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Propagation of Nonequilibrium Phonons with Frequency Down-conversion

Anthracene

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A theory is constructed describing the propagation of nonequilibrium phonons under conditions when propagation of phonons proceeds simultaneously with their spontaneous decay into phonons with lower frequencies.

1. INTRODUCTION

The experiments carried out by V. L. Broude and his collaborators on nonequilibrium luminescence of anthracene[1–3] stimulated the development of a theory presenting a new viewpoint on the propagation of nonequilibrium phonons in crystals. To our profound belief this point of view based on the idea of phonon generations should be very important in the interpretation of a whole number of experiments associated with highly excited states of dielectrics and semiconductors.

The present paper is the account of the semi-quantitative theory of the nonequilibrium phonon propagation. Such a level of theory is quite sufficient for developing a complete picture of the phenomena and for estimating the relevant parameters.

To illustrate more clearly in what way the experiments of V. L. Broude and his collaborators stimulated to develop the above-mentioned theory we shall describe in short most eminent results of these experiments, without touching the experimental procedures, which are often very much complicated.

2. EXPERIMENT

A thin antracene crystal (of $d = 10 - 50\ \mu$m in thickness) is placed in a liquid helium bath. One crystal surface (the front one) is illuminated with a powerful laser pumping pulse; the pulse duration is $t_P = 10$ ns and the pulse intensity is $I = (10 \div 100)\ \text{kW/cm}^2$; the diameter of the spot is about 0.5 mm. On the other crystal surface (the rear one) a weak probing pulse of the same duration delayed relative to the pumping pulse excites luminescence which is detected; the delay time t_D varies from 0 to 60 ns, the diameter of the probing beam is the same as that of the pumping beam. If required, the probing beam can be directed to the front surface of the crystal as well.

The following characteristics of the luminescence spectrum have been measured:

1) the width $\Delta\nu$ of the vibronic band 23692 cm^{-1}, located 1400 cm^{-1} lower than the exciton absorption edge;

2) the ratio R of the intensities of two lines: 25036 cm^{-1} and 25051 cm^{-1}, the components of the triplet located just below the exciton absorption band.

The nonequilibrium state of the crystal region where the probing beam is directed and from which luminescence arises is characterized by the values $\Delta\nu$ and R distinguished from those values $(\Delta\nu)_0$ and R_0 which are typical of the equilibrium luminescence and which are observed when the pumping pulse is blocked. While measuring the dependence of $\Delta\nu$ and R on the probing pulse delay, it is possible to monitor the time dynamics of the nonequilibrity development at the place of probing. In case the rear surface is probed, the place where nonequilibrity is excited and the place where it is probed are separated in space. This makes the experiment space-resolved and allows one to study propagation of nonequilibrity in space.

Let us now turn to the description of the results of the experiments which turned out to be the most important for stimulating the development of the theory of nonequilibrium phonon propagation.

Figure 1 shows the dependence of R at the rear surface of the crystal on t_D for a crystal with $d = 45\ \mu$m at the pumping $I = 40\ \text{kW/cm}^2$ and the bath temperature $T = 5$ K. So far as $t_D < t_1 = 15$ ns, the R value does not differ from R_0, i.e. the luminescence is coming from an equilibrium state. When the delay is larger, nonequilibrity arises, since $R > R_0$. Even though the duration of the probing pulse and of the pumping pulse is rather large, as is seen from the same figure, the steps at $t_2 = 23$ ns and $t_3 = 31$ ns are clearly seen.

The results of measurements of $\Delta\nu$ are shown in Figure 2, for a thinner crystal $d = 12\ \mu$m at $I = 30\ \text{kW/cm}^2$ and $T = 5$ K. Both at the front and the rear surfaces oscillations of $\Delta\nu$ with the period $\Delta t \simeq 20$ ns can be distinctly seen. The mean levels, in the vicinity of which oscillations occur, depend on T

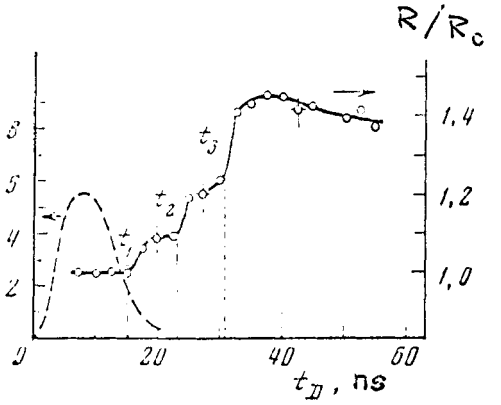

FIGURE 1 Dependence of the ratio R on the delay time t_D. The pumping pulse is shown by a dotted line.

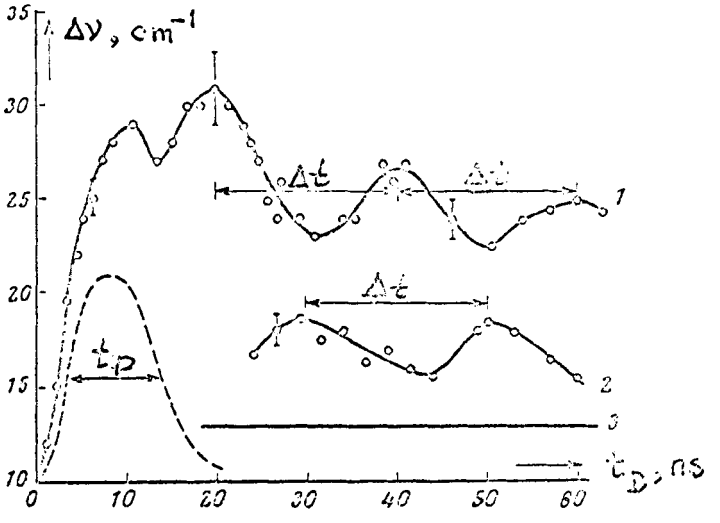

FIGURE 2 Oscillations of the width $\Delta\nu$ with respect to the delay time at the probing of the front surface are shown by curve 1, and at the probing of the rear surface by curve 2. The mean oscillation level on both surfaces with weak pumping 9 kW/cm^2 is straight line 3. The dotted line shows the pumping pulse.

and I. At low temperatures T and weak pumpings I these levels on both surfaces practically coincide. With increasing T and/or I the levels move apart and the level at the front surface becomes higher than that on the rear surface. The oscillation amplitude decreases at the same time. At $I \simeq 100\,\text{kW/cm}^2$ or $T \simeq 20\,\text{K}$ for delay times $t_D < 60\,\text{ns}$ no nonequilibrity at the rear surface is observed. Meanwhile at the front surface the nonequilibrity is very strong (Figure 3).

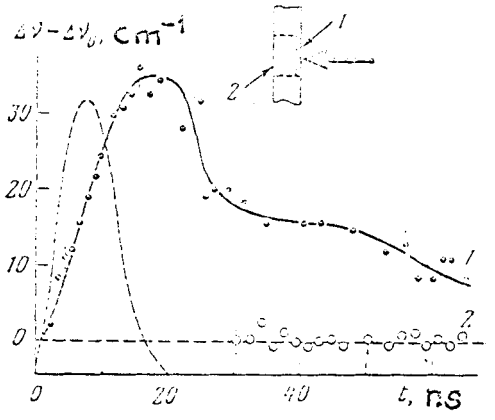

FIGURE 3 Absence of oscillations $\Delta\nu$ and localization of nonequilibrity in the vicinity of the front surface. The probing of the front surface—curve 1, of the rear surface—straight line 2. The pumping pulse is indicated.

3. NONEQUILIBRIUM PHONONS AND PHONON SCATTERING

The characteristic times observed in the experiment of the order of 10 ns are close to d/s, the acoustic phonon time of flight from one crystal surface to the other (the sound velocity in antracene is $s = (1 \div 4) \cdot 10^5$ cm/s). Therefore an assumption arises that nonequilibrity transfers from the front surface of the crystal to the rear one by nonequilibrium phonons.

The absorption of the pumping light leads to production of nonequilibrium excitons in the absorption layer $\kappa^{-1} = 0.5\ \mu$m; the exciton lifetime is about 0.5 ns. For this time a greater part of the absorbed energy transfers from the electron system into the phonon one. In such a situation high-frequency optical phonons (intramolecular oscillations) are produced first. Then the high-frequency optical phonons are converted into low-frequency optical ones, and at the end there arise acoustical phonons with frequencies of the order of the Debye frequency ω_D. All the process of relaxation up to the acoustical phonon appearance is localized in the absorption layer, since the group velocities of excitons and of optical phonons are small. Besides, this process proceeds very fastly—the characteristic time of the optical phonon decay is of the order $10^{-12} - 10^{-11}$ s; so that even taking account of the great number of down-conversion acts, it is unlikely that more than 10^{-10} s passes until the first acoustical phonons are produced.

Production of acoustical phonons means that excitations with an appreciable group velocity of the order of the sound velocity arise and non-

equilibrity may go now beyond the absorption layer. According to the results of the experiments nonequilibrity achieves the rear crystal surface, for the time of the order d/s; this means that phonons fly ballistically without scattering, i.e., the phonon free path is $l > d$. Whereas the free path of acoustical phonons with the frequency of the order of the Debye one is small, of the order of 10^{-6} cm; so that in reality it turns out that $l < d$. If one follows the common point of view, the last inequality means that phonons propagate diffusively and so the time of nonequilibrity propagation is not d/s, but d^2/sl, which is in evident disagreement with the experiment. Besides, it is clear in the case of diffusive propagation we are unlikely to obtain oscillations.

It is the attempt to solve this contradiction that resulted in the development of a new picture of the nonequilibrium phonon propagation.

First of all it should be recalled which scattering processes define the character of phonon propagation. All the scattering processes are divided into normal (N-processes) and resistive (R-processes) ones. The normal processes are phonon–phonon interaction without umklapp. The main property of N-processes is the conservation of the total momentum and of the total energy of the system. Resistive processes include phonon–phonon interaction with umklapp (U-processes) and phonon scattering by crystal defects, in particular, by isotopic atoms. R-processes conserve the energy, but not the momentum. It is essential that the probabilities of all the scattering processes decrease fastly with decreasing the phonon frequency: for N-process it is as ω^5, for U-processes as $\exp\{-\omega_D/\omega\}$, for scattering on static defects as ω^4.

According to the common point of view the comparison of free paths l_N and l_R one with another and with d, the length of propagation, defines the regime of the nonequilibrium phonon propagation—ballistic ($d < l_N, l_R$), diffusive, i.e., heat conduction ($d > l_N, l_R$), or hydrodynamic one, i.e., the second sound ($l_R > d > l_N$).

The new property of the nonequilibrium phonon propagation is based on the concept that if $\hbar\omega > kT$, then simultaneously with the phonon propagation in space there occurs frequency down-conversion and the phonon spectrum is continuously shifted to lower frequencies. In this case all the free paths fastly change and the process of propagation is continuously rearranged. As a result, new regimes of propagation appear, which cannot be identified with the known ones.

4. PHONON GENERATIONS

Let us first evaluate the occupation numbers of the first acoustical phonons with the frequency of the order of the Debye one. The lifetime of these phonons τ_0 is shorter than the duration of the pumping pulse, and therefore

the pumping may be considered as stationary. Assuming for simplicity that all these phonons have one and the same energy $\hbar\omega_0$, their occupation numbers n_0 can be found from the energy balance

$$\dot{Q} = n_0 \hbar\omega_0 / a_0^3 \tau_0, \tag{4.1}$$

where a_0^3 is the volume of the elementary cell, $\dot{Q} = \kappa I$ is the power absorbed in 1 cm^3. For antracene $\omega_D = 30\ \text{cm}^{-1}$, $a_0^3 = 0.5 \cdot 10^{-21}\ \text{cm}^3$. By analogy with other crystals we assume that $l_0 = s\tau_0 \simeq 100\, a_0 \simeq 0.1\ \mu\text{m}$. Then $n_0 = 1$ at $I = 1400\ \text{kW/cm}^2$. Thus, at all the pumpings employed $n_0 < 1$.

When the phonon energy is high: $\hbar\omega > kT$, and the occupation numbers are small: $n(\omega) < 1$, the dominating N-process is the spontaneous decay into two phonons: $\omega \to \omega' + \omega''$. In a simple model the rate of such a decay is[4]

$$\frac{1}{\tau_N(\omega)} = \frac{1}{\tau_N^0}\left(\frac{\omega}{\omega_0}\right)^5, \tag{4.2}$$

i.e., the decay rate fastly decreases with decreasing the phonon frequency. The frequencies of the phonons ω' and ω'' are of the same order, and for simplicity they may be considered to be equal: $\omega' = \omega'' = \frac{1}{2}\omega$.

Then we may say that the phonons ω_0 (the first generation) decay into phonons $\omega_0/2$ (second generation), they in their turn decay into phonons $\omega_0/2^2$ (third generation) and so on. The generation ω transforms into the generation $\omega/2$ in the time $\tau_N(\omega)$. Since $\tau_N(\omega)$ fastly increases with the change of generations, then at the moment t only the phonons for which $\tau_N(\omega) = t$ survive, i.e., phonons with the frequency†

$$\omega = \omega_0\left(\frac{t}{\tau_N^0}\right)^{-1/5}. \tag{4.3}$$

Process of decay will continue until the occupation numbers $n(\omega)$ become of the order of unity. When this occurs, the N-processes of phonon coalescence will compete with the N-processes of phonon decay: $\omega' + \omega'' \to \omega$ and the down-conversion stops. The simultaneous action of decay and coalescence will lead to the Plank distribution. The temperature of this distribution is $k\tilde{T} = \hbar\tilde{\omega}$, where $\tilde{\omega}$ is the frequency of the generation for which $n(\tilde{\omega}) = 1$.

Such a termination of the down-conversion takes place, provided $\hbar\tilde{\omega} > kT$, i.e., when the down-conversion stops at the level of phonon frequencies $\tilde{\omega}$ much higher than that of the equilibrium phonons $\omega_T = kT/\hbar$. This situation apparently takes place at a sufficiently strong pumping and sufficiently low bath temperature. In case the pumping is weak or the bath temperature is high, the down-conversion terminates at the level of equilibrium phonon frequencies ω_T and the nonequilibrity is totally "absorbed" by the bath.

† Here and below the sign = means equality by the order of magnitude. and the signs > and < mean strong inequalities.

Hence, it follows that the time of establishing the complete equilibrium is $\tau_N(\omega_T)$, irrespective of the frequency of the initial nonequilibrium phonons. At helium temperatures this time is above microseconds.

5. QUASIBALLISTIC REGIME

Let us assume that there are no other scattering processes aside from spontaneous decay. Then the phonon generations with some frequency ω for the lifetime of this generation shifts in space by the distance of the order of the free path with respect to the decay: $l_N(\omega) = s\tau_N(\omega)$. The first generation phonons occupy a layer of the thickness $l_N^0 = l_N(\omega_0)$; the second generation phonons occupy a layer of much greater thickness $l_N(\omega_0/2)$, and so on (Figure 4). While the generations change the length $l_N(\omega)$ fastly increases, and it becomes clear that the rear surface of the crystal is achieved by those phonons whose frequency ω satisfies the equality $l_N(\omega) = d$, i.e., they are the phonons with the frequency†

$$\omega = \omega_0 \left(\frac{d}{l_N^0} \right)^{-1/5} \equiv \bar{\omega}. \tag{5.1}$$

Using (4.3) we may see that this will occur by the moment $t = d/s \equiv t_B$. And we may call such a regime of propagation as "quasiballistic" one.

Therefore, though the free path of the initial acoustical phonons ω_0 is very small, this does not mean that the phonon propagation regime is diffusion. A sharp increase in the free path during the decay leads to the fact that the velocity of the phonon nonequilibrity propagation turns out to be the same as if the phonons would move ballistically.

The quasiballistic regime of propagation makes it possible to explain the fast propagation of phonon nonequilibrity in the experiments described above and the oscillations of nonequilibrity. However, a question arises why these effects vanish with an increase in the bath temperature and/or in pumping.

The quasiballistic regime exists until the occupation numbers are small: $n(\omega) < 1$, and the frequencies are above the thermal ones: $\omega > \omega_T$. An increase in the bath temperature T may lead to the case when ω becomes comparable with ω_T before the phonons reach the rear surface. An increase in pumping I increases the occupation numbers and $n(\omega)$ may become equal to unity before the phonons reach the rear surface. An increase in T and I may thus distort the quasiballistic regime.

Naturally, there arise two questions: when does this occur and how will the regime of propagation be rearranged? The limitation imposed upon the

bath temperature may be written immediately. It is $kT < \hbar\bar{\omega}$, or

$$kT < \hbar\omega_0 \left(\frac{d}{l_N^0}\right)^{-1/5} \tag{5.2}$$

Taking $d = 12\ \mu$m and $l_N^0 = 0.1\ \mu$m we find the limitation: $T < 16$ K, which seems quite reasonable in the sense of the experimental results. In order to obtain the limitation imposed upon the pumping one should find the phonon occupation numbers.

6. PHONON OCCUPATION NUMBERS

Let us estimate the occupation numbers in a quasiballistic regime, first for a semiinfinite crystal. Let us write the energy density of the generation ω:

$$\varepsilon(\omega) = n(\omega)\hbar(\omega)\left(\frac{\omega}{s}\right)^3 = n(\omega)\frac{\hbar\omega_0}{a_0^3}\left(\frac{\omega}{\omega_0}\right)^4. \tag{6.1}$$

If $\tau(\omega) > t_P$, then by the moment when the generation ω exists, all the pumping energy is already contained in the sample and is turned to the energy of this generation. Since the generation occupies a layer in the crystal of the thickness $l_N(\omega)$, then

$$\varepsilon(\omega)l_N(\omega) = \int_0^\infty dt I(t) \equiv P, \tag{6.2}$$

where P is the total energy absorbed by 1 cm^2 of the surface. From (6.1) and (6.2) we have:

$$n(\omega) = \frac{P}{I_0\tau_0}\left(\frac{\omega}{\omega_0}\right), \qquad I_0 \equiv \frac{\hbar\omega_0}{a_0^3}s, \tag{6.3}$$

i.e., for the generations arising after the termination of the pump the occupation numbers decrease with changing the generations.

Suppose now $\tau(\omega) < t_P$. Then the pumping may be considered to be stationary and $n(\omega)$ can be found from the condition of constant phonon energy flux. The flux should be equal for all the generations and equals the light flux incident upon the sample:

$$\varepsilon(\omega)s = I. \tag{6.4}$$

From (6.1) and (6.4) we have

$$n(\omega) = \frac{I}{I_0}\left(\frac{\omega}{\omega_0}\right)^{-4}, \tag{6.5}$$

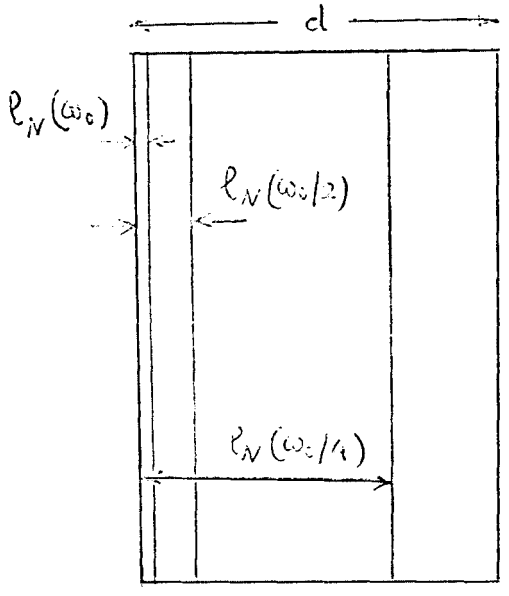

FIGURE 4 Arrangement of phonon generations in space.

i.e., for the generations arising before termination of the pump the occupation numbers increase with changing generations.

So, first when the pumping is in process, the occupation numbers increase, and then after its termination they decrease (Figure 5). The maximum occupation numbers exist by the moment $t = t_P$, when (6.3) is equal with (6.5).

Thus,

$$\max n(\omega) = \frac{I}{I_0}\left(\frac{t_P}{\tau_N^0}\right)^{4/5}. \tag{6.6}$$

While this value is small, in a semiinfinite crystal the quasiballistic regime is realized.

In a crystal plate the $n(\omega)$ calculation is valid for the generations for which $l_N(\omega) < d$, i.e. until the rear surface is not achieved. The generations with the free path $l_N(\omega) > d$, existing at $t > t_B$, differ from the generations for which $l_N(\omega) < d$, since in the crystal they occupy a layer of the thickness d, and not of $l_N(\omega)$. Therefore, for such generations we have

$$\varepsilon(\omega)d = \int_0^{\tau(\omega)} dt I(t). \tag{6.7}$$

If the pumping pulse is short, i.e., $t_P < t_B$, then the generations occupying the whole crystal exist only after termination of the pump. Then the integral in

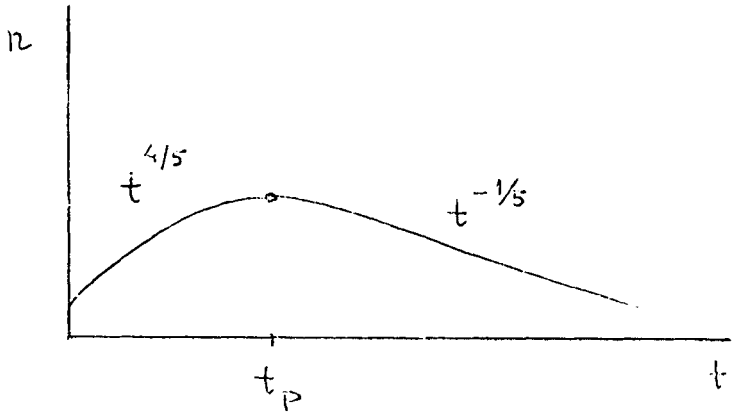

FIGURE 5 Variation of the occupation numbers with time for a semi-infinite crystal. Time dependences are obtained from the frequency ones with the substitution (4.3).

(6.7) is equal to P and

$$n(\omega) = \frac{P}{I_0 t_B}\left(\frac{\omega}{\omega_0}\right)^{-4}. \tag{6.8}$$

This means that at $t = t_B$ the decrease of $n(\omega)$ according to (6.3), is replaced by an increase according to (6.8), see Figure 6.

If the pumping pulse is long, i.e., $t_P > t_B$, then the generations occupying the whole crystal arise before the termination of the pumping pulse. For these generations the integral in (6.7) is equal to $I\tau(\omega)$ and we have

$$n(\omega) = \frac{I}{I_0}\frac{\tau_0}{t_B}\left(\frac{\omega}{\omega_0}\right)^{-9}. \tag{6.9}$$

In this case the maximum (6.6) practically does not arise, since the increase of $n(\omega)$ according to (6.5) at $t = t_B$ is changed by a still faster increase of $n(\omega)$ according to (6.9). For the generations occupying the whole crystal and for those existing after the termination of the pump, the integral in (6.7) is equal to P and we again receive (6.8). Thus, at $t = t_P$ the fast increase according to (6.9) is changed by a slower one, according to (6.8), see Figure 6.

Now it is possible to obtain the limitation for the pumping, resulting from the inequality $n(\bar{\omega}) < 1$ for all $\omega > \bar{\omega}$. From Figure 6 it is clear that if the pumping pulse is short, then this requirement is reduced to max $n(\omega) < 1$, i.e., to

$$I < I_0\left(\frac{\tau_N^0}{t_P}\right)^{4/5}, \qquad (t_P < t_B). \tag{6.10}$$

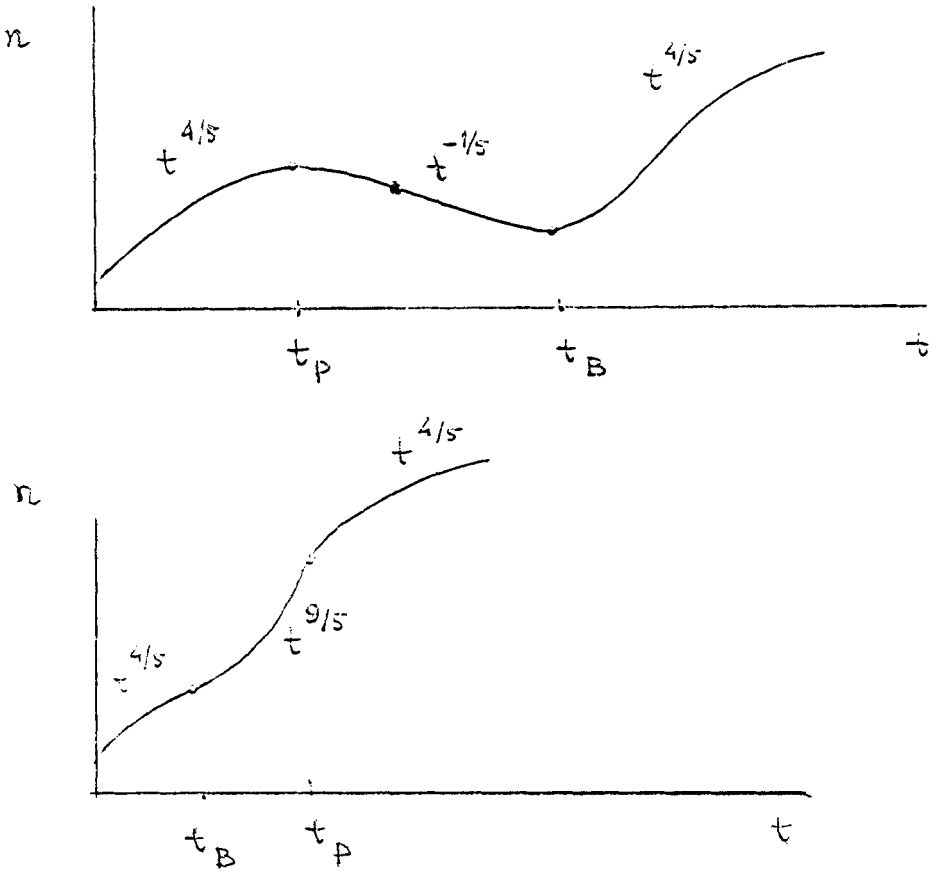

FIGURE 6 Variation of occupation numbers with time in a plate of a finite thickness. The upper figure illustrates a thick crystal ($t_B > t_P$); the lower figure shows a thin crystal ($t_B < t_P$).

If the pulse is long, then from Figure 6 it is seen that the requirement is reduced to $n(\bar{\omega}) < 1$, which after using (6.5) and (5.1) yields

$$I < I_0 \left(\frac{l_N^0}{d}\right)^{4/5}, \qquad (t_P > t_B). \tag{6.11}$$

Let us estimate the corresponding parameters for the experimental situation. Assuming $s = 1.10^5$ cm/s (in order to obtain the observed semi-period of Δv-oscillations) we yield $I_0 = 120\,\mathrm{kW/cm^2}$ and $t_B = 12$ ns. This t_B is of the same order as t_P, so that we may equally use both (6.10) and (6.11), which provides limitation to the power: $I < 3\,\mathrm{kW/cm^2}$. These estimates are certainly very rough, first due to a simplified model, and secondly, due to arbitrariness of the assumed l_N^0. In case to consider them seriously, we admit

that limitation on the power is not fulfilled. Why are then the ballistic times of the order of d/s still observed?

7. HYDRODYNAMIC REGIME

If the limitation on the power (6.10) or (6.11) is not fulfilled there exists a certain generation $\tilde{\omega}$, for which $n(\tilde{\omega}) = 1$. As has been already noted in Section 4, the phonons of this generation will not undergo a further decay, but will be "thermalized." This means that the Plank phonon distribution takes place with the temperature $k\tilde{T} = \hbar\tilde{\omega} > kT$. The phonons of this distribution differ from the thermal ones by the fact that they are hot; and besides, the distribution as a whole drifts with some velocity u. Further propagation of phonon nonequilibrity is of a hydrodynamic character and occurs with the velocity u.

The value of the velocity u can be estimated using the fact that in N-processes the ratio of the total phonon momentum to the total energy is conserved, i.e., the anisotropy of the phonon distribution is conserved. The first generation phonons fly only at one direction, from the front surface, and have, therefore, a total momentum oriented toward the rear surface, the total momentum energy ratio being of the order $1/s$. For the shifted Plank distribution this ratio is u/s^2. Hence, it follows that $u = s$, and therefore it is clear that the phonon nonequilibrity propagation is characterized by the times of the order t_B.

Let us consider the case when the limitation on temperature (5.2) is not fulfilled, but the limitation on power holds. In this case for a generation which has not yet reached the rear surface, the frequency is equal to the thermal one: $\omega = \omega_T$ and the occupation numbers for this generation are still small: $n(\omega) < 1$. This generation will interact with thermal phonons and thermalize with them. A shifted Plank distribution arises again, but now its temperature will only slightly differ from T, and the drift velocity will be small: $u < s$. This follows from the fact that the nonequilibrium phonons whose occupation numbers are small introduce little energy and momentum into the thermal bath and, therefore, weakly perturb the equilibrium Plank distribution. However, one should not assume that the phonon nonequilibrity will propagate with a small velocity u; small perturbations on the background of an equilibrium phonon gas propagate with the second sound velocity less than s, but of the same order.

We may thus draw the following conclusion: if the limitations on power or on temperature are not fulfilled, the "quasiballistic regime" turns into a hydrodynamic one, but no essential change in the velocity of the phonon nonequilibrity propagation takes place in this case. So, it is most probable that in the experiment conducted by V. L. Broude the nonequilibrity oscillations occur in a hydrodynamic regime of hot phonon motion.

It is easy to show which is the frequency of the hot phonons $\tilde{\omega}$, at what distance from the front surface $\tilde{z}$ and at which moment $\tilde{t}$ the rearrangement into the hydrodynamic regime takes place. Apparently, we are interested in the rearrangement at $\tilde{t} < t_B$. In case the rearrangement occurs at $\tilde{t} > t_B$, then at the moment $\tilde{t}$ the phonons have already repeatedly reflected from both surfaces; in this case the phonon distribution has already lost its oriented momentum and after thermalization it turns out that $u = 0$, i.e., there is not any hydrodynamic motion. In case $\tilde{t} < t_B$, then from Figures 5 and 6 it may be seen that the frequency $\tilde{\omega}$ can be found by putting $n(\omega) = 1$ in (6.5), which gives

$$\tilde{\omega} = \omega_0 \left(\frac{I}{I_0}\right)^{1/4}. \tag{7.1}$$

Now we find

$$\tilde{z} = s\tilde{t} = l_N(\tilde{\omega}) = l_N^0 \left(\frac{I}{I_0}\right)^{-5/4}. \tag{7.2}$$

At $I = 30$ kW/cm^2 this gives $\tilde{\omega} = 0.7\ \omega_0$ and $\tilde{z} = 0.6\ \mu$m. It may be calculated that $\tilde{T} = 30$ K which is appreciably higher than the bath temperature $T = 5$ K. The hydrodynamic regime establishes almost near the front surface.

Now the question remains to be elucidated what is the reason of vanishing of oscillations and of ballistic times of the order d/s? Why does this occur at elevated temperatures and pumpings? In order to answer this question we should take into account R-processes.

8. RESISTIVE PROCESSES

The main property of R-processes is dissipation of momentum. A decrease in the total momentum of the phonons with conserving the total phonon energy decreases the anisotropy of the phonon distribution and decelerates the phonon nonequilibrity propagation.

The simplest R-process is the phonon scattering by static defects when[4]

$$\frac{1}{\tau_R(\omega)} = \frac{1}{\tau_R^0}\left(\frac{\omega}{\omega_0}\right)^4. \tag{8.1}$$

The U-processes are not considered here, since for them it is always hard to estimate $\tau_U(\omega)$, and in a nonequilibrium situation, which mostly is realized in the experiments, it is particularly hard. The thing is that U-processes are determined by high-energy tails of the distribution functions which in our semiquantitative theory are not present at all.

What is changed, if the scattering on static defects only is taken into account? First of all it is clear that since the phonon frequency is not changed

by this scattering, the picture of phonon generations formulated in Section 3 holds too. The quasiballistic regime of propagation retains, provided

$$\tau_R(\tilde{\omega}) > \tau_N(\tilde{\omega}). \tag{8.1}$$

The last inequality refers to the generation which has already reached the rear surface. Its fulfillment provides the same inequality for all the previous generations with $\omega > \tilde{\omega}$. The hydrodynamic regime of hot phonon motion with the velocity $u = s$ holds, if for the time of motion t_B the scattering may be neglected i.e., if

$$\tau_R(\tilde{\omega}) > t_B. \tag{8.2}$$

Analogously, the second sound regime on the background of the thermal bath is retained, if

$$\tau_R(\omega_T) > t_B. \tag{8.3}$$

As the pumping increases, $\tilde{\omega}$ increases also, and with increasing the bath temperature, ω_T increases. It is evident that in this case the inequalities (8.2) and (8.3) may be violated. This explains qualitatively at least the vanishing of oscillations and deceleration of the phonon propagation observed in the experiments.

The isotope scattering produced a contribution to the R-scattering; in this case $\tau_R^0 = 30$ ns.[3] The critical power from (8.2) equals then 380 kW/cm^2. In fact the characteristic times of the order of d/s vanish in the experiment at pumpings less than 100 kW/cm^2, which is quite understandable, since there exist other R-processes, besides the isotope scattering. We thus assume $\tau_R^0 = 5$ ns, which corresponds to the critical power of 60 kW/cm^2. At such a τ_R^0 the critical temperature from (8.3) is equal to 36 K, which reasonably corresponds to the experiment.

9. QUASIDIFFUSIVE REGIME

Most impressive results of the experiments carried out under V. L. Broude refer to the cases of fast phonon propagation with the velocities of the order of the sound velocity. However, in interpretating the experimental results it is very important to understand what is the nature of the slow phonon propagation when the resistive scattering dominates.

Let us consider[5] the phonon propagation, in case the scattering by static defects predominates over the decay, i.e., $\tau_R(\omega) < \tau_N(\omega)$. In this case the phonons of the generation ω for the period of their lifetime will be repeatedly scattered by the defects. Therefore, their displacement in space will not be ballistic, but diffusive. That is, they spread not for the free path $l_N(\omega)$, but for a

diffusive length corresponding to the diffusion coefficient $s^2\tau_R(\omega)$ and to the lifetime $\tau_N(\omega)$, i.e., for the length $l_D(\omega) = [l_N(\omega)l_R(\omega)]^{1/2}$. Precisely the layer of such a thickness in the crystal will be occupied by the phonons of the generation (ω).

It is evident that those phonons $\bar{\omega}$ will reach the rear surface of the crystal, for which $l_D(\bar{\omega}) = d$. Hence,

$$\bar{\omega} = \omega_0\left(\frac{d^2}{l_N^0 l_R^0}\right)^{-1/9}. \tag{9.1}$$

The lifetime $\tau_N(\bar{\omega})$ of the generation with such a frequency determines the time of the nonequilibrity propagation through a crystal. It is:

$$\left(\frac{d}{s}\right)^{10/9}\frac{(\tau_N^0)^{4/9}}{(\tau_R^0)^{5/9}}. \tag{9.2}$$

This time exceeds the ballistic time t_B, but is less than the naive diffusive time estimated in Section 3. We deal with a specific regime which is called quasi-diffusive one. It is of importance that the time (9.2) depends much less on the distance of propagation d, than at usual diffusion ($d^{10/9}$ instead of d^2). Quasidiffusion proceeds essentially faster than a common diffusion, since during the phonon decay the diffusion coefficient rapidly increases.

By analogy with Section 6 the occupation numbers of quasidiffusive propagation can be calculated. The only difference is that in case of diffusion the anisotropy of phonon propagation is less than with a ballistic propagation. therefore, the energy phonon flux will not be $\varepsilon(\omega)s$, as is the case in (6.4), but $\alpha(\omega)\varepsilon(\omega)s$, where

$$\alpha(\omega) = \left[\frac{\tau_R(\omega)}{\tau_N(\omega)}\right]^{1/2} < 1 \tag{9.3}$$

is the degree of the distribution anisotropy for the generation ω.

CONCLUSION

A shortcoming of the theory formulated above is not, as it seems at first sight, the apparent complete absence of "mathematical" argumentation. The argumentation based on the picture of generations replacing one another can be justified with the help of the phonon kinetic equation, which has scaling solutions of a definite type. V. L. Broude believed, as it seems to me, that the absence of sofisticated mathematics is rather an advantage than a limitation of the theory, and I agreed with him.

An actual shortcoming of the theory is that it does not take account of the crystal anisotropy and the presence of three phonon branches. The first steps

in obviating these limitations have been done on the initiative of V. L. Broude: the account taken of anisotropy has made it possible to elucidate for the steps on the curve in Figure 1.[6] However, the account of the acoustic anisotropy is a hard problem, though very perspective, and it is a great pity that V. L. Broude will not any more strive, with the enthusiasm we all remember very well, with the reluctance of theoretician to work at such "trifles" as anisotropy.

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