PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON THE PHYSICS OF SEMICONDUCTORS, KYOTO, 1966
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 21, SUPPLEMENT, 1966

XI-9. Behavior of Phonons in Cadmium Sulfide

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The decaying features of excited phonon flux in CdS were observed using pairs of pulses: exciters and tracers. Some physical properties of the lowest nonlinear loss term in the rate equation for phonons were investigated as a function of carrier density and temperature. A preliminary calculation of the damping coefficient of a phonon based upon the phonon-electron interaction was made also.

§ 1. Introduction

Current saturation and ultrasonic amplification phenomena in various piezoelectric materials have been the subjects of many works. It has been proposed by Hutson[1], that the acoustic flux, accompanying current saturation, is limited in steady state by a nonlinear loss mechanism involving the interaction with carriers. Yamashita and Nakamura[2], based upon phonon picture, have shown that this nonlinear process can not stem from simple three phonon processes, and Mikoshiba[3] calculated the three phonon processes assisted by conduction electrons. McFee[4] has investigated the ultrasonic buildup accompanying current saturation, but actually the ultrasonic waves lie at so high frequencies that his observations are short of any direct informations concerning the kinetics of "active phonons." This paper reports that we observed the decaying features of the active phonons, using double pulse technique. This means, the first pulse makes the phonon flux build up, and the second one traces the active phonon states, if we let the interval of the two pulses vary. In this case, only the initial current intensity of the second pulse is concerned. This method has a merit that the observed quantities are not disturbed by the oscillatory behavior of the current.

§ 2. Experiments

A vapor-phase grown, photoconducting CdS crystal ($\rho \sim 10^6 \Omega$.cm in the dark) was cut to the dimensions $5 \times 5 \times 0.5$ mm^3. Electric contact of 1 mm^2 in area was made at the center of the crystal plate with indium. Electric field was parallel to the crystal *c*-axis. Carrier density was controlled by illuminating the sample with a tungsten lamp equipped with combined filters which allow wavelengths between 540–750 mμ to pass. Thus uniform carrier generation was performed. Infrared part of the spectrum was cut off to avoid sample heating. Various light levels were obtained using neutral filters.

Our observations were made by applying pairs of voltage pulses of 4 μsec in duration and 50 cps in repetition rate. The interval of a pair could be chosen arbitrary. Figure 1 (a) shows an oscilloscope trace of the current, when the applied field exceeds the threshold for the current saturation. The peak of the second current pulse stands lower than that of the first pulse. This is because of extra scattering of conduction electrons by "excess phonons," which have been excited by the first pulse and are decaying during the interval of the pulses. The peaks of both current pulses would reach an equal Ohmic value, if sufficient time interval were allowed.

Photograph, Fig. 1(b), was taken by continuously changing the interval between a pair of pulses. The locus of the peaks of the second pulses is very similar to the decay curve of, so to say, photoconductivity. This curve shows the change in the number of scatterers against electrons, as a function of time. By a proper transformation, we can obtain decay curves of the excess phonons, which have been excited through acoustoelectric effect.

§ 3. Method of Analysis

Consider a system consisting of conduction electrons, ordinary phonons (optical and acoustic) and excess phonons. In the absence of the excess phonons the current is described well by a relaxation time τ_0, which is of the order of 10^{-13} sec or less. This relaxation is mainly due to optical phonons, at room temperature. We assume that the scattering due to the excess phonons is also described by a single relaxation time τ_e. This is the case, when the excess phonon band is narrow enough in comparison with the thermal phonon distribution. Then

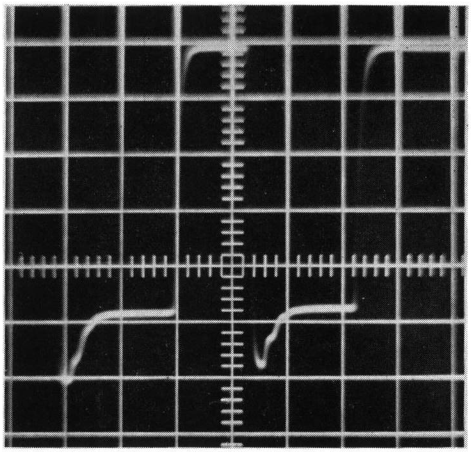

Fig. 1(a). Oscilloscope trace of a pair of current pulses. Time runs from left to right. The second pulse is applied 3 μsec later than the first one.

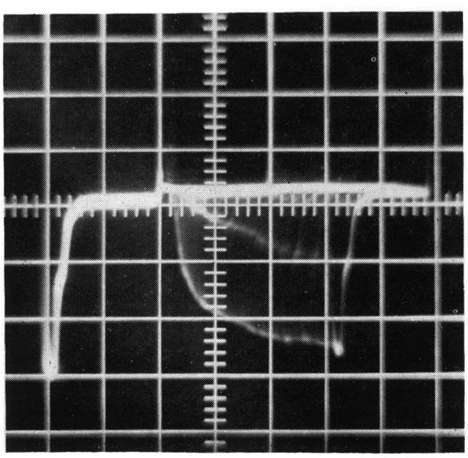

Fig. 1(b). Multiplely exposed photograph of the above double pulse oscilloscope traces. The interval was changed upto about 6 μsec manually.

the time dependence of the current is introduced only through τ_e. τ_e varies along with the number of the excess phonons of wave number $\boldsymbol{q}$, $N_{\boldsymbol{q}}^e$, as

$$1/\tau_e = \kappa N_{\boldsymbol{q}}^e \tag{1}$$

with a suitable time independent factor κ. κ must be the product of the collision cross-section and the mean velocity of carriers. Since

$$1/j \propto 1/\tau = 1/\tau_0 + \kappa N_{\boldsymbol{q}}^e\,, \tag{2}$$

we obtain the desired relations between the number of the excess phonons and the current density j, as

$$\kappa N_{\boldsymbol{q}}^e = 1/\tau - 1/\tau_0 = \frac{ne^2E}{mj_0}\cdot\frac{j_0 - j}{j}\,, \tag{3}$$

where n is the carrier density, m the effective mass of the carrier, E the applied electric field and j_0 the Ohmic current.

Let's introduce the maximum excess phonon number $N^e{}_{\text{max}}$. This is equivalent to $N_{\boldsymbol{q}}^e(\infty)$ under the condition of ultrasonic amplification, and corresponds to the saturation current j_{sat}, as

$$\kappa N^e{}_{\text{max}} = \frac{ne^2E}{mj_0}\cdot\frac{j_0 - j_{\text{sat}}}{j_{\text{sat}}}\,. \tag{4}$$

Finally from (3) and (4) we have an equation

$$\frac{N^e{}_{\text{max}}}{N_{\boldsymbol{q}}^e} = \frac{j(j_0 - j_{\text{sat}})}{j_{\text{sat}}(j_0 - j)}\,. \tag{5}$$

Equation (5) describes the behaviors of the phonon flux along time, connected with the non-Ohmic current, not only in case the current saturates (phonon flux builds up), but also the peak of the second pulse recovers toward the Ohmic value (phonon flux decays).

§ 4. Results

Decay processes of particles are usually analized by means of rate equations. As is well known decay curves of monomolecular processes are exponential, whereas those of bimolecular, hyperbolic, with respect to time.

Figure 2 shows that the decay curve of the excess phonons is almost hyperbolic. It is also found that for the first one or two microseconds

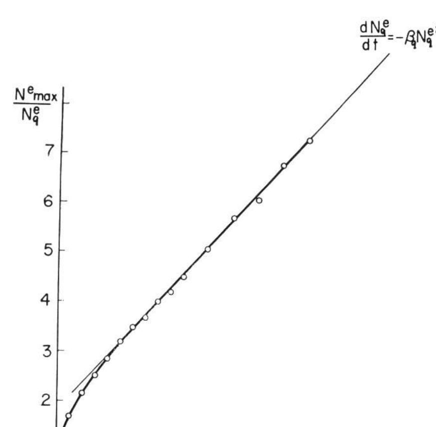

Fig. 2. Observed $N^e_{\text{max}}/N^e_{\boldsymbol{q}}(t)$ versus t curve. The solution of eq. (6), $N^e_{\text{max}}/N^e_{\boldsymbol{q}}(t) - 1 = \beta_{\boldsymbol{q}} N^e_{\text{max}}\, t$ is compared with the observations. The initial condition here is taken to be at the end of the first pulse.

the decay curve is very neatly described as trimolecular process. No doubt monomolecular process will be dominant, when the excess phonon density is more lessened. We propose, therefore, a rate equation for the excess phonons:

$$\frac{dN_q^e}{dt} = -\alpha_q N_q^e - \beta_q (N_q^e)^2 - \gamma_q (N_q^e)^3 - \cdots, \quad (6)$$

where α_q, β_q and γ_q are numerical constants, if the phonon band is monochromatic (otherwise, they should be proper integral operators.) Our main concern here is on the second term. This is the lowest nonlinear term in the decay process.

From the slope of the linear portion on the curve in Fig. 2, $\beta_q N^e{}_{max}$ can be estimated using eq. (5). Relative value of $N^e{}_{max}$ is obtained from the ratio of saturation current to Ohmic current. Thus relative value of β_q is readily estimated. Figure 3 shows the carrier density dependence of $\beta_q N^e{}_{max}$, $N^e{}_{max}$ and β_q.

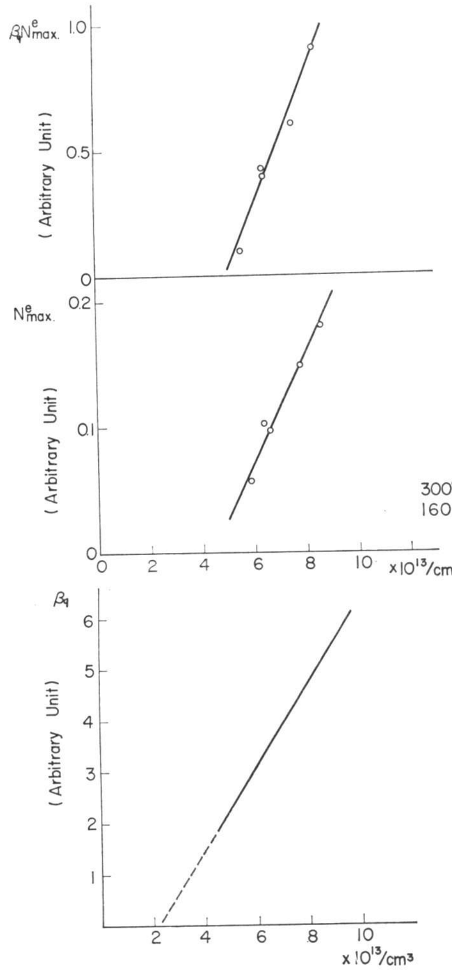

Fig. 3. Observed carrier density dependence of decay constant β_q. Experiments were performed at room temperature. Carrier density n was estimated from the Ohmic conductivity, assuming $\mu = 260\,\text{cm}^2/\text{V sec}$.

The most remarkable feature is carrier density dependence of β_q, since β_q would be insensitive to n, provided this bimolecular process is not any three phonon process involving conduction electrons, as discussed by Hutson.[1)]

§ 5. Theoretical Considerations and Discussions

(1) With respect to the nonlinear terms appearing in the phonon Boltzmann equation, we have had a dilemma: though Abe[5)] calculated the voltage dependent current beyond the threshold voltage, the effects due to the three phonon processes among excess phonons and ordinary phonons were too small to explain the dependence. On the other hand, the same processes among the excess phonons themselves do not satisfy the energy and the momentum conservation laws simultaneously.[2)] We should look for other mechanism. Upon phonon picture the electron-phonon interaction to 2nd order causes the phonon system to become unstable. However, the same interaction to higher orders does not necessarily have negative damping effects. In fact we calculated the most dominant damping coefficient of a phonon of (q, $z = \cos(\boldsymbol{q}, \boldsymbol{v}_d)$) to 4th order and obtained

$$-\int \beta_{qq'} N_{q'} d\boldsymbol{q}' = -\frac{5}{2\pi^3}\frac{1}{z^2}\sqrt{\frac{k_B T}{2\pi m v_s^2}} \cdot \frac{m}{\hbar^2 q^2} E_q \frac{1}{n} \times \int_0^{\sim m v_s/\hbar} E_{q'} N_{q'}^{(1)} \log\frac{m v_s (1+3z)^2}{\hbar_{q'} z} \cdot q'^2 dq' \quad (7)$$

where $E_q = K^2 \hbar\omega_q q_D^2/\varepsilon_q^2 q^2$, $\omega_q = q v_s$ and $\varepsilon_q = 1 + q_D^2/q^2$ in terms of the electromechanical coupling constant K^2, and $N_{q'}(Z) = \sum_{l=0}^{\infty} N_{q'}^{(2l+1)} P_{2l+1}(Z)$ with $Z = \cos(\boldsymbol{q}', \boldsymbol{v}_d)$. Here $P_{2l+1}(Z)$ are Legendre functions and $N_{q'}^{(1)} \gg N_{q'}^{(3)}, N_{q'}^{(5)}, \cdots\cdots$ was assumed. From eq. (7) and the well-known α_q,

$$\left|\int \beta_{qq'} N_{q'} d\boldsymbol{q}' \Big/ \alpha_q \right| \cong \frac{N_q^{(0)}}{n}\int_0^{\sim m v_s/\hbar} E_{q'} \frac{N_{q'}^{(1)}}{\hbar\omega_{q'}} \log\left(\frac{m v_s}{\hbar q'}\right) \times q'^2 dq', \quad (8)$$

with $N_q^{(0)} = k_B T/\hbar\omega_q$, is obtained except for some numerical factor.

(2) We estimate eq. (8) assuming that $N_q^{(1)}$ is narrow enough and the integral is replaced by $E_q N_q^{(1)} q^3 \log(m v_s/\hbar q)/\Omega$ with a suitable volume in the reciprocal space $1/\Omega$. Then the ratio (8)

becomes of the order of unity, if we take $q = q_D \cong 10^5\,\mathrm{cm}^{-1}$ for $n = 10^{14}\,\mathrm{cm}^{-3}$ and $N_q^{(1)}/N_q^{(0)} n\Omega \cong 10^{-4}$. Thus this damping effect no doubt seems intense enough to suppress the buildup of phonon flux. We are trying to make estimate of the effect without using pertubation.

(3) Thus obtained β_q is proportional to carrier density n so long as $q^2 \gg q_D^2$. But if we take the condition of the maximum gain to be $q = q_D \propto n^{1/2} T^{-1/2}$, then β_q should be proportional rather to $n^{1/2}\ T^{-2}$. Experimentally β_q is linear in n in the range of $2 \sim 10 \times 10^{13}\,\mathrm{cm}^{-3}$ and decreases with temperature for $T = 77° \sim 300°\mathrm{K}$. In this respect more data on n-dependence should be required to compare with the theory.

(4) In our experiments β_q is quite insensitive to applied voltage, whereas $N^e{}_{\max}$ increases with it. Also $N^e{}_{\max}$ is proportional to $n^a T^b$ with $a \gtrsim 1$ and $b \cong 2.3$.

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