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Optical pumping of hot LO phonons in GaAs has been studied as a function of the excitation photon frequency. The experimental results are in good agreement with a model calculation which includes both inter- and intra-valley electron-phonon scatterings. The Γ-L and Γ-X intervalley electron-phonon interactions in GaAs have been estimated.

1. INTRODUCTION

Optical pumping of the polar longitudinal optical (LO) phonon in GaAs was first observed by Shah et al.[1] Using a tightly focussed cw Ar$^+$ laser beam they monitored the population of zone-center LO and TO (transverse optical) phonon modes by the ratio of the Antistokes to Stokes Raman scattering intensities. By increasing the incident laser power from 0.1 to 1 watt they observed a linear increase in the population of the LO phonon but not of the TO phonon. They concluded that a non-equilibrium LO phonon population is created by the relaxation of photoexcited carriers and the carrier-LO phonon scattering time was short compared to the LO phonon lifetime. Subsequent experiments[2,3] found that optical pumping of both the LO and TO phonons could be observed in varying degrees depending on the materials studied. Since in these experiments the lattice temperatures remain considerably lower than the effective temperature of the phonons which are optically pumped, these non-equilibrium phonons are often referred to as 'hot phonons'.

Recently von der Linde et al.[4] studied the time dependence of the generation and decay of hot LO phonons in GaAs using picosecond (psec) laser pulses. The 2.5 psec long dye laser pulses are split into two parts. One was used to excite the hot LO phonons while the other, after a variable time delay, was used to probe the phonon population by Raman scattering. Von der Linde et al.[4] found that the hot LO phonon relaxation time in GaAs was 7 psec equal to the LO phonon lifetime deduced from the Raman linewidth.

In this paper we report an experimental and theoretical study of the dependence of the hot phonon population in GaAs on the exciting photon energy $\hbar\omega_1$. We have observed an interesting drop in the hot phonon population at $\hbar\omega_1 \simeq 2.1$eV. By comparing our experimental results with a model calculation we conclude that this decrease in hot phonon population is due to the scattering of electrons in the Γ valley to the X valleys. Values of Γ-X and Γ-L electron-phonon scattering matrix elements in GaAs are deduced.

2. EXPERIMENTAL DETAILS AND RESULTS

The experiment was performed in a backscattering geometry on a high purity (N_D, $N_A \le 10^{14}$ cm^{-3}), 4μ thick layer of GaAs grown by liquid phase epitaxy. The sample surface orientation is (100) and the incident laser is polarized along (010). The Raman selection rules[5] forbid TO phonon scattering in this geometry while the LO phonon scattering is allowed for scattered radiation *polarized perpendicularly to the incident radiation*. The sample is mounted on a cold finger and cooled to ~10° K by He exchange gas. The hot LO phonons are excited and probed by the same modelocked dye laser beam which consists of a train of ~4 psec long pulses at a repetition rate of ~ 82 MHz. The dyes we used were Rhodemine 6G and DCM allowing the photon energy to be varied from 1.75 to 2.15eV with typical average powers of ~ 80mW. The laser pulse lengths are continuously monitored by an autocorrelation setup using second harmonic generation to ensure that the pulse lengths do not vary with time and photon energy. The Raman scattering is analyzed with a double monochromator and photon counting electronics both interfaced with a microcomputer. The Raman cross sections are calculated with corrections for the spectral response of the monochromator and photomultiplier tube, the background due to luminescence from the $E_o + \Delta_o$ transition and variation in the penetration depths of the incident and scattered radiation.

The LO phonon population N_q is computed from the corrected Antistokes and Stokes Raman cross sections (σ_{AS} and σ_S) via the formula $N_q = \sigma_{AS}/(\sigma_S - \sigma_{AS})$. The LO phonon population of GaAs determined in this way are plotted as solid circles in Fig.1 as a function of the incident *photon energies. We note that the observed* values of N_q are many orders of magnitude higher than expected from the sample temperature of ~ 10° K. To rule out the possibility of local sample heating due to the focussed laser beam we estimated the lattice temperature from the photoluminescence spectra and from the LO phonon frequency. In both cases the lattice temperature is found to be ≤ 15° K. To further confirm that the large N_q we observed is due to optical pumping and not to lattice heating the modelocker

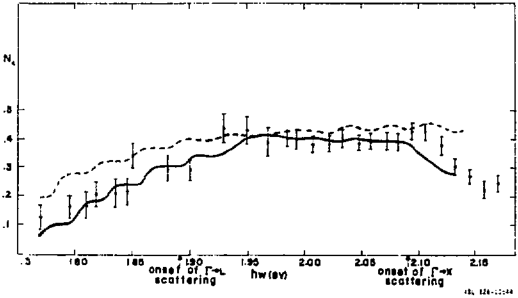

Figure 1: Phonon occupation number, N_q vs. incident photon energy:

Broken curve - theory, parabolic conduction band with no intervalley scattering

Solid curve - theory including nonparabolic conduction band and intervalley scattering

Solid circles - experimental result

was switched off so the laser becomes cw. In this case the Antistokes scattering was undetectable.

3. THEORETICAL MODELS AND CALCULATIONS

To understand the large N_q we obtained in GaAs by optical pumping and especially the decrease in N_q at 2.1eV we performed a calculation based on the following model.

Figure 2 shows schematically the band structure of GaAs. For $\hbar\omega_i < E_0 + \Delta_0$ the incident laser pulses excite electrons from the heavy and light hole bands to the conduction band. For $\hbar\omega_i > E_0 + \Delta_0$ the split-off band can contribute also. These optical transitions create an excess population of electrons and holes which relax in ~ 1 psec to the band minima via emission of LO phonons. This relaxation process is shown as a arrow labelled 1 in Fig. 2 and is responsible for creating the hot LO phonon population. This scattering process involves carriers in the Γ valley only and is referred to as intravalley scattering. As $\hbar\omega_i$ is increased, the photoexcited electrons in the Γ valley may be sufficiently high in energy to relax also to the L and X conduction band minima via emission of an intervalley phonon (indicated by arrows 2 and 3 in Fig. 2). From high field transport studies in GaAs it is known that the Γ-X intervalley scattering is faster than intravalley scatterings.[6] Because of the large density-of-states of the L and X valleys carriers will return to the Γ valley at a much slower rate. Thus we expect a decrease in N_q when $\hbar\omega_i$ is larger than the onsets for Γ-X and Γ-L intervalley scatterings.

To calculate N_q quantitatively we have to solve a set of 3 coupled rate equations involving the LO phonons, the electrons and the holes.[7] To simplify the problem we assume (1) Only spontaneous emission of LO phonons by electrons is important. (2) Optical transitions from the light hole and split-off hole bands are negligible. The validity of these assumptions will be discussed elsewhere.

Under these assumptions the Boltzmann equations for the electron distribution f_k and the phonon occupation number N_q reduces to:

$$\frac{df_k}{dt} = \left(\frac{\partial f_k}{\partial t}\right)_g - \left(\frac{\partial f_k}{\partial t}\right)_{e-ph} - \frac{f_k}{\tau_k} \qquad (1)$$

and

$$\frac{dN_q}{dt} = \left(\frac{\partial N_q}{\partial t}\right)_{e-ph} - \frac{N_q}{\tau_p} . \qquad (2)$$

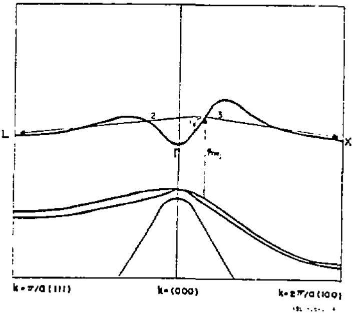

Figure 2: Scattering processes for an electron injected into the Γ valley by a photon of frequency $\hbar\omega_i$: (1) intravalley scattering by emission of LO phonons; (2) intervalley scattering to the L valleys for $\hbar\omega > 1.89$eV; (3) intervalley scattering to the X valleys for $\hbar\omega > 2.11$eV.

The rates $\left(\frac{\partial f_k}{\partial t}\right)_g$ *and* $\left(\frac{\partial f_k}{\partial t}\right)_{e-ph}$ stand for the rate of generation and the rate of decay due to electron-LO phonon scattering, respectively. τ_p is the LO phonon lifetime while $1/\tau_k$ is the rate of decay of an electron with wave vector k due to any process not involving the LO phonons. For electrons not energetic enough to undergo intervalley scattering τ_k is >> the electron-LO phonon scattering time so that f_k/τ_k can be neglected in Eq. (1). The broken curve in Fig. 2 was obtained by numerically integrating Eqs. (1) and (2) assuming parabolic conduction and valence bands and neglecting intervalley scatterings. The excitation pulse is assumed to be a δ-function in time. Since the density of photoexcited electrons is not known exactly it has been adjusted to fit the experiment at $\hbar\omega_I \sim$ 1.95eV. The electron density we obtained is $\sim 3 \times 10^{16}$ cm^{-3} which was roughly what we estimated from our laser intensity. We should point out that we have kept the power density on the sample low enough so that carrier-carrier interaction is not important.[8-10] At higher power densities the Raman spectra are complicated by the presence of single-particle scattering and screening of LO phonons by the photoexcited carriers.

The agreement between the experimental points and the above simple model is satisfactory except for $\hbar\omega_I \geq 2.1$eV. *The discrepancy is expected* since we have neglected the intervalley scatterings and the conduction band nonparabolicity. *For electrons energetic enough to scatter* into another valley τ_k^{-1} is equal to the inter*valley scattering rate. These rates for the* Γ-L and Γ-X intervalley scattering have been calculated assuming an average phonon energy $\simeq$ 0.8 x the zone center LO phonon energy and average deformation potentials $D_{\Gamma L}$ and $D_{\Gamma X}$ respectively.[11] The photon energies corresponding to the onset of intervalley scatterings depend on the band structure of GaAs. For the separations of the Γ, X and L valleys we have used the electroreflectance result of Aspnes.[12] The nonparabolic Γ electron band energy has been calculated by the Kane model.[13] The warped heavy hole band is assumed to be parabolic with a spherically averaged effective mass. The resultant theoretical curve (solid curve) is obtained by adjusting $D_{\Gamma L}$ plus an overall scaling factor. Since we did not observe a decrease in N_q at the onset of the Γ-L intervalley scattering at ~ 1.89eV we can only set an upper limit on $D_{\Gamma L}$ of $\sim 1.4 \times 10^8$eV/cm. For $D_{\Gamma X}$ we have used the value of 1.1×10^9eV/cm obtained by Vinson et al.[14] from Gunn effect measurements on GaAs. Although our upper bound on $D_{\Gamma L}$ is about a factor of 2 smaller than the value deduced by Vinson et al. it is consistent with recent experiments on optically pumped GaAs lasers by Koch et al.[15]

There is a small discrepancy of ~ 15 meV between theory and experiment regarding the onset of Γ-X intervalley scattering. Since the uncertainty in the separation of the Γ and X conduction minima is only ~ 5meV, we attribute this discrepancy to our approximation in using an average effective mass for the highly anisotropic heavy hole band. The sensitivity of N_q to the band structure is also shown by the better agreement between the nonparabolic electron band theory (solid curve in Fig. 2) and experiment for $\hbar\omega_I < 2.1$eV. Detailed discussions of our calculation will be presented elsewhere.

In conclusion we have measured the hot LO phonon population in GaAs as a function of the excitation photon energy. Our experimental results are in good agreement with a simple model calculation which includes both intravalley and intervalley relaxation of the photoexcited electrons. We have shown that this kind of 'hot phonon spectroscopy' can provide information on the band structure and electron-phonon interactions in semiconductors.

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