Kinetics of nonequilibrium acoustic phonons in a thin semiconductor sample

N. N. Zinov'ev, D. I. Kovalev, V. I. Kozub, and I. D. Yaroshetskiĭ

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 23 January 1986)
Zh. Eksp. Teor. Fiz. **92**, 1331–1350 (April 1987)

A study was made of the kinetics of the spectral distribution on nonequilibrium acoustic phonons injected into a thin ($\sim 2\times 10^{-3}$ cm) semiconductor from a metal film (of thickness ≈ 1000–2000 Å) excited by a short laser pulse ($\approx 10^{-8}$ sec). After injection the initial phonon spectrum in the semiconductor was not in equilibrium. The deviation from the Planck distribution function was manifested by a considerable weakening in the high-frequency part of the spectrum. It was found for the first time that this non-Planck distribution function was retained for up to several microseconds, which was much longer than the energy relaxation time of the system estimated allowing for possible return of phonons to the metal film. The observed effects were explained on the assumption that the metal-semiconductor interface was not ideal. A study was made of the spatial, spectral, and time dependences of the number of nonequilibrium phonons in the semiconductor when superfluid, liquid, or gaseous helium was used in a thermostat and the degree of heating of the metal film was varied. The results obtained were a demonstration of the dominant role of heat transfer to helium in large-scale long-term evolution of the nonequilibrium phonon distribution compared with the diffusion of phonons along a sample and the processes of energy relaxation in the system.

One of the central tasks in the physics of the condensed state is the study of the kinetics of phonon systems.[1] Evolution of a nonequilibrium phonon distribution itself has a number of special features. However, equally interesting is the problem of how a particular nonequilibrium is established. Nonequilibrium acoustic phonons (which will be the subject of our investigation) may be created in condensed media by a variety of methods.[2] A thermal generator is used most frequently to create a phonon nonequilibrium in a crystal. The generator is a metal film of thickness $d \approx 500$–3000 Å evaporated on the surface of a semiconductor or an insulator and excited by Joule heating as a result of the passage of a current pulse or by illumination with a sufficiently powerful laser radiation pulse. This heats the phonon system of the metal and nonequilibrium phonons are next injected into the semiconductor or insulator as well as into the ambient medium (thermostat). The nonequilibrium phonon spectrum in the semiconductor (insulator) is then governed by the physical processes in the generator, by the conditions under which phonons cross the interface, by the rate and spectral dependence of the transfer of heat to the thermostat, and by relaxation processes in the semiconductor (insulator) sample itself. In the case of strong heating of a phonon system we can expect changes in the properties of the helium surrounding the sample and this should alter also the phonon kinetics.

In view of this, it would be of considerable interest to investigate the phonon kinetics of the structures formed from a metal film and a pure semiconductor sample of small thickness, in which both impurity and phonon-phonon scattering processes are not very effective; semiconducting properties of the sample make it possible to determine the nonequilibrium phonon spectrum by an optical method. Such an investigation makes it possible to identify the contribution of the processes associated with the phonon generator itself and, in particular, with the characteristics of the metal-semiconductor interface. A system of this kind is also very convenient for the study of the processes occurring at the sample-helium interfaces. It should be pointed out that the emission of nonequilibrium phonons into helium under transient conditions and the dependence of its efficiency on the phonon flux intensity have not yet been investigated sufficiently thoroughly. We have particularly in mind the spectral characteristics of the processes of phonon transfer across the interface, particularly at high frequencies in excess of 100 GHz. Therefore, in the experiments reported below the situation was complex and an important factor governing the nature of the kinetics was the interface.

We concentrated our attention on the kinetics of nonequilibrium phonons, particularly on the spectral evolution of their distribution and its dependence on the intensity of the photon flux under conditions such that the influence of the interfaces was strongest.

EXPERIMENTAL METHOD. DETERMINATION OF THE PHONON SPECTRUM

We investigated CdS single crystals with a residual donor concentration $N_D \lesssim 10^{15}$ cm^{-3}, a residual acceptor concentration $N_A \lesssim 10^{14}$ cm^{-3}, characteristic dimensions of the plates 5×3 mm, and a thickness of $L \approx 20$–40 μ. Samples were grown from the vapor phase and the hexagonal **c** axis was in the plane of the plate; the outer surfaces of a plate were perfect natural growth planes. A metal film of tin was evaporated in 10^{-6} Torr vacuum on substrates kept at 50–100°C and the rate of evaporation was 10–100 Å/sec. The diameter of the Sn film on a sample was $D_0 \approx 0.5$–1 mm. Samples were placed in a helium cryostat operating in the range of temperatures T_0 corresponding to liquid He ($T_0 = 1.3$–4.2 K), and also to gaseous He ($T_0 = 4.2$ K). The temperature was always kept constant to within 0.05 K. The metal film was heated by a nitrogen laser pulse of $\tau_0 \sim 10^{-8}$ sec duration characterized by a power density (allowing for the reflection coefficient) amounting to $\sim 10^2$–10^4 W/cm^2.

The spectrum of nonequilibrium acoustic phonons injected into a sample from the metal film was determined by an analysis of the phonon-induced changes in the lumines-

0038-5646/87/040746-12$04.00
© 1987 American Institute of Physics

cence of exciton-impurity complexes. We studied the luminescence spectra of excitons localized at acceptor impurities (line I_1 according to Ref. 3). We also recorded two signals: the luminescence spectrum of exciton-impurity complexes under equilibrium conditions in the phonon system at $T = T_0$ and the difference luminescence spectrum, formed as a result of modulation of the phonon distribution function by nonequilibrium phonons injected to a sample. We shall discuss how to reconstruct the distribution function of acoustic phonons, knowing the spectral distribution of the intensity of the difference luminescence due to emission processes involving participation of acoustic phonons.

First of all, we note that in the case when radiative recombination involving acoustic phonons is governed by one-phonon processes,[1)] the intensity of the difference luminescence (Stokes band $\delta\Phi_s(\nu,\Delta t)$, anti-Stokes band $-\delta\Phi_{as}(\nu,\Delta t)$) may be linked to the nonequilibrium part of the phonon distribution function $\delta N(\nu, \Delta t)$ by the relationship

$$\delta N(\nu, \Delta t)=\delta\Phi_{as}(\nu; \Delta t)/k(I_1)\alpha V_{as}(\nu), \tag{1}$$

where V_{as} is the probability of the anti-Stokes interaction of electron-impurity complexes with phonons of energy $h\nu$ (Ref. 4) ($h\nu \equiv \hbar\omega = E - E_0$; E_0 and E are, respectively, the values of the photon energy corresponding to the zero-phonon luminescence line and of the running value of the photon energy); Δt is the delay time between the moments of phonon injection and recording of the difference optical signal; $k(I_1)$ is a coefficient dependent on the probability of radiative recombination localized at an impurity center with an *e-h* pair, and also dependent on the light intensity $I = I_1$ governing the concentration of the excited centers; $\alpha = \alpha[\delta N(\nu, \Delta t), T_0]$ is the nonequilibrium value of the Debye-Waller factor. The expression for α obtained using the results of Ref. 4 can be represented as follows:

$$\begin{gathered}\alpha=\exp[-(f(T_0)+\delta f)],\\ f(T_0)=\int_{-\infty}^{\infty}\{[N_{T_0}(\nu')+1]V_s(\nu')+N_{T_0}(-\nu')V_{as}(-\nu')\}d\nu',\\ \delta f=\int_{-\infty}^{\infty}\{V_s(\nu')\delta N(\nu',\Delta t)+V_{as}(-\nu')\delta N(-\nu',\Delta t)\}d\nu',\\ N(\nu,\Delta t)=N_{T_0}(\nu)+\delta N(\nu,\Delta t),\end{gathered} \tag{2}$$

$N_{T_0}(\nu)$ is the equilibrium phonon distribution function. We can determine $\delta N(\nu, \Delta t)$ if we know the dependence $V_{as}(\nu)$, and also the values of k and α. A method for the experimental determination of nonequilibrium occupation numbers was proposed in Ref. 3 (and used subsequently on many occasions—see, for example, Refs. 2 and 5); this method is based on the relationship

$$\delta N(\nu, \Delta t)=\delta\Phi_{as}(\nu, \Delta t)/\Phi_s(\nu), \tag{3}$$

where $\Phi_s(\nu)$ is the luminescence signal in the Stokes wing under equilibrium conditions:

$$\Phi_s(\nu)=k(I_2)\alpha_0 V_s(\nu)[N_{T_0}(\nu)+1]|_{T_0\to 0}\approx k(I_2)\alpha_0 V_s(\nu)$$

(α_0 is the equilibrium value of the Debye-Waller factor at $T = T_0$—see Ref. 4). We can see from Eqs. (1) and (2) that the relationship (3) is satisfied only under the following conditions: a) the intensity of the radiation used to excite luminescence should be the same in recording the dependences $\delta\Phi_{as}(\nu, \Delta t)$ and $\Phi_s(\nu)$: $I_1 = I_2$; b) the values of the factor α are the same for equilibrium and nonequilibrium cases; c) the wings $\Phi_s(\nu)$ and $\Phi_{as}(\nu)$ are due to one-phonon processes; d) the probability of the processes involving absorption and emission of phonons is the same: $V_s(\nu) = V_{as}(\nu)$. The equality of the intensities I_1 and I_2 is usually quite easy to ensure by the use of modulation methods. In the case of the other factors, their influence may be very considerable and requires separate discussion. For example, a sensitive indicator of the equality of $V_s(\nu)$ and $V_{as}(\nu)$ is

$$\delta\Phi_s(\nu, \Delta t)=\delta\Phi_{as}(\nu, \Delta t).$$

We observed experimentally a number of special features particularly in the Stokes part of the spectrum,[5-7] indicating at least at first sight that the proposed simple model does not represent properly the real situation. We have in mind principally the observed asymmetry of the Stokes and anti-Stokes parts of the spectrum of the difference luminescence, as well as the deviation from the laws of conservation of the areas under the spectra of electron-vibrational transitions. Hence, we may suggest that the formation of the observed luminescence wings may not be due to just one-phonon processes but due to multiphonon processes and even the processes not associated with the electron-phonon interaction. A similar complex situation occurs to a greater or lesser extent also in other cases (see, for example, Refs. 5 and 8) when electron-vibrational luminescence bands of bound electron states are used in phonon spectroscopy. It is also very important to note the difference between the Debye-Waller factors in the case of equilibrium and nonequilibrium cases, but this difference does not allow us in principle to find the absolute values of nonequilibrium occupation numbers with the aid of Eq. (3). Therefore, the method for the determination of the nonequilibrium phonon spectrum based on Eq. (3) and described above is strictly speaking not universal. We therefore investigated the characteristic features due to the electron-phonon interaction in CdS (Ref. 9). According to the conclusions reached in Ref. 9, the absence of symmetry between $\delta\Phi_s(\nu)$ and $\delta\Phi_{as}(\nu)$ is due to the presence in the Stokes part of the spectrum, measured relative to the zero-phonon luminescence line of electron-exciton complexes, of a weak zero-phonon luminescence band related both to a deviation from the distribution of the substitutional impurity in the lattice and to different types of acceptor centers. On the other hand, the spectral dependences in the anti-Stokes region do not differ greatly in any of these cases and are due to one-phonon processes. This makes it possible to modify the system for the determination of $V_{as}(\nu)$ as follows[6]:

$$V_{as}(\nu)=\Phi_{as}(\nu)|_{T_0^*}/k(I_2)\alpha_{T_0^*}N_{T_0^*}(\nu), \tag{4}$$

where

$$N_{T_0^*}(\nu)=[\exp(h\nu/T_0^*)-1]^{-1}, \quad \Phi_{as}(\nu)|_{T_0^*}$$

is the anti-Stokes luminescence spectrum at temperatures $T_0^* > T_0$ under equilibrium conditions. The selection of the value of T_0^* is not of fundamental importance and it is governed by the requirement that in the range of energies of interest to us the signal $\Phi_{as}(\nu)|_{T_0^*}$ should exceed the noise level. This selection of T_0^* makes it possible to avoid indeter-

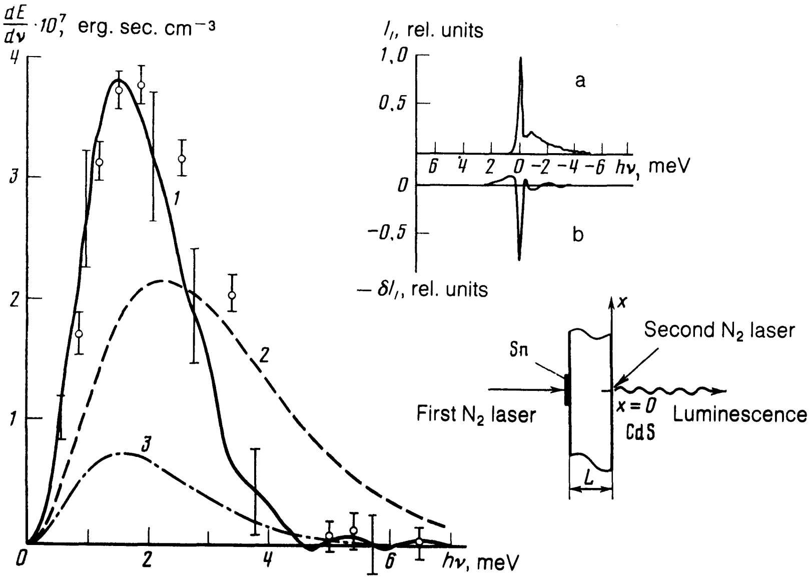

FIG. 1. Spectral distribution of the bulk energy density of nonequilibrium acoustic phonons in a sample of CdS: curve 1 and the symbols (O) are the experimental dependences obtained by different methods; curve 2 is the Planck spectrum with $T = T_E$; curve 3 is the Planck spectrum with $T = T_{\max}$ and $T_0 = 1.3$ K. The inset shows the experimental geometry and the luminescence spectrum in the absence of the effect of nonequilibrium phonons (a) and the difference spectrum induced by nonequilibrium acoustic phonons when the maximum rate of excitation of the metal film was 10^4 W/cm^2 (b).

minacy in the values of $\delta N(\nu, \Delta t)$ calculated from Eq. (1). In the analysis of the experimental dependences of the difference luminescence we used the data on $\Phi_{as}(\nu)\,|_{T_0^*}$ at $T_0^* = 10$ K. The range of phonon frequencies accessible to analysis by such a phonon spectrometer was 0.25 meV $\lesssim h\nu \lesssim 7$ meV. The lower limit was set by the width of the zero-phonon luminescence line and could in principle become $\sim 10^{-3}$ meV. The upper limit was governed by the width of the part of the spectrum free of the luminescence due to other impurity centers. It should be noted that a calculation of $\delta N(\nu, \Delta t)$ carried out using Eqs. (1) and (4) gave only the relative values of the occupation numbers in view of the difference between the values of α. However, this method utilizing—in contrast to Eq. (3)—only the characteristics of the anti-Stokes luminescence eliminates possible errors in the determination of the dependence $\delta N(\nu, \Delta t)$.

An independent experimental method for the determination of the phonon spectrum can be based on the relationships

$$\gamma(\nu, \Delta t) = \Phi_{as}(\nu, \Delta t)/J(0) = V_{as}(\nu) N(\nu, \Delta t), \qquad (5)$$

where $J(0) = k(I_2)\alpha$ and $\Phi_{as}(\nu, \Delta t)$ are, respectively, the intensity and the maximum of the zero-phonon luminescence line and the intensity of the anti-Stokes luminescence;

$$N(\nu, \Delta t) = N_{T_0}(\nu) + \delta N(\nu, \Delta t), \quad \delta N \gg N_{T_0}.$$

We can see from Eq. (5) that the ratio γ is a universal quantitative characteristic governed by the nonequilibrium occupation numbers corresponding to a given energy $h\nu$. In turn, the absolute values of $\gamma(\nu, \Delta t)$ obtained for the nonequilibrium case can be associated with the corresponding values of $\gamma(\nu, T_0^*)$ obtained from a series of luminescence spectra recorded under equilibrium conditions at various thermostat temperatures T_0^*. A comparison of $\gamma(\nu, \Delta t)$ and $\gamma(\nu, T_0^*)$ can be used to find the effective phonon temperature $T_{\text{eff}} = T_0^*$, corresponding to a given energy $h\nu$ $[\gamma(\nu, \Delta t) = \gamma(\nu, T_0^*)]$, and the corresponding occupation numbers:

$$\delta N = [\exp(h\nu/T_{\text{eff}}) - 1]^{-1}.$$

Therefore, an analysis of the difference anti-Stokes luminescence and of the equilibrium spectra recorded at different temperatures can give both the functional dependences $\delta N(\nu, \Delta t)$ and the absolute values of the nonequilibrium occupation numbers. It is important to stress that both methods used by us give dependences $\delta N(\nu, \Delta t)$ which are practically identical (see the example in Fig. 1). An analysis of the experimental dependences allowed us to find the nature of the spectrum $\delta N(\nu, \Delta t)$ from Eqs. (4) and (1); we then used the above-mentioned identity of the form of the dependences $\delta N(\nu, \Delta t)$ determined by both methods to normalize the resultant spectrum $\delta N(\nu, \Delta t)$ to the absolute values of the occupation numbers obtained from Eq. (5) when the energy $h\nu$ is fixed (we selected $h\nu = 1.5$ meV).

Although the experiments give the absolute values of the phonon occupation numbers, the experimental data could be analyzed conveniently introducing a weighting factor ν^3 allowing for the relative contribution of a given group of phonons to the total energy density in the Debye model (which is definitely valid in the investigated range of frequencies). By way of example, we shall give the spectral dependences

$$dE/d\nu = \rho(\nu) h\nu \delta N(\nu),$$

where $\rho(\nu) = 6\nu^2/\bar{s}^3$ is the density of the phonon states in the Debye model; $\bar{s}^{-3} = (s_l^{-3} + s_{t\parallel c}^{-3} + s_{t\perp c}^{-3})/3$; $s_l = 4.3 \times 10^5$ cm/sec, $s_{t\perp c} = 3.35 \times 10^5$ cm/sec, $s_{t\parallel c} = 1.8 \times 10^5$ cm/sec.

The luminescence due to exciton-impurity complexes I_1 was excited on the opposite surface of a sample by using pulses of $\approx 10^{-8}$ sec duration from a second nitrogen laser. Since the lifetime of exciton-impurity complexes was $\tau \approx 10^{-9}$ sec (Ref. 10), these experiments corresponded to quasisteady excitation regimes. The steady-state concentration of exciton-impurity complexes was governed by the rate of excitation, the concentration of nonequilibrium e-h pairs, their cross section for binding to an impurity center, and the concentration of the impurity centers of the relevant type. Since the diffusion length of free excitons L_{DX} did not exceed $(1\text{-}5) \times 10^{-4}$ cm (Ref. 11) and the depth of penetration of the exciting N_2 laser radiation was $\alpha^{-1} \sim 10^{-5}$ cm, under our conditions such exciton-impurity complexes were concentrated in an excited layer of a crystal with a characteristic depth $\lesssim L_{DX} \sim 10^{-4}$ cm. The diameter of the excitation spot on a sample was $(5\text{–}10) \times 10^{-3}$ cm and the power density in the spot did not exceed $(0.5\text{–}1) \times 10^2$ W/cm^2.

The difference luminescence spectrum was determined by the operation

$$\delta I(\nu_i, t) = \frac{1}{M} \sum_{k=1}^{M} \left(I_k(\nu_i, t) - I_{k-1}(\nu_i, t) \right), \tag{6}$$

where

$$\{k\} = \begin{Bmatrix} 0 \\ 1 \end{Bmatrix}, \quad \begin{Bmatrix} 1 \\ 1 \end{Bmatrix}$$

denotes the states of the first and second (I and II) lasers, respectively (the laser denoted by I was used to excite the luminescence and the laser denoted by II excited the metal film; the number 1 corresponds to irradiation of a sample or film with a laser pulse, and the number 0 indicates the absence of such irradiation); $i = 250$. The luminescence at the output of a double diffraction monochromator of the DFS-24 type with a spectral slit width of 0.25 meV was recorded using a fast ÉLU-24FM photomultiplier with a time resolution of $\lesssim 10^{-9}$ sec. After pulsed analog-digital conversion the signal was accumulated in the memory of a DZ-28 microcomputer. A program was used to control a step motor in the monochromator and the moment of ignition of the lasers. A typical number M of the accumulation cycles at a fixed wavelength was ~ 250. The clock frequency of the whole apparatus was set by an external master oscillator. In addition to triggering the system, the master oscillator provided also a triggering pulse for the system controlling the motor of the mechanical modulator (chopper) which, firstly, automatically divided the repetition frequency of the laser II by a factor of 2 and, secondly, in accordance with the angular position of the chopper disk, selected the voltage needed to set the sign of the quantity $I_k(\nu_i, t)$ occurring in Eq. (6), which made it possible to identify the difference luminescence and at the same time suppress various perturbations.

FORMATION OF THE SPECTRAL DISTRIBUTION AND KINETICS OF NONEQUILIBRIUM ACOUSTIC PHONONS

In studies of kinetic effects in a nonequilibrium phonon system the following two aspects are most interesting: the spatial and spectral kinetics. We shall begin with discussion of the latter.

Figure 1 gives not only the experimental spectrum recorded at a moment[2)] $\Delta t = t_1 = 3 \times 10^{-8}$ sec, but also the energy spectra corresponding to the Planck distribution at two values of the temperature T. In one case the value of $T = T_{\max}$ was selected from the condition $\hbar\nu_{\max} \equiv \hbar\omega_{\max} = 2.83 T_{\max}$ (where $\nu_{\max}$ is the frequency corresponding to the maximum of the experimental spectrum $dE/d\nu$). This condition ensures coincidence of the maximum of the corresponding equilibrium spectrum with $h\nu_{\max}$. Another value of $T = T_E$ is selected from the requirement that the area under the $dE/d\nu$ curve in the case of the Planck distribution should be equal to the area under the experimental spectrum, i.e., the total energy of the phonon system for these distributions should be the same. It should be noted that since all the spectra are plotted using the same value of $\bar{s}$, they can be compared without allowance for the error in the determination of this parameter. We can see from the experimental data that at times $\Delta t \sim t_1$ the phonon spectrum differs considerably from equilibrium. The occupation numbers in the energy range $h\nu \sim h\nu_{\max}$, which are of the order of 0.1–0.5 (in a situation corresponding to Fig. 1 we have $\delta N \approx 0.42$) are considerably greater than those corresponding to the maximum of the Planck distribution (of the order of 0.067). It should be noted that the error in the determination of δN at $\nu = \nu_{\max}$ did not exceed 20%. Turning to the Planck distribution corresponding to the same total energy of the phonon system (Fig. 1), we note that the nature of the experimental spectrum can be interpreted as a considerable reduction in the number of phonons in the high-frequency region compared with the Planck distribution (Fig. 1). It should be pointed out that the value of t_1 is of the order of the time for ballistic transit of phonons across the whole semiconductor sample between the surfaces, i.e., $t_1 \sim L/s$. Figure 2 shows the spectra recorded at a fixed rate of pumping of the metal film at different moments (the time was measured from the beginning of a pump pulse). On increase in t the occupation numbers decrease and the maximum of the energy distribution $dE/d\nu$ shifts slightly toward lower energies. The general nature of the spectrum associated with the "deficit" of high-frequency phonons is however retained. Figure 3 shows the phonon spectra obtained for different pumping rates. We can see that at $t \sim t_1$ the distribution $dE/d\nu$ differs from the Planck form. Reduction in the pumping rate alters considerably the total number of phon-

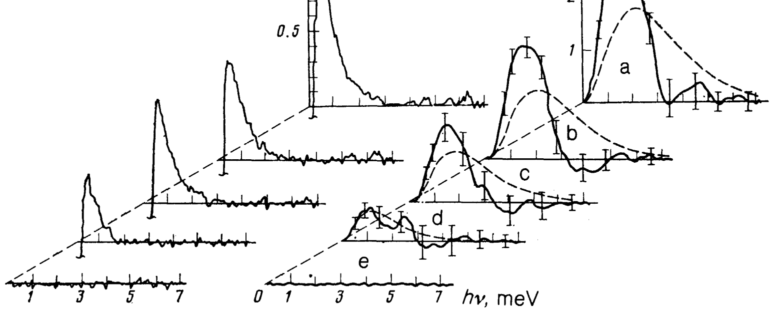

FIG. 2. Spectral distribution of the bulk energy density and the spectra of the anti-Stokes luminescence for a fixed rate of excitation of the metal film $I \approx 10^4$ W/cm^2, plotted for different delays Δt relative to the moment of injection of phonons into a sample kept at $T_0 = 1.3$ K: a) $\Delta t = 30$ nsec, $T_E \approx 9.3$ K; b) $\Delta t = 1$ μsec, $T_E = 8.6$ K; c) $\Delta t = 2$ μsec, $T_E = 7.6$ K; d) $\Delta t = 5$ μsec, $T_E = 5.1$ K; e) $\Delta t = 9$ μsec. The dashed curves represent the Planck spectra with $T = T_E$.

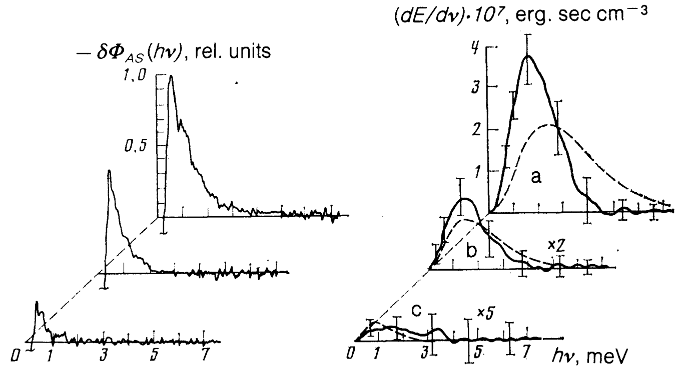

FIG. 3. Spectral distributions of the bulk energy density and the anti-Stokes luminescence spectra obtained for a fixed delay of $\Delta t \approx 30$ nsec and different rates of pumping of the metal film ($I_0 \approx 10^4$ W/cm^2, $T_0 = 1.3$ K): a) $I = I_0$, $T_E = 9.9$ K; b) $I = 0.14 I_0$, $T_E \approx 6.5$ K; c) $I = 0.02 I_0$, $T_E \approx 3.4$ K. The dashed curves represent the corresponding Planck spectra with $T = T_E$.

ons in a sample, but hardly changes the value of $\nu_{\max}$. We can summarize these results by as follows.

1) At short times $t \sim L/s \sim 10^{-8}$ sec the distribution of nonequilibrium phonons differs considerably from the Planck distribution with a temperature $T = T_E$. This is unexpected, for example, in the light of the data reported in Refs. 3, 5, and 12.

2) The evolution of the spectrum representing a reduction in the value of $dE/d\nu$ is observed beginning from the ballistic transit time of $\sim 10^{-8}$ sec right up to $\sim 10^{-5}$ sec.

3) In the process of evolution the non-Planck nature of the spectrum is maintained.

4) When time is increased (and when the pumping rate is reduced) the total energy of phonons decreases.

5) There is no significant change in the energy $h\nu_{\max}$ when the pumping rate is varied.

The physical picture behind these experimental observations can be provided if we analyze the processes of relaxation in the phonon system. We shall begin with the answer to the question of whether it is possible to establish a quasiequilibrium in the system under consideration at times $t \lesssim 10^{-5}$ sec. First of all, we note that because of the deficit of high-frequency phonons the processes of attachment should dominate the establishment of an equilibrium in the phonon system. The characteristic relaxation time is then naturally dependent on the nature of the distribution function[1]:

$$\tau_N^{-1} \propto \int d\mathbf{k}'\, d\mathbf{k}''[N(\mathbf{k}') - N(\mathbf{k}'')]F(\mathbf{k}, \mathbf{k}', \mathbf{k}''),$$

where $F(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$ is the probability of the interaction process. We shall also bear in mind that the bulk of the phonons is concentrated in the frequency interval $\nu \sim \nu_{\max}$ and the form of the spectral dependence deduced from Fig. 1 can be described quite accurately by the Planck distribution with $T = T_{\max}$. The difference between the occupation numbers discussed above can be allowed for in the calculation of the integral characteristics if we introduce the factor

$$\eta = \delta N(\omega_{max})[\exp(h\omega_{max}/T_{max}) - 1]$$

(in the case corresponding to Fig. 1, we have $\eta \approx 6.3$). Therefore, the order of magnitude is

$$\tau_N^{-1} \propto \eta \tau_{0N}^{-1}(T_{max})$$

(where τ_{0N} is the value obtained in the equilibrium case). As far as the estimate of τ_{0N} is concerned, it follows from the theory of Ref. 1 that

$$\tau_{0N}^{-1} \propto (\hbar\omega)^m T^n / \rho \hbar^4 s^5,$$

where $n + m = 5$ and the value of m depends on the crystal symmetry, on the phonon polarization,[3)] and on the relationship between $\hbar\omega$ and T; in the interpolation region $\hbar\omega \sim T$ such an estimate is fairly rough. According to this estimate, we find that for

$$h\omega \sim T \sim T_{max},$$

we have $\tau_{0N} \sim 10^{-5}$ sec. An estimate of τ_{0N} for CdS from the experimental data on the thermal conductivity[13] gives $\tau_{0N}^{-1} = (1.4 \times 10^{-22}\ \text{sec/K})\omega^2 T^3$, which corresponds to $\tau_{0N} \sim (2\text{-}3) \times 10^{-6}$ sec for $\omega \sim \omega_{\max}$ at $T = T_{\max}$; this estimate of τ_{0N} can be regarded as an order-of-magnitude estimate. It is clear from these discussions that the experimentally observed ineffectiveness of the phonon-phonon relaxation at times $\lesssim 10^{-6}$ sec is not in conflict with current ideas.

However, in the investigated semiconductor-metal system we can expect manifestation of a very effective thermalization mechanism associated with phonon-electron processes in the metal film. In fact, since the ballistic transit time of phonons between the surfaces of the semiconductor sample is relatively short ($\sim 10^{-8}$ sec), compared with the lifetime of a phonon nonequilibrium ($\sim 10^{-5}$ sec), this system may be characterized by the return of phonons reflected from the semiconductor-helium interface to the metal film. Bearing in mind that the phonon-electron interaction in the metal is very effective and can be much more important in the low-frequency range than the phonon-phonon interaction, we can expect a significant influence of the electron-phonon processes on the phonon spectrum. In the case of an arbitrary relationship between the mean free path of phonons in the metal $l_{ph-e} \sim (s/\omega)(V_F/s)$ and the film thickness d, an estimate of the effective phonon-electron relaxation time of the system gives

$$\tilde{\tau}_{ph-e}^{-1} \approx \frac{s}{L} \min\left(\frac{\omega d}{V_F}, 1\right). \qquad (7)$$

In the case when $\hbar\omega \approx 1.5$ meV, we have $\tilde{\tau}_{ph-e} \sim 3 \times 10^{-8}$ sec. However, in this estimate we are assuming that the coefficient k_t representing the passage of phonons from the semiconductor to the metal and back again is of the order of unity. An approximate calculation of the coefficient k_t carried out using the theory of acoustic mismatch[15] in the specific case of the CdS–Sn interface gives $k_t \gtrsim 0.3$. An analysis of the simplest model in which the value of k_t is assumed to be the same for all the phonons demonstrates that, if $k_t \ll 1$, the time needed to establish an equilibrium is governed by the quantity $\max(\tilde{\tau}_{ph-e}, k_t^{-1} L/s)$ and in our case it amounts to $\lesssim 10^{-7}$ sec. It therefore follows that the return of phonons to the metal film should establish an equilibrium in the investigated structure already after $\lesssim 10^{-7}$ sec; however, this is not confirmed by the experimental results. This behavior should be attributed to the frequency dependence of heat transfer (we shall discuss later the influence of the latter on the phonon kinetics); however, estimates and the experimental results indicate that this factor begins to have an influence only after $\gtrsim 1\,\mu$sec. Therefore, we can draw the conclusion that the return of phonons to the metal film is hindered by some factor. This may happen, for example, if the thermal contact between the film and the semiconductor is not established over the whole area of the film, but in certain narrow regions. This assumption is quite realistic since thin metal films prepared by vacuum evaporation have usually a granular structure with granules of the order of the film thickness[16] and the semiconductor surface is not wetted by the metal. On the other hand, if we assume that in the vicinity of such narrow contacts the metal is largely disturbed so that the elastic scattering of phonons on defects and dislocations and also on defect clusters becomes important, then—as shown below—we can account also for the frequency spectrum of the phonons emitted by the film. The importance of the contribution of the phonon elastic scattering process at metal microcontacts, allowance for which makes it possible to explain a number of experimental observations, was pointed out by Kulik *et al.*[17]

We shall consider the following model which can explain the set of the experimental data reported above. We shall assume that a metal film of thickness d is deposited on the surface of a semiconductor and that the thermal contact between this film and the substrate is established at certain parts on the film surface with a characteristic diameter a and the distance between such parts of the surface is $\mathscr{L}$ (Fig. 4). We shall assume that the mean free path of phonons in a film in the case of elastic scattering (by defects) $l_{ph,i}$ is much less than the characteristic length of the inelastic phonon-electron interaction (for electrons the corresponding inequality is practically always satisfied in the films). In this case the electron-phonon system of the metal film is described by the equations

$$\partial n_\varepsilon/\partial t - D_\varepsilon{}^e \Delta n_\varepsilon + I_{e-ph}(n_0+n_\varepsilon{}^1,\ N_0+N_\omega{}^1)=0, \tag{8}$$

$$\partial N_\omega/\partial t - D_\omega{}^{ph} \Delta N_\omega + I_{ph-e}(N_0+N_\omega{}^1,\ n_0+n_\varepsilon{}^1)=0, \tag{9}$$

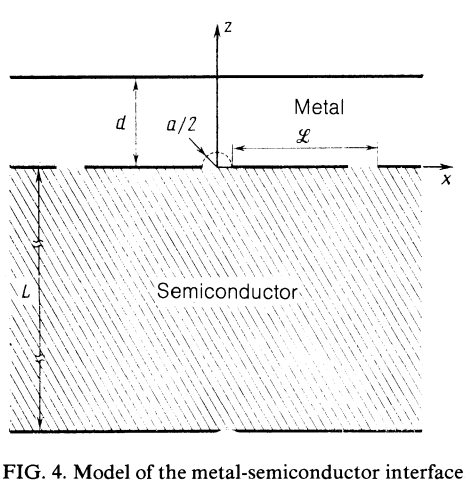

FIG. 4. Model of the metal-semiconductor interface.

where $n_\varepsilon = n_0 + n_\varepsilon^1$, and $N_\omega = N_0 + N_\omega^1$ are, respectively, the distribution functions of electrons and phonons averaged over a constant energy surface; n_0 and N_0 are the local-equilibrium parts of the functions corresponding to a temperature $T = T(\mathbf{r})$ dependent on the coordinates; we then have

$$I_{e-ph}(n_0, N_0)=0;\ I_{ph-e}(N_0, n_0)=0;$$

D_ε^e and D_ω^{ph} are the diffusion coefficients of electrons and phonons, respectively. We shall assume that the diffusion length of electrons

$$L_{De}=(D^e\tau_{e-ph})^{1/2}$$

is much less than the distance $\mathscr{L}$. In this case the transport of heat in a metal film can be described by the heat conduction equation obtained if we multiply Eqs. (8) and (9) by $\varepsilon_\mathbf{p}$ and $\hbar\omega$ and then sum over the electron and phonon states using the expressions

$$\sum_\mathbf{p} \varepsilon_\mathbf{p} I_{e-ph} = -\sum_\mathbf{q} \hbar\omega I_{ph-e},$$
$$(C_{ph}+C_e)\,dT/dt-(\varkappa_e+\varkappa_{ph})\Delta T=0, \tag{10}$$

where C_{ph} and C_e are the phonon and electron specific heats, and $\varkappa_{ph}$ and $\varkappa_e$ are the thermal conductivities; we shall assume here that $C_{ph} \gg C_e$, $\varkappa_e \gg \varkappa_{ph}$. [We are assuming here that $\varkappa \sim DC$; if

$$C_{ph}/C_e \sim \pi^2 T^2/\Theta_D m s^2, \quad D_e/D_{ph} \gtrsim V_F/s,$$

where $D_e \sim V_F d/3$ and $D_{ph} \lesssim sd/3$, then for $\Theta_D \sim 100$ K and $T < 100$ K the above inequalities are obeyed.]

The initial condition for Eq. (10) will be $T = T_m = \text{const}$ (corresponding to instantaneous laser heating to a temperature T_m). In the absence of a thermal contact with the semiconductor and in the case of negligible (over time intervals of $\sim 10^{-8}$ sec) heat transfer to helium the boundary condition is $\nabla_z T\,|_{z=0,\,d} = 0$. Since we shall subsequently assume that $a \lesssim L_{De} \ll \mathscr{L}$, a thermal contact with the sample will be regarded as a point-like sink the efficiency of which generally depends on the local temperature. In other words,

$$I_t = d\varkappa_e \oint \nabla T\, dl = f(T), \tag{11}$$

where the integral is taken over a contour surrounding the sink point (if $d \lesssim L_{De}$, the problem of propagation of heat in a film can readily be shown to reduce to the two-dimensional case).

We can find the function $f(T)$ and determine the nature of the frequency distribution of the emitted phonons by solving the transport equations [see Eqs. (8) and (9)] in the region of such a contact. The boundary conditions for electrons is $\nabla_z n_\varepsilon\,|_{z=0,\,d} = 0$. The boundary condition for phonons can be written as follows:

$$\nabla_z N_\omega|_{z=0,\,d}=0, \quad D_{ph}\nabla_z N_\omega|_{z=0}=k_{t\omega} N_\omega s$$

for the regions outside the contact and in the contact, respectively ($z = 0$, $x^2 + y^2 < a^2/4$). We shall introduce a coefficient $k_{t\omega}$ representing the penetration of phonons across the interface (in general, this coefficient may be frequency-dependent). Such a formulation of the boundary conditions implies that:

a) the temperature of the metal film is much higher than the effective temperature of phonons in the semiconductor;

b) the propagation of phonons in the semiconductor is ballistic.

An analysis of the equation for electrons gives the following estimate:

$$n_\varepsilon{}^1/n_0 \leqslant \min[\nabla T L_{De}/T,\ (N_\omega{}^1/N_0) L_{Dph}/L_{De}] \ll 1.$$

We shall now derive an equation for phonons which can be represented in the form

$$\frac{\partial N_\omega}{\partial t} - D_\omega{}^{ph} \Delta N_\omega + \frac{N_\omega{}^1}{\tau_{ph-e}(T,\omega)} + I_{ph,e}(N_0, n_\varepsilon{}^1) = 0. \qquad (12)$$

Here, the term proportional to $\tau_{ph\text{-}e}^{-1}$ describes the contribution of a local-equilibrium electron distribution function n_0 and the term $I_{ph\text{-}e}(N_0, n_\varepsilon^1)$ gives the contribution of the correction n_ε^1. Bearing in mind the above discussion, we can conclude that the ratio of these terms is large ($\gtrsim L_{De}/L_{Dph} \gg 1$), so that we can ignore the last term in Eq. (12). On the other hand, the characteristic scale of the spatial variation of the function $N_0(T)$ is greater than or of the order of $L_{De} \gg L_{Dph}$, and the characteristic times of the changes in T, N_0, and N_ω^1 are much greater than $\tau_{ph\text{-}e}$. This allows us to ignore the corresponding spatial and temporal derivatives, so that we obtain a homogeneous equation

$$D_\omega{}^{ph} \Delta N_\omega{}^1 - (1/\tau_{ph-e}) N_\omega{}^1 = 0 \qquad (13)$$

subject to the boundary condition in the region of the contact:

$$D_\omega{}^{ph} \nabla_z N_\omega{}^1|_{z=0} = k_{t\omega}(N_0 + N_\omega{}^1) s. \qquad (14)$$

A detailed calculation requires fairly complex computation, in which the real geometry is used. However, estimates obtained for two limiting cases on the assumption that

$$k_{t\omega} \gg D_\omega{}^{ph} \nabla_z N_\omega{}^1 / s N_\omega{}^1$$

yield the following interpolation expression for the partial phonon flux, which is correct to an order of magnitude:

$$I_\omega = {}^1/_4 k_{t\omega} N_\omega s \pi a^2 \rho(\omega) \approx D_\omega{}^{ph} N_0 a \psi(a, d, L_{Dph}) \rho(\omega),$$

$$\psi(a, d, L_{Dph}) = \min\{(\pi/4)(1 + a/L_{Dph}),\ (\pi d/a)(1 + a^2/4L_{Dph}^2)\}, \qquad (15)$$

where $\rho(\omega)$ is the density of the phonon states. Hence, in the case of the function $f(T)$ occurring in Eq. (11) we obtain an estimate

$$f(T) \sim (T/a) \varkappa_{ph} a^2 \psi. \qquad (16)$$

Denoting by δT the characteristic scale of the temperature inhomogeneity and using Eqs. (10) and (15), we obtain the inequality

$$\frac{\delta T}{T} \approx \frac{a}{2\pi d} \frac{\varkappa_{ph}}{\varkappa_e} \psi \ll 1.$$

In other words, in this system the equalization of the temperature along the metal is much faster than the transfer of heat to the semiconductor and the inhomogeneity of the temperature distribution in the film can be ignored. Consequently, the problem of heat transfer from the film reduces to the solution of the equation

$$\mathscr{L}^2 d \frac{dT}{dt} \approx -a \frac{\bar{C} T}{C(T)} \bar{D}^{ph} \psi \qquad (17)$$

and in the case of the characteristic time for the transfer of heat to the semiconductor we obtain

$$\Delta t \approx \frac{\mathscr{L}^2 d}{a \bar{D}^{ph} \psi} \frac{C(T_m)}{\bar{C}}. \qquad (18)$$

Here, $\bar{D}$ and $\bar{C}$ are the values corresponding to characteristic phonon frequencies $\bar{\omega}$, which dominate the thermal conductivity. If $D_\omega^{ph} \propto \omega^{-n}$, then in the case when $n \leqslant 3$, we have $\hbar\bar{\omega} \sim 2.83T$, whereas in the case when $n \geqslant 3$, we find that

$$s\tau_i(\omega = \bar{\omega}) = \min(a, d),$$

so that $\bar{D}^{ph} \approx s \min(a,d)/3$, and $\bar{C} = C(T = \hbar\bar{\omega}/2.83)$.

We shall now consider whether this model can be used to interpret the experimental results. It follows from the experiments that heat is transferred practically completely from the film to the sample in a time $\lesssim 3 \times 10^{-8}$ sec (there is no increase in the energy of the phonon system during the observation time). Using Eq. (18) and the assumption that $a \lesssim d$, we then obtain the following restrictions on the parameter of the model:

$$3d\mathscr{L}^2/sa(s\tau_{ph-i}) \leqslant 3 \cdot 10^{-8} \text{ sec}$$

or for $d \sim 10^{-5}$ cm, we find that $s\tau_{ph-i} \sim a$; $a^2/\mathscr{L}^2 \gtrsim 1/300$. We note that the ratio $a^2/\mathscr{L}^2$ governs the relative area of those parts of the film where a thermal contact is established with the semiconductor and, consequently, it governs the probability of the return of phonons to the metal. If this ratio is sufficiently small, we can account for the ineffectiveness of the return processes and, therefore, of the absence of thermalization of the phonon distribution by phonon-electron processes in the film. An estimate of the return thermalization time $\bar{\tau}_{ph-e}^{-1}$ is in fact then obtained from Eq. (7) if this equation is multiplied by a small parameter $a^2/\mathscr{L}^2$. If $a^2/\mathscr{L}^2 \sim 10^{-2}$, then $\bar{\tau}_{ph\text{-}e}$ exceeds 10^{-5} sec (at $\hbar\omega \sim 1.5$ meV).

The frequency spectrum of the phonons emitted by the film is given by Eq. (15). Assuming that $a^2/\mathscr{L}^2 \ll 1$, it follows from this equation that during periods longer than $\max(2L/s, \Delta t)$ but shorter than the characteristic phonon-phonon relaxation times, the following phonon distribution is established in a sample:

$$N_\omega \approx \frac{I_\omega \Delta t}{\mathscr{L}^2 L \rho(\omega)} \approx \frac{D_\omega{}^{ph}}{\bar{D}^{ph}} \frac{d}{L} N_0(T_m) \frac{C(T_m)}{\bar{C}} \frac{\rho_m(\omega)}{\rho_0(\omega)}, \qquad (19)$$

where ρ_0 and ρ_m are the densities of the phonon states in the semiconductor and in the metal [we recall that the frequencies ω are limited from below by the condition $s\tau_{ph-i}(\omega) \leqslant \min(a,d)$.] We can see that the phonon distribution in the sample may differ considerably from the Planck equation and because of the strong frequency dependence of D_ω^{ph}, we can expect a considerable shift of this distribution toward lower frequencies. For example, if

$$D_\omega^{ph} \propto \omega^{-n}, \quad \hbar\omega > T_m$$

we obtain from Eq. (19)

$$\partial E/\partial\omega \propto \omega^{3-n} \exp(-\hbar\omega/T_m),$$

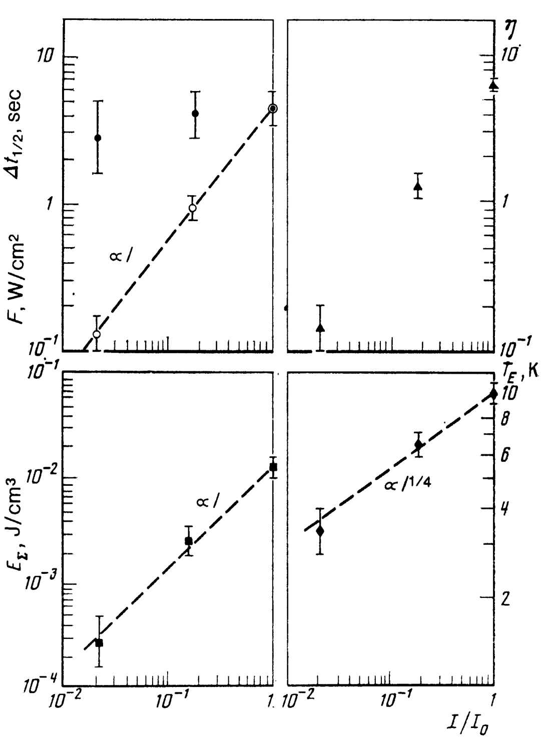

FIG. 5. Dependences of the bulk energy density E, of the effective temperature of a nonequilibrium phonon distribution T_E, of the characteristic heat transfer time $\Delta t_{1/2}$ (open circles), of the heat flux F to the thermostat (black symbols), and of the value of η on the rate of excitation relative to $I_0 \approx 10^4$ W/cm². The thermostat was superfluid He⁴.

and if $\hbar\omega < T_m$, then

$$\partial E/\partial\omega \propto \omega^{2-n}$$

in contrast to the dependence $\partial E/\partial\omega \propto \omega^2$, typical of the Planck distribution). Low-frequency cutoff of this dependence occurs at frequencies $\omega_{\rm lim}$ defined by the condition

$$s\tau_{ph-i}(\omega_{\rm lim}) \sim \min(a,\ d),$$

below which we have $I_\omega \propto \omega^2$. Therefore, if $n > 2$, the frequency dependence of $\partial E/\partial\omega$ corresponding to Eq. (19) has a maximum at $\omega \sim \omega_{\rm lim}$. If $T_m > \hbar\omega_{\rm lim}/2.83$ the position of this maximum should not depend significantly on T_m, i.e., it should not be greatly affected by the pumping rate I. This is precisely the behavior which is exhibited by the experimental data in Fig. 3. A reduction in I results primarily in a reduction of the occupation numbers $N(\nu_{\max})$, where

$$\nu_{\max} = \omega_{\max}/2\pi\ .$$

It follows from the theory that such a dependence can be expected up to values $T_m \sim \hbar\omega_{\rm lim}/2.83$. At lower generator temperatures the frequency dependence of the efficiency of the transmission of phonons to the semiconductor sample becomes unimportant:

$$N_\omega \sim (d/L) N_0(T_m,\ \omega)\,[\rho_m(\omega)/\rho_0(\omega)].$$

Figure 5 shows the experimental dependences of the ratio

$$\eta = N(\nu_{\max})/N_0(T_{\max})$$

on I; we can see that this is described quite satisfactorily by the law $\eta \propto I$, which follows from Eq. (19). In turn, we can use the experimental results to show that in the case of the investigated phonon generator, we have

$$s\tau_{ph-i}'(\omega \sim 1.5\ {\rm meV}/\hbar) \sim 10^{-5}\ {\rm cm}.$$

Unfortunately, because the experimental values of the ratio $T_m/\hbar\omega_{\rm lim}$ are not very high, a reliable estimate of the power exponent n on the $\tau_{ph-i}(\omega)$ is not possible. Therefore, our model reflects the main qualitative features of the processes of penetration of phonons from the generator into the sample which have been observed experimentally. It should be noted that certain difficulties have been encountered in explaining the inefficiency of the processes of phonon return to the metal if the metal-semiconductor interface is assumed to be homogeneous. This inefficiency can then be related to the existence of a "passive" group of phonons in the semiconductor sample because the penetration of such phonons to the metal is unlikely. Some of the observations supporting the formation of such a group include focusing of phonons in the sample, the difference between the phonon spectra of the sample and film, and the absence of a transverse rigidity of the metal-semiconductor interface. However, it is not possible to develop a self-consistent model without invoking some particular concepts which are to some extent artificial.

For example, a metal generator creates a non-Planck distribution of phonons. As pointed out above, in the lowest approximation such a distribution can be described (at least in the case when $n > 3$ or at temperatures $T_m \lesssim \hbar\omega_{\rm lim}$) by two parameters: the most probable phonon energy $\hbar\omega_{\max} = 2.83 T_{\max}$ and by the ratio $\eta = N(\omega_{\max})/N_0(\omega_{\max}, T_{\max})$. For clarity, we shall estimate also the temperature T_E of the Planck distribution to which a given nonequilibrium distribution would relax if only phonon-phonon processes take place in the system (Fig. 1). Clearly, the order of magnitude is given by the relationship $T_E \propto T_{\max}\eta^{1/4}$. On the other hand, the value of T_E corresponding to the initial moment may be related to the total energy E received from a laser pulse:

$$E \approx I\tau_0/L \approx \pi^2 T_E^4/10(\hbar\bar{s})^3. \tag{20}$$

The experimental values of T_E and η are plotted in Fig. 5 as a function of the pumping rate I. We can see that the dependence $T_E(I)$ is described quite well by the $I^{1/4}$ law. On the other hand, if $\tau_0 \sim 10^{-8}$ sec, $I \sim 10^4$ W/cm², $\bar{s} \sim 2.4 \times 10^5$ cm/sec, and $L \sim 2 \times 10^{-3}$ sec, we find from Eq. (20) that $T_E \sim 10$ K, which again agrees well with the experimental results. Finally, we shall estimate the temperature of a thermal generator in the form of a metal film:

$$T_m \sim T_E[Ld^{-1}(\bar{s}_m/\bar{s})^3]^{1/4}, \tag{21}$$

so that for $T_E \sim 10$ K we have $T_m \sim 31$ K (here, $\bar{s}_m$ is the average velocity of sound in a metal which amounts to 1.87×10^5 cm/sec for Sn) when the thickness of the Sn film is $d \approx 10^{-5}$ cm.

PHONON PROPAGATION IN A SEMICONDUCTOR PLATE. HEAT EXCHANGE WITH THE THERMOSTAT

The spatial and temporal evolution of a nonequilibrium phonon distribution created in a semiconductor sample by a thermal generator is determined by three factors:

a) phonon-phonon processes;

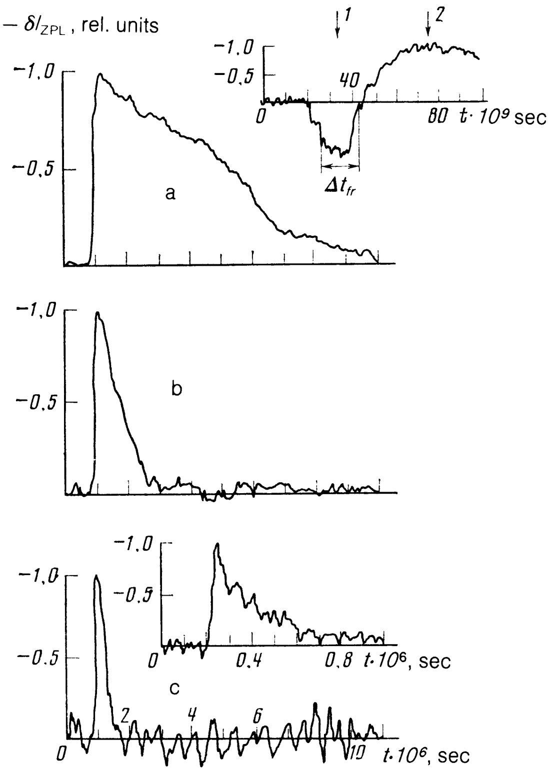

FIG. 6. Time dependences of the difference luminescence at the wavelength of the zero-phonon line obtained for different rates of pumping of the metal film: a) $I = I_0$; b) $I = 0.14I_0$; c) $I = 0.02I_0$; $I_0 \approx 10^4$ W/cm^2, $L = 3\times10^{-3}$ cm, superfluid He^4 used as the thermostat. The inset at the top shows the shape of a laser pulse exciting the metal film (1) and the signal representing the difference luminescence at the zero-phonon line wavelength. The inset at the bottom of the figure shows the shape of the pulse *b* on a scale enlarged (×10) along the *t* axis.

b) phonon diffusion along a sample (spreading of a cloud of phonons initially localized in the region of their injection with a characteristic diameter $\mathscr{D}$);

c) heat transfer representing the loss of phonons to the thermostat kept at a temperature T_0 (our experiments were carried out in superfluid He with $T_0 = 1.3$ K, in liquid He with $T_0 = 4.2$ K, and in gaseous He with $T_0 = 4.2$ K).

First of all, we note that in accordance with the experimental evidence (Fig. 2) the temporal evolution of the phonon distribution for times in excess of 30 nsec is accompanied by a reduction in the total energy of the phonon system. It follows that up to $\lesssim L/s \sim 30$ nsec practically all the injected phonons reach the rear surface of the semiconductor crystal (where they are detected). This conclusion is confirmed also by the kinetics of the signal of the difference luminescence represented by the zero-phonon line $\delta I_{\rm ZPL}$ (Fig. 6), governed by the total number of phonons per unit volume of the sample. The inset at the top of Fig. 6 shows the relative positions, on the time scale, of the pulses representing excitation of the metal film (1) and the leading edge of the zero-phonon luminescence line $\delta I_{\rm ZPL}$ (2); for clarity, the pulse (1) is represented with its polarity reversed. A comparison of the parameters of these pulses shows that the durations of the edges are not significantly different, reflecting the ballistic nature of the propagation of phonons across the CdS sample. On the other hand, the time separation between them Δt_{fr} is ~ 15 nsec, i.e., it is of the order of the ballistic transit time L/s. The fall of the signal $\delta I_{\rm ZPL}$ becomes significant already for $t > 30$-50 nsec. We can therefore conclude that the propagation of phonons across the sample is basically ballistic and the processes of inelastic scattering by impurities and defects are not important in CdS. The motion along the sample is of diffuse nature because of the diffuse reflection of phonons by the surface; the diffusion coefficient is estimated to be

$$D_0^{ph} \approx s^2 L/3; \qquad s \approx 1 - p,$$

where p is the parameter representing the specularity of the reflection by the surface; in the case of specular reflection we have $p = 1$, in the case of the completely diffuse reflection we find that $p = 0$.

In the case of heat transfer this factor may, in particular, affect the nature of the phonon spectrum: it is known that an increase in the phonon energy increases the probability of their transfer to helium.[18] This may favor retention of deviations from the Planck distribution also after longer times.

We shall now consider the experimental results obtained in a study of phonon propagation along a semiconductor film. Figure 7 shows the time dependences of the difference luminescence signal at the wavelength of the zero-phonon line recorded at two points separated by a distance of ~ 0.5 mm under different external conditions (superfluid helium, liquid helium, and gaseous helium). It is clear from the experimental results that the pattern is qualitatively similar in all cases: the transport of phonons along the film by diffusion is demonstrated clearly and the change from superfluid to gaseous helium increases the delay of attainment of the signal maximum at points with the coordinates $x = 0.5$ mm and $y = 0$ when the rate of excitation of the metal film is kept constant. We shall now consider the nature of the phonon spectrum in these cases. It is clear from

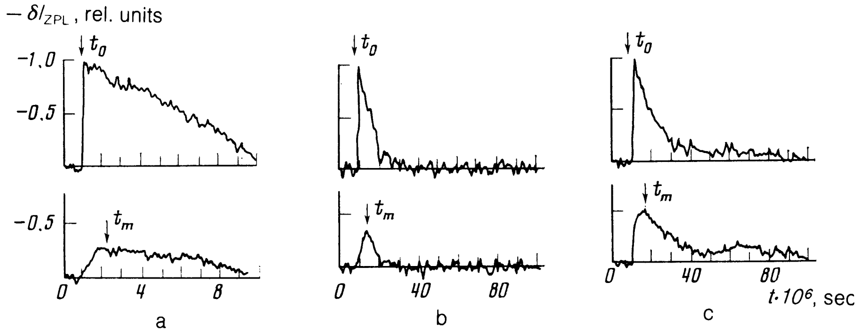

FIG. 7. Time dependences of the difference luminescence at the wavelength of the zero-phonon line recorded at two points when nonequilibrium phonons were detected: $x = 0$ (upper curves) corresponds to the region opposite the center of the metal film and the other point is at $x = 0.05$ cm (lower curves); t_0 is the moment of excitation of the metal film and t_m is the time corresponding to the maximum of the dependence $I_{\rm ZPL}(t)$ at the point $x = 0.05$ cm; $I \approx 0.14\times10^4$ W/cm^2. Thermostat: a) super fluid He^4 at $T_0 = 1.3$ K; b) liquid He^4 at $T_0 = 4.2$ K; c) gaseous He^4 at $T_0 = 4.3$ K.

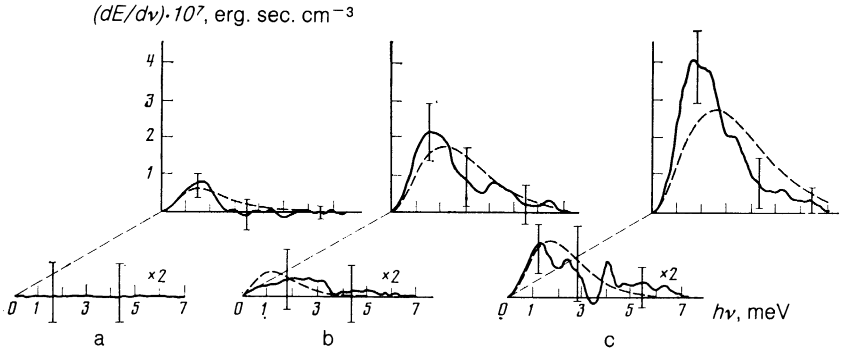

FIG. 8. Spectral distributions of the bulk energy density obtained using different thermostats: a) superfluid He^4 at $T = 1.3$ K; b) liquid He^4 at $T_0 = 4.2$ K; c) gaseous He^4 at $T_0 = 4.3$ K; $I \approx 0.14 \times 10^4$ W/cm^2. The time delay in recording the spectra at $x = 0$ (upper curves) relative to the moment t_0 was 1 μsec and at $x = 0.05$ cm (lower curves) it was $t_m - t_0$ (compared with Fig. 7). The dashed curves are the Planck spectra characterized by $T_E = 6.2$ K (a), 9.2 K (b, cupper curve), 5.2 K (b, lower curve), 10.8 K (c, upper curve), 6.9 K (c, lower curve).

Fig. 8 that a reduction in the rate of transfer of heat to the thermostat has two effects: firstly, the absolute values of the phonon occupation numbers increase in the region of $x = 0$ and $y = 0$ after a long time; secondly, when the sample is in contact with liquid and gaseous helium the relative number of high-frequency phonons begins to increase with time. It should be pointed out that the absolute values of $\partial E/\partial \nu$ obtained for large values of t and also in the peripheral parts of the crystal film are comparable (see Figs. 2, 3, and 8) with the values expected on the basis of the Planck distribution. Unfortunately, the smallness of the signal does not make it possible to analyze the subscript and kinetics of the phonon nonequilibrium in such cases. The results obtained demonstrate definitely a significant influence of the transfer of heat on the large-scale kinetics; moreover, we shall show that these results can be used to obtain a number of quantitative estimates.

We shall now consider in greater detail the experimental results on the large-scale kinetics. Phonon propagation along a film can be described by the following two-dimensional diffusion equation:

$$\partial N/\partial t - D_0{}^{ph}\nabla^2 N = -\beta_\omega N + \hat{I}_{ph-ph}. \tag{22}$$

Here, the quantity β_ω determines the phonon lifetime in a crystal (in the case of loss of phonons to the thermostat); the operator $\hat{I}_{ph-ph}$ allows for the transfer of energy along the spectrum because of phonon-phonon processes: this operator can be ignored for times $t < \tau_N$. An analysis of Eq. (22) for $t > \tau_N$ can be simplified if we multiply this equation by $\hbar\omega$ and integrate with respect to ω, which gives the heat transport equation

$$\partial Q/\partial t - D_0{}^{ph}\nabla^2 Q = -F, \tag{23}$$

where

$$Q = \sum_\omega \hbar\omega N,$$

and the function F describes heat transfer. The solutions of Eqs. (22) and (23) are easily obtained in the case when they reduce to linear equations with constant coefficients. This can be done in the case of Eq. (22) if $t < \tau_N$ and if β_ω is independent of N_ω; in the case of Eq. (23) such a simplification is achieved if $F = \bar{\beta}Q$, where $\bar{\beta} =$ const (which is true, in particular, if $\beta\omega =$ const). We then have

$$f(x, y=0, t) = \frac{\exp(-\beta t)}{4\pi D_0{}^{ph} t} \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \varphi(x', y') \times \exp\left[-\frac{(x-x')^2 + y'^2}{4D_0{}^{ph} t}\right] dx'\, dy', \tag{24}$$

where $f = N_\omega$ or $f = Q$ and $\beta = \beta_\omega$ or $\beta = \beta$. We can easily see that $\partial f/\partial t < 0$ at $x = 0$; if $x \neq 0$, the dependence $f(t)$ may be nonmonotonic and if the point x is outside the excitation spots, the maximum is given by the following order-of-magnitude condition:

$$t_{max} \approx (1/4\beta^2 + x^2/4D_0{}^{ph}\beta)^{1/2} - 1/2\beta, \tag{25}$$

i.e., if $x^2/D_0^{ph} < 1/\beta$ (when heat transfer is unimportant), we have

$$t_{max} \approx x^2/4D_0{}^{ph},$$

whereas if $1/4\beta \ll x^2/D_0^{ph}$, then

$$t_{max} \approx (x^2/4D_0{}^{ph}\beta)^{1/2}.$$

In reality however the nature of heat transfer may depend on the degree of heating of the sample, i.e., β_ω may be a functional of N_ω and $\bar{\beta}$ may depend on Q. In particular, in the case of liquid helium the heating of a sample may alter the properties of helium near the surface of the sample. In accordance with Eq. (24), we note that if $\beta =$ const, then the time dependence of the luminescence intensity cannot be affected significantly by the pumping rate. It therefore follows that the experimentally observed change in the time dependence of the intensity $\delta I_{\rm ZPL}$ due to a change in I (Fig. 6) is evidence of the dependence of the time representing the loss of excitons to the thermostat on the degree of heating (this is at least true of the superfluid-helium thermostat). The nonlinear nature of the heat transfer kinetics makes it much more difficult to analyze quantitatively Eqs. (22) and (23). If this nonlinearity is important, then estimates of the type given by Eqs. (24) and (25) are valid only when a certain characteristic value of β can be defined, for example, during the initial stage of the process for times shorter than $\lesssim \mathscr{D}^2/4D_0\beta$.

We shall now consider the qualitative conclusions that follow from the experimental data presented in Fig. 7. First of all, we note that the maximum characteristic time of the decay of the signal $\Delta t_{1/2}$ (which is determined by the total number of phonons per unit volume of the sample) is of the order of 10 μsec and it applies in the case when the thermostat is in the form of gaseous helium. (The characteristic time of any given change will be considered to be the time in which the signal decreases by a factor of 2.) Hence, we may conclude that the characteristic time of diffusion spreading of the heated region is $t_D \sim \mathscr{D}^2/11.2D_0^{ph} \gtrsim 10\,\mu$sec. Moreover, estimates show that the maximum possible value of l_D in the case when $\mathscr{D} \sim 10^{-1}$ cm, corresponding to $p = 0$, is also of the order of $10\,\mu$sec. Hence, we can draw the following conclusions: firstly, the value of β^{-1} in the case of gaseous helium is considerably greater than $10\,\mu$sec and the decrease in the number of phonons at the point $x = 0$ is governed mainly by diffusion. Secondly, reflection of phonons from the boundaries of a sample is largely diffuse. On the other hand, the reduction in the time $\Delta t_{1/2}$ when a change is made from liquid to superfluid helium shows that the value of β^{-1} (for a given pumping rate I) in such situations is $\lesssim 10$ μsec and it is heat transfer that determines the phonon lifetime at the point $x = 0$. Moreover, the presence of a maximum in the time dependence $\delta I_{\rm ZPL}$ at the point $x = 0.05$ cm and a shift of this maximum toward shorter times on improvement in heat transfer (Fig. 7) are in qualitative agreement with the conclusions stated above.

We shall now estimate the efficiency F of heat transfer in the specific case of superfluid helium using the data on the kinetics of $\delta I_{\rm ZPL}$ (Fig. 6). Obviously, the order-of-magnitude estimate is $F \sim E_\Sigma L/2\Delta t_{1/2}$, where $\Delta t_{1/2}$ is the characteristic decay time of the $\delta I_{\rm ZPL}$ signal (strictly speaking, $\delta I_{\rm ZPL}$ is more likely to be governed by the number of phonons rather than by their total energy, but the corresponding difference can be ignored). The total energy density in the phonon system E_Σ at the point $x = 0$ can be estimated at a time $t = 30$ nsec using the phonon spectra (Fig. 2). The results of such estimates are presented in Fig. 5. An analysis of these results and a comparison with those obtained from other experiments, particularly with the behavior of heat transfer under equilibrium conditions, is complicated by the circumstance that the change in heat transfer on increase in the heating rate may generally be associated with two factors:

1) the frequency dependence of the coefficient β_ω, i.e., the change in the transfer of heat on increase in the characteristic phonon energy[18];

2) the change in the properties of the heat transfer medium during heating.[19]

Therefore, the nature of heat transfer should generally depend on the nature of the phonon distribution function. We have seen already that in our case the change in the pumping rate has practically no effect on the characteristic phonon energy but simply alters the occupation numbers and, consequently, the total energy of the phonon system (and, therefore, the parameter T_E). Consequently, the factor associated with the frequency dependence β_ω can be ignored in the situation under discussion. As far as the characteristic heat transfer time $\Delta t_{1/2}$ is concerned, it follows from Fig. 5 that it rises almost linearly on increase in the pumping rate in the investigated range of excitation intensities. This means that the flow of heat F to the thermostat is not altered by changes in the pumping rate (Fig. 5). The observed constancy of heat evolution is clearly entirely due to the change in the properties of the medium near the interface between the sample and helium. It is worth noting the results of Ref. 19, where bubbles of gaseous helium were observed near the heated surface under approximately the same excitation conditions as in our experiments (at pumping rates $I \sim 10^2 - 10^4$ W/cm^2 in the form of pulses of $\sim 10^{-8}$ sec duration). In the light of this circumstance we can explain the constancy of F when I is varied by the constancy of the rate of transfer of heat to helium in the course of formation of bubbles in a thin layer of this liquid near the surface of a crystal[19] when the vapor formation rate remains constant; the amount of the vapor which is formed should then increase linearly on increase in I.

CONCLUSIONS

1. The spectrum of nonequilibrium phonons injected into a semiconductor sample from a thin metal film is shown to differ considerably from the Planck spectrum. This difference is retained for times up to $\sim 1\,\mu$sec and is characterized mainly by a deficit of high-frequency phonons, as well as by a very weak dependence of the characteristic phonon energy on the pumping rate. A model, based on the hypothesis that the metal-semiconductor interface is not ideal, can explain the main characteristics of the kinetics of nonequilibrium phonons in a system of this kind.

2. It is shown that spatial and temporal evolution of the distribution of phonons after long times is governed both by the processes of their diffusion along a sample and by the transfer of heat to the thermostat (helium). A determination of the efficiency of heat transfer to superfluid helium is reported. It is shown that this efficiency is practically independent of the rate of pumping of the metal film.

1) In the opposite case the validity of the method is limited to small values of the occupation numbers $\delta N(\nu, \Delta t) \ll 1$.

2) Here and later the experimental spectrum represents the results of smoothing over 250 points of the experimental frequency dependence of $dE/d\nu$. The smoothing was carried out using the expression $y_i' = (1/4)(y_{i-1} + 2y_i + y_{i+1})$, where y_i is the initial value of the function and y_i' is smoothed value. The outer points were analyzed using the formulas $y_0' = (2y_0 + y_1 + y_2)/4$ and $y_{250}' = (2y_{250} + y_{249} + y_{248})/4$. The initial scatter of the points is represented in the graphs by vertical segments.

3) It should be noted that because of mode conversion as a result of reflection of phonons by the boundaries of a sample, at times $t > L/s$ we have to allow for the processes which ensure the minimum value of τ_{0N} (Ref. 14).

[1] V. L. Gurevich, Kinetics of Phonon Systems (in Russian), Nauka, Moscow (1980).

[2] W. Eisenmenger, in: *Physical Acoustics*; *Principles and Methods* (ed. by W. P. Mason and R. N. Thurston), Vol. 12, Academic Press, New York (1976), p. 79; W. E. Bron, Rep. Prog. Phys. **43**, 301 (1980).

[3] J. Shah, R. F. Leheny, and A. H. Dayem, Phys. Rev. Lett. **33**, 818 (1974).

[4] I. S. Osad'ko, Usp. Fiz. Nauk **128**, 31 (1979) [Sov. Phys. Usp. **22**, 311 (1979)].

[5] W. E. Bron and W. Grill, Phys. Rev. B **16**, 5303, 5315 (1977).

[6] V. I. Arakin, N. N. Zinov'ev, U. Parmanbekov, and I. D. Yaroshetskii, Solid State Commun. **55**, 733 (1985).

[7] N. N. Kovaleva, K. P. Meletov, and E. F. Sheka, Zh. Eksp. Teor. Fiz. **87**, 2085 (1984) [Sov. Phys. JETP **60**, 1204 (1984)].

[8] A. V. Akimov, S. A. Basun, A. A. Kaplyanskiĭ, and R. A. Titov, Fiz. Tverd. Tela (Leningrad) **20**, 220 (1978) [Sov. Phys. Solid State **20**, 122 (1978)].

[9] N. N. Zinov'ev, D. I. Kovalev, and I. D. Yaroshetskiĭ, Fiz. Tverd. Tela (Leningrad) **28**, 3595 (1986) [Sov. Phys. Solid State **28**, 2026 (1986)].

[10] V. B. Timofeev and T. N. Yalovets, Fiz. Tverd. Tela (Leningrad) **14**, 481 (1972) [Sov. Phys. Solid State **14**, 399 (1972)].

[11] N. N. Zinov'ev, L. P. Ivanov, I. G. Lang, S. T. Pavlov, A. V. Prokaznikov, and I. D. Yaroshetskiĭ, Zh. Eksp. Teor. Fiz. **84**, 2153 (1983) [Sov. Phys. JETP **57**, 1254 (1983)].

[12] T. E. Wilson, F. M. Lurie, and W. E. Bron, Phys. Rev. B **30**, 6103 (1984).

[13] G. E. Moore Jr and M. V. Klein, Phys. Rev. **179**, 722 (1969).

[14] Y. I. B. Levinson, in: *Nonequilibrium Phonons in Nonmetallic Crystals* (ed. by W. Eisenmenger and A. A. Kaplianskii), North-Holland, Amsterdam (1986), p. 91 [Modern Problems in Condensed Matter Sciences, Vol. 16].

[15] J. D. N. Cheeke, H. Ettinger, and B. Hebral, Can. J. Phys. **54**, 1749 (1976).

[16] I. O. Kulik and I. K. Yanson, Josephson Effect in Superconducting Tunnel Structures [in Russian], Nauka, Moscow (1970), p. 153.

[17] I. O. Kulik, A. N. Omel'yanchuk, and I. K. Yanson, Fiz. Nizk. Temp. **7**, 263 (1981) [Sov. J. Low. Temp. Phys. **7**, 129 (1981)]; I. O. Kulik, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 302 (1985) [JETP Lett. **41**, 370 (1985)].

[18] P. L. Kapitza, Zh. Eksp. Teor. Fiz. **11**, 1 (1941); A. C. Anderson, in: *Phonon Scattering in Solids* (Proc. Second Intern. Conf., Nottingham, 1975, ed. by L. J. Challis, V. W. Rampton, and A. F. G. Wyatt), Plenum Press, New York (1976), p. 1.

[19] V. A. Tsvetkov, A. S. Alekseev, M. M. Bonch-Osmolovskiĭ, T. I. Galkina, N. V. Zamkovets, and N. N. Sibel'din, Pis'ma Zh. Eksp. Teor. Fiz. **42**, 272 (1985) [JETP Lett. **42**, 335 (1985)].

Translated by A. Tybulewicz