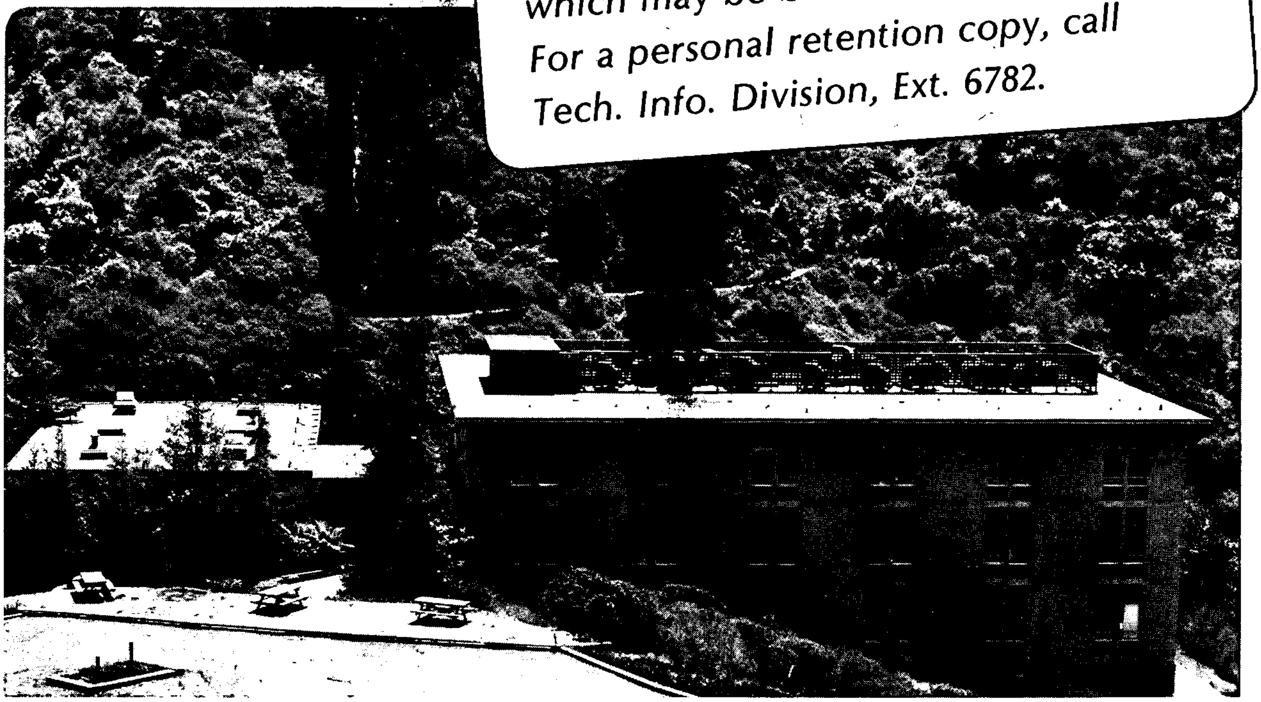

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Light Scattering from Non-equilibrium Electron-Hole Plasma Excited by Picosecond Laser Pulses in GaAs

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Abstract

A picosecond dye laser is used to excite electron-hole plasma of density between 10^{18} to 10^{19} cm^{-3} in GaAs. The plasma density and distribution function within $\leq$ 20 psec of excitation is probed by Raman scattering. The lineshape of the single particle excitation spectra of the plasma can be explained only by assuming that the electron distribution function is in nonthermal equilibrium.

This research is supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract Number DE-AC03-76SF00098. The loan of a cavity dumper from the San Francisco Laser Center (supported by the National Science Foundation, NSF Grant No. CHE79-16250, and the National Institute of Health, NIH Grant No. P41 RR01613-02, awarded to the University of California at Berkeley in collaboration with Stanford University.), GaAs samples from the Hewlett-Packard Company and helpful discussion with R. Bray are gratefully acknowledged.

Photoexcited electron-hole plasma (EHP) in GaAs has been the subject of intensive studies recently using both picosecond (psec) and nanosecond laser excitations.[1] In most of these studies it is found that the EHP attained thermal equilibrium within one psec of excitation and the subsequent behavior of the EHP can be explained by its cooling and expansion. Although in a few studies[2] it was noted that in GaAs the EHP distribution did not reach thermal equilibrium for as long as 10 psec when the plasma density was higher than 10^{18} cm^{-3}, the nonequilibrium plasma distribution was not determined. In this Communication we have utilized light scattering from the photoexcited plasma to confirm that the plasma distribution was indeed nonequilibrium and furthermore to determine this nonequilibrium distribution. In addition we have measured parameters such as density and expansion velocity of the EHP.

Our experiment was performed on a 4μ thick, high purity (N_A, $N_D \leq 10^{14} cm^{-3}$) epitaxial layer of GaAs oriented perpendicular to the (100) direction. The sample is cooled by He exchange gas in an optical dewar. The lattice temperature as estimated from the time-resolved luminescence spectra is ~20 K. The EHP is excited by the output of a modelocked and cavity-dumped Rhodamine 6G dye laser. The length of the output pulses is typically ~20 psec long and separated from each other by 250 nsec. The energy of the dye laser pulses is typically ~20 nJ. Power density as high as 10^7 W/cm^2 can be obtained by focusing the laser beam to a spot of ~100μ diameter on the sample surface. By changing the focus we can vary the EHP density from 10^{18} to 10^{19} cm^{-3}. The same dye laser pulses which excite the EHP are used also to scatter from the EHP. Since Raman scattering occur instantaneously there is zero time delay between the excite and probe pulses. As a result the Raman signal represents an average of the EHP behavior within ~20 psec after excitation. The incident and scattered radiations are polarized respectively along the (010) and (001) axes. The backscattered Raman radiation is analysed first by a Spex double monochromator and then detected by a gated photon counting system. By collecting only light scattered within 2 nsec of the incident laser pulse we reduce the dark counts by a factor of ~125.

It is well known that there are two kinds of elementary excitations in a plasma which can scatter radiation.[3] The first kind are known as collective modes and involve collective oscillations such as plasmons and acoustic plasmons. In crystals like GaAs a slight complication results from the fact that the longitudinal electric field of the plasmon can couple to the corresponding field of the longitudinal optical (LO) phonon to form coupled plasmon-LO phonon modes (to be abbreviated as coupled modes). The second kind are known as single particle excitations (SPE) and involve excitations of carriers from a filled state to an empty state. Thus in principle it is possible to determine the plasma density from the coupled modes and the plasma distribution function from the SPE scattering. We will show that this ability of light scattering to measure both the density and distribution function of the EHP independently is crucial to our conclusion that the EHP distribution is nonthermal equilibrium.

We first present the coupled mode spectra we obtained at different excitation intensities in Fig. 1(a). We found that when GaAs is strongly excited energetic carriers are created which produce intense photoluminescence extending into the visible.[4] To minimize this hot luminescence we have tuned the dye laser into the green (ω=17018 cm^{-1}). In the higher excitation spectra in Fig. 1(a) a background due to SPE scattering appears. It has been shown that the coupled plasmon-LO phonon frequency is given by:[5]

$$\omega_\pm^2 = \frac{1}{2}(\omega_{LO}^2+\omega_p^2) \pm \frac{1}{2}[(\omega_{LO}^2+\omega_p^2)^2 - 4\omega_p^2\omega_{TO}^2]^{\frac{1}{2}} \quad (1)$$

where ω_{LO} and ω_{TO} are respectively the frequencies of the transverse optical (TO) and LO phonons and ω_p is the bulk plasmon frequency:

$$\omega_p^2 = \frac{4\pi Ne^2}{\varepsilon_s}\left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right) \quad (2)$$

In Eq.(2) N is the density of electron-hole pair excited, ε_s is the bound electron dielectric constant and m_e^* and m_h^* are respectively the effective masses of the electron and hole (both assumed to be isotropic for simplicity). In GaAs $m_e^* << m_h^*$ for the heavy hole band so ω_p is approximated by $(4\pi Ne^2/m_e^*\varepsilon_s)^{\frac{1}{2}}$. A plot of Eq.(1) for GaAs is shown in the inset of Fig.1(a). Our result appears to contradict Eq. (1) because instead of observing coupled modes whose frequencies $\omega_\pm$ varies with density and hence laser intensity according to Eq. (1) we observe two peaks at ω_{LO} and ω_{TO} whose relative height varies with laser intensity. Our results can be understood if we include the spatial distribution in the plasma density in our analysis. The density distribution of the plasma is influenced by two factors: the penetration depth of the incident laser is only ~0.2μ so the plasma is essentially created at the sample surface and the radial intensity distribution of the incident laser beam which is a Gaussian. As pointed out by Turtelli and de Castro[6] the coupled mode lineshapes are influenced more strongly by the radial distribution in the plasma density. We have therefore assumed that the radial dependence of the EHP density, N(r), has the same Gaussian profile as the laser:

$$N(r) = N_o \exp[-(r/r_o)^2] \qquad (3)$$

where $r_o \gtrsim 50\mu$ is the beam size. The scattered spectra $I(\Delta\omega)$ can be calculated from the expression:

$$I_s(\Delta\omega) \propto \int_0^\infty I_\ell(r)\eta(r)2\pi r\, dr \qquad (4)$$

where $\Delta\omega$ is the Raman shift, I_ℓ is the incident laser intensity profile and $\eta(r)$ is the scattering efficiency of the EHP with density N(r) which determines $\Delta\omega$. The expression for $\eta(r)$ can be found in Ref. 7. The calculated spectra with N_o and the plasma damping constant (fixed at 250 cm^{-1} for all the curves) as the only adjustable parameters are shown in Fig. 1(b). As can be seen the calculated curves reproduce quite well the experimental curves with values of N_o typically a factor of 3-5 smaller than the nominal values N_o' calculated from the laser intensity. The good correlation between N_o determined from the coupled modes and N_o' will be assumed when we study the SPE spectra.

To make it easier to measure the SPE spectra we have lowered the laser frequency to 15793 cm^{-1} to take advantage of the resonance enhancement due to the $E_o+\Delta_o$ transition in GaAs[8] Two typical SPE spectra we obtained after subtracting off a smooth hot luminescence background are shown in Fig. 2. For comparison the theoretical SPE spectra for thermal equilibrium distributions of electrons are shown qualitatively in the inset of Fig. 2.[9] Curve (a) represents the result for degenerate electron at $T \gtrsim 0$ K. Curve (b) which is essentially a Gaussian distribution is for a nondegenerate electron gas with a Maxwell-Boltzmann velocity distribution. In both cases if the plasma is expanding rapidly in the direction of the incident radiation, the expansion velocity can be determined from the Doppler shift of the scattering spectra. If we assume that the EHP distribution is in equilibrium and its density is adjustable we find that the experimental SPE spectra can be fitted satisfactorily by assuming that the electron distribution is a drifted Maxwell-Boltzmann corresponding to $T \lesssim 500$ K and a drift velocity of $\sim 10^7$ cm/sec. Because of the small electron mass in GaAs in order the EHP distribution be Maxwell-Boltzmann N_o must be less than 10^{17} cm^{-3} even at $T \sim 500$ K. However based on the laser intensity we expect a EHP density in excess of 10^{18} cm^{-3}!

To try to resolve this discrepancy we have considered corrections to the theoretical model due to spatial variation in the EHP density; spatial variation in the plasma temperature analogous to the density variation and increase in the plasma damping constant.[10] Details of these analyses will be published elsewhere.[11] In Fig. 3 we show how the theoretical SPE spectra assuming thermal equilibrium distribution functions compare with the experimental SPE spectra at the highest laser intensity. Note how the calculated spectra always have a steeper edge on the Stokes side than the experimental spectrum because of the degenerate nature of the distribution. Thus we are led to conclude that in GaAs the EHP has not reached thermal equilibrium within ~20 psec after excitation at density larger 10^{18} cm^{-3} as found by Leheny et al.[2] It is beyond the scope of this short article to discuss the reasons why the equilibration time of the EHP in GaAs, which is known to be less than 1 psec at low densities ($<10^{18}$ cm^{-3}) and at high densities ($>10^{21}$ cm^{-3}), becomes longer than 10 psec at intermediate densities. Presumably this is because at these intermediate densities the electron density is high enough to screen the electron-LO phonon interaction[2,12] but not high enough for Auger processes to occur in less than 1 psec.

One advantage of the SPE spectra is that it is a rather direct measure of the distribution function. Since equilibrium distributions fail to explain the experimental spectra satisfactorily, we have adopted a phenomenological approach of fitting the experimental curves with trial nonequilibrium distributions. One class of distributions which we found to work rather well is a power law of the form:

$$n(E) = AE_o^r(E+E_o)^{-r} \qquad (5)$$

where A, r and E_o are adjustable parameters subject to the only condition that $A \leq 1$. In Fig. 2 the smooth solid curves are obtained with a drifted nonequilibrium distribution of this form. The values of the parameters chosen are: $E_o = 800$ cm^{-1}, r=2 and drift velocity=10^7 cm/sec for both curves. The values of A have been chosen to be 0.67 and 0.22 for the two curves to correspond to estimated EHP densities of 6×10^{18} cm^{-3} and 2×10^{18} cm^{-3} respectively. The uncertainty in the choice of r is $\sim\pm 1$. For example equally good fits can be obtained with r=3 by changing A. The significance of the parameters E_o and A is that E_o determines the width of the SPE spectra and plays the role of temperature in the equilibrium distribution while A is proportional to the plasma density. The crucial feature in the nonequilibrium distribution of Eq.(5) as compared to the equilibrium distribution is that the electrons are more spread out in energy. As a result even for densities as high as 10^{19} cm^{-3} the occupancy of any electronic state is less than one. In other words the nonequilibrium distribution is nondegenerate so that all electrons, irrespective of their energy, can participate in the scattering. The nonequilibrium distribution we propose can also explain qualitatively the transmission results of Leheny et al.[2]

Finally we note that the plasma expansion we determine from the SPE spectra is consistent with previous experiments in GaAs.[13] The fact the plasma is expanding does not invalidate our analysis of the coupled mode results. We estimated that the plasma should expand by no more than ~2μ during light scattering. This amount is not signifcant for the radial distribution of the EHP since $r_o \gtrsim 50\mu$. However the expansion of the EHP into the bulk has the effect of lowering the plasma density and can possibly explain the difference between the density calculated from the laser intensity and the density determined by fitting the coupled modes. A caveat in our calculation of the SPE spectra based on the nonequilibrium distribution is that we have assumed that the SPE scattering cross-section derived by the fluctuation dissipation theorem[14] remains valid for nonequilibrium carrier distributions. This is still an open question worthy of further investigations.

Acknowledgement--This research is supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division of the U.S. Department of Energy under Contract number DE-AC03-76SF00098. The loan of a cavity dumper from the San Francisco Laser Center (supported by the National Science Foundation, NSF Grant No. CHE79-16250, and the National Institute of Health, NIH Grant No. P41 RR01613-02, awarded to the University of California at Berkeley in collaboration with Stanford University.), GaAs samples from the Hewlett-Packard Company and helpful discussion with R. Bray are gratefully acknowledge.

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[+] Present address: Shell Development Company, P.O.Box 481, Houston, TX 77001.

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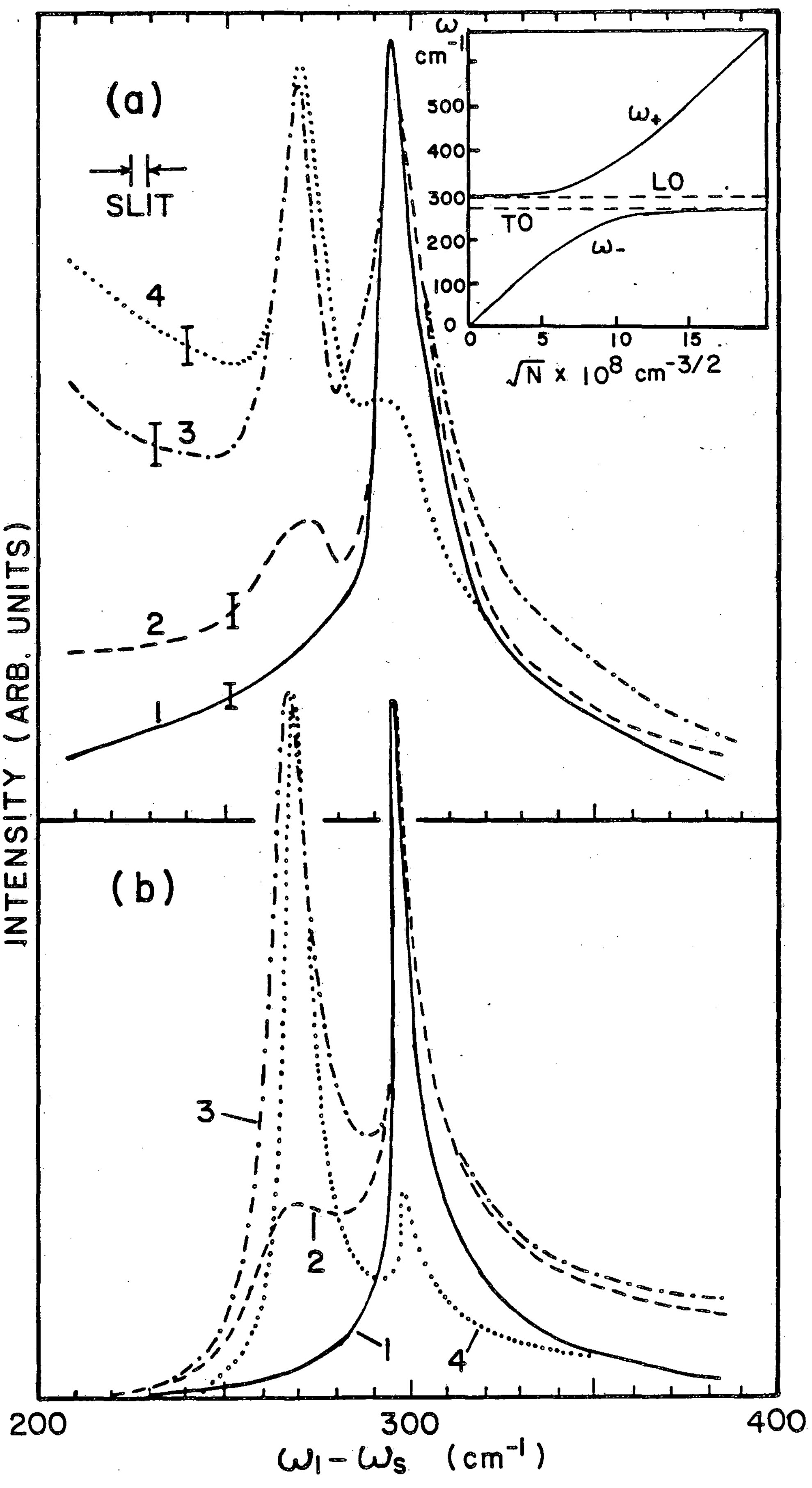
Figure Captions

Fig. 1: Experimental (a) and theoretical (b) Raman spectra of the coupled modes of EHP in GaAs. The nominal plasma densities N'_o (in $10^{18}cm^{-3}$) calculated from the laser intensity for the four experimental curves are: 1-3.4, 2-7.0, 3-13.6 and 4-28. The corresponding plasma densities N_o (in $10^{18}cm^{-3}$) used in computing the theoretical curves are: 1-1.0, 2-2.5, 3-4.6 and 4-9×10^{18}. The inset in (a) shows the frequencies of the coupled modes vs electron density calculated from Eq.(2) and confirmed experimentally in Ref. 5.

Fig. 2: Experimental (light curve with noise) and theoretical (heavy smooth curve) SPE Raman spectra of EHP in GaAs. The nominal plasma densities for the curves 1 and 2 are repectively 2.7 x $10^{18}cm^{-3}$ and 2.4 x $10^{19}cm^{-3}$. The inset shows qualitatively the SPE spectra for a degenerate (curve a) and non-degenerate (curve b) electron gas. V_{TH} and V_F are respectively the thermal and Fermi velocities of the electron gas.

Fig. 3: Two theoretical fits to the experimental SPE spectra (same as curve 2 in Fig. 2) using thermal equilibrium distribution functions. For theoretical curve A a Fermi-Dirac distribution corresponding to T=400 K, density of 2.8×10^{18} cm^{-3} and electron damping constant of 560 cm^{-1} is assumed. For curve B the Fermi-Dirac distribution is further assumed to be drifting at 6×10^{7} cm/sec along the direction of observation.


(a)
SLIT
4
3
2
1
(b)
3
2
1
4
INTENSITY (ARB. UNITS)
200
300
400
$\omega_l - \omega_s$ (cm^{-1})
ω cm^{-1}
500
400
300
200
100
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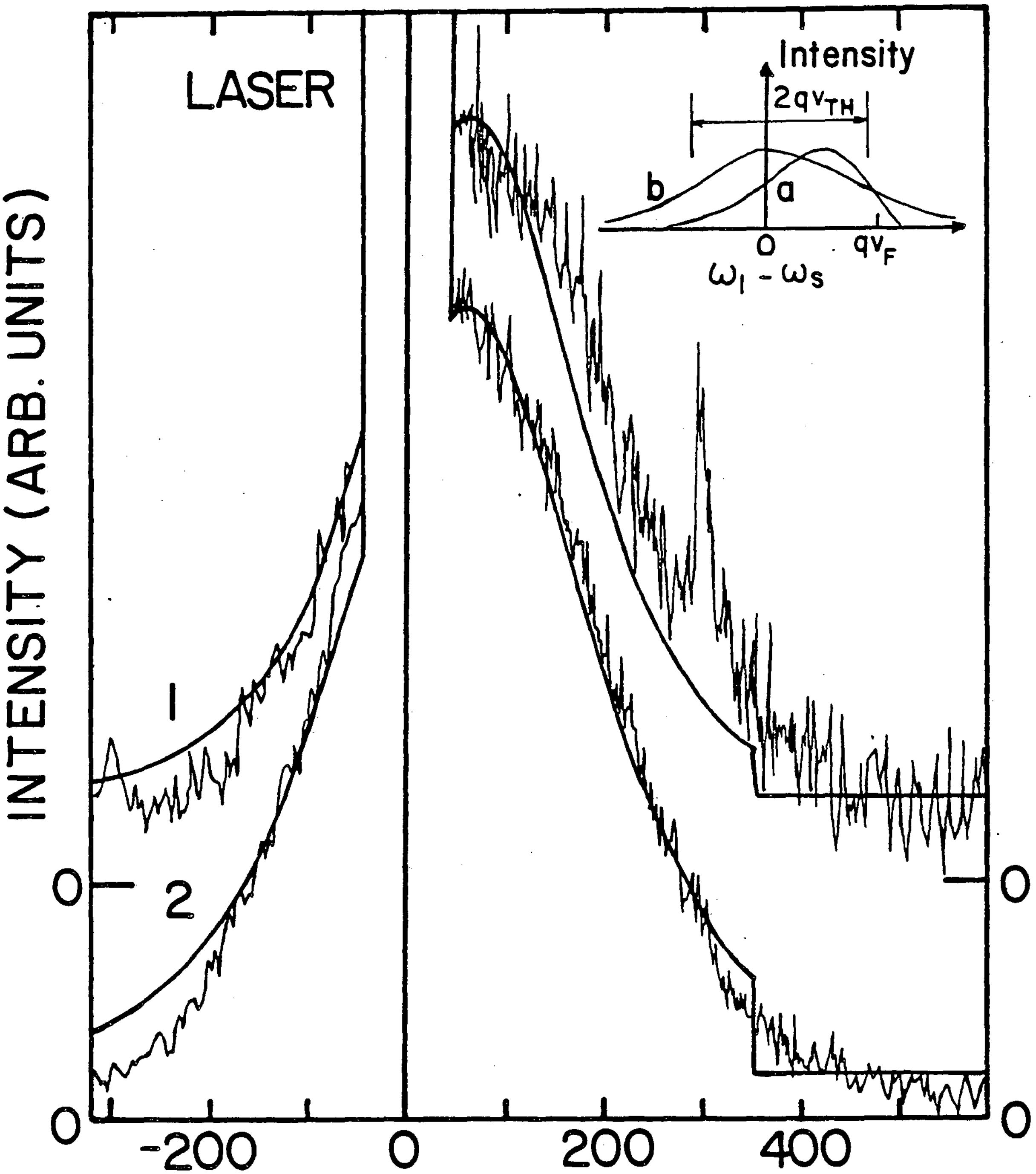
15
$\sqrt{N} \times 10^8$ cm$^{-3/2}$

Figure 1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$\omega_l - \omega_s$ (cm^{-1})
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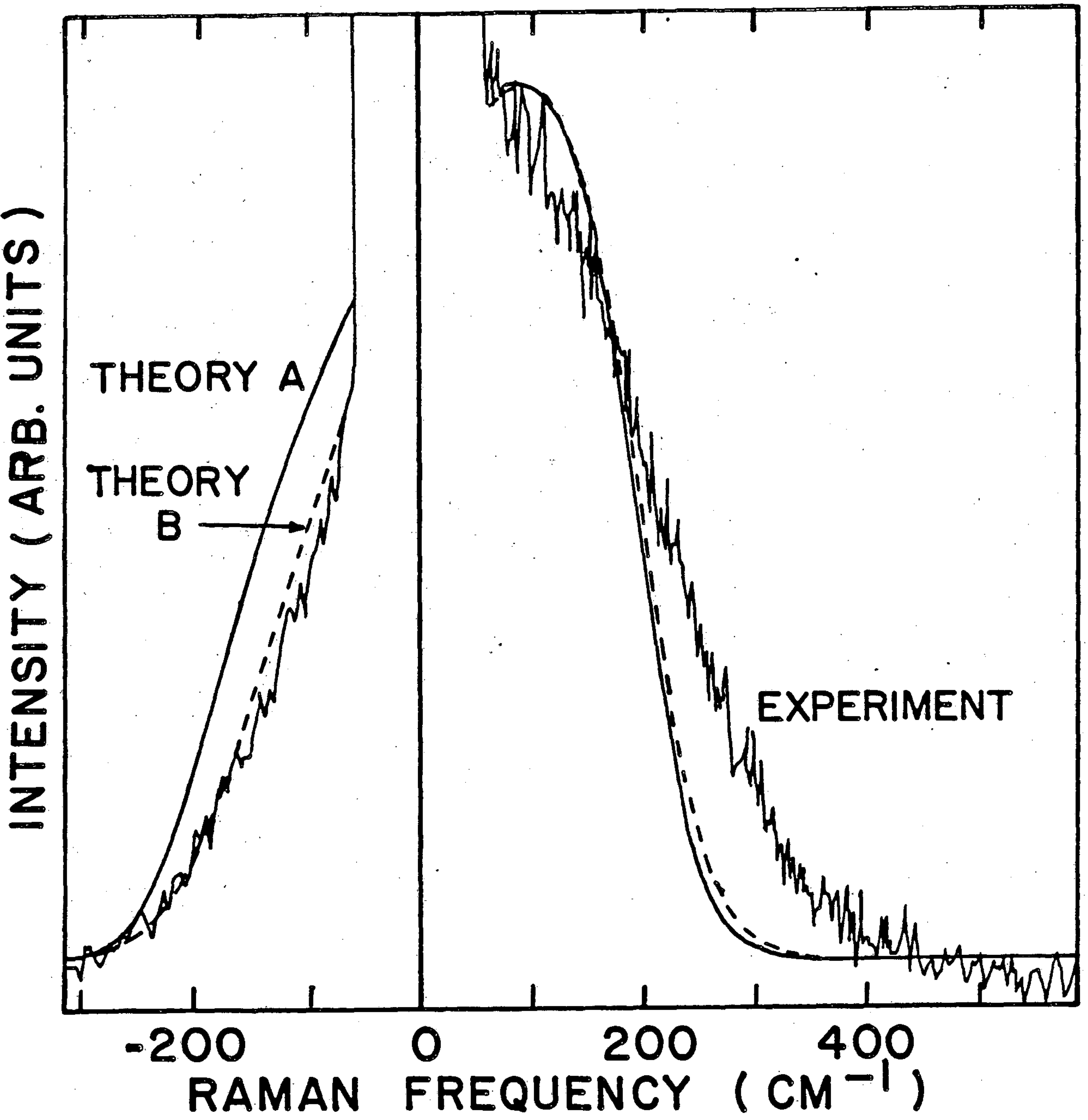
$\omega_l - \omega_s$

Figure 2


INTENSITY (ARB. UNITS)
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THEORY
B
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0
200
400
RAMAN FREQUENCY (CM^{-1})

Figure 3

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