Dynamics of the development of high-frequency nonequilibrium phonons in a thin film

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(Submitted 13 February 1991; resubmitted 8 July 1991)
Zh. Eksp. Teor. Fiz. **101**, 540–544 (February 1992)

The propagation, in a semiconductor film, of high-frequency nonequilibrium phonons that have been formed as a result of cooling of a photoexcited electron-hole plasma is investigated. The only anharmonic process for the nonequilibrium phonons is assumed to be spontaneous decay. Scattering of phonons by impurities is neglected. The film thickness is assumed to be a small quantity $v\tau_l(\omega)$, where v if the phonon group velocity and $\tau_l(\omega)$ is the decay time for a longitudinal phonon. It is established that energy is transported in space by phonons moving almost parallel to the film surfaces, and the characteristic length scale of the phonon nonequilibirum is $\bar{r} \sim vt$.

Upon interband absorption of light, a photoexcited electron-hole plasma emits long-wavelength optical phonons as it cools, and these decay into pairs of short-wavelength acoustic phonons with frequency of the order of the Debye frequency ($\omega \sim \omega_D$).

In the case when the temperature of the equilibrium state of the crystal is low ($T_{eq} \ll \hbar\omega_D$), and the distribution function of the nonequilibrium phonons satisfies the relation $n_{T_{eq}}(\omega) \ll n(\omega) \ll 1$, where $n_{T_{eq}}(\omega) \approx \exp(-\hbar\omega/T_{eq})$ is the distribution function of the equilibrium phonons, the only anharmonic process for the nonequilibrium phonons is spontaneous decay.

We consider thin, pure crystalline films, such that the relative magnitudes of the frequency $1/\tau^*(\omega)$ of scattering by defects, the decay frequency $1/\tau_l(\omega)$, and the quantity v/d (where v is the average group velocity of the phonons and d is the film thickness) can be assumed to be as follows:

$$\frac{1}{\tau^*(\omega)} \ll \frac{1}{\tau_l(\omega)} \ll \frac{v}{d}. \tag{1}$$

The theory of the propagation of superthermal phonons in bulk crystals, where spontaneous decay dominates over scattering by defects, was considered in Refs. 1 and 2. In the case of films, allowance for the scattering of phonons by the boundaries will play an essential role.

For certain values of the light-pumping parameters (the power and duration of the light pulse), as a result of multiple phonon decays a generation of phonons with distribution function $n(\omega) \sim 1$ can appear. The processes of phonon-phonon coalescence and induced decay become just as probable as spontaneous decay. Quasiequilibrium, describable by a Planck function with a certain initial temperature $T_0 \gg T_{eq}$, is established as a result of three-phonon interactions. A phonon hot spot (PHS) is formed. The results of the calculations show that the PHS is established before the nonequilibrium phonons begin to emerge appreciably from the region of the light excitation. The heat transport will be determined by the Casimir thermal diffusivity.[3] The spatial expansion of the PHS occurs in a diffusive manner, and the characteristic size of the phonon nonuniformity is on the order of

$$\bar{r}(t) \sim (^1/_3 vdt)^{1/2}. \tag{2}$$

Consider the case when the light-pumping power is sufficiently low, so that a PHS is not established and, among the phonon-phonon scattering processes, only spontaneous decay is important. It might appear that, when the inequality (1) is fulfilled, the problem reduces to one with two-dimensional diffusion, for which the diffusion coefficient is determined by the phonon mean free path (of order d) and the characteristic size of the phonon nonuniformity is determined by Eq. (2). It may happen, however, that over the characteristic lifetime $\tau_l(\omega)$ of the phonon generation, as a result of repeated diffuse scattering of phonons at the boundaries there is an accumulation of a sufficient number of phonons moving ballistically almost parallel to the surfaces of the film, and the principal contribution to the spatial transport of energy will be made by phonons moving along the film. The anisotropic part of the distribution function of these phonons will not be small in comparison with the isotropic part, and the use of the diffusion equation is not justified. To elucidate this question, it is necessary to consider the kinetic equation.

THE KINETIC EQUATION

As already noted, the nonequilibrium phonons that are created during the thermalization of the electrons have a frequency on the order of the Debye frequency. The phonons of the thermal background can be neglected. The nonequilibrium phonons have small occupation numbers. With these assumptions, it is clear that the main type of phonon–phonon interaction is phonon decay.

We neglect the scattering of phonons by impurities, and also the leakage of phonons through the boundary, which is possible in the cooling of a film without a substrate in helium gas. Elastic scattering of phonons occurs principally on the film surfaces. Since elastic scattering can occur with conversion of the polarization of the phonons, frequent scattering on the surfaces leads to rapid "mixing" of the polarizations. Therefore, instead of the system of kinetic equations, we can consider one equation for the phonons of the decay mode:

$$\frac{\partial n}{\partial t} + \mathbf{v}\nabla n = -\frac{n}{\tau_l} + \int_\omega^\infty d\omega' \rho(\omega') P(\omega' \to \omega) \bar{n}(\omega'). \tag{3}$$

Here, $n(\omega, \mathbf{r}, z, \mathbf{e}, t)$ is the distribution function of the phon-

 0038-5646/92/020286-03$05.00

ons, $\mathbf{r}$ is the position vector, lying in the plane of the film, $\mathbf{e}$ is the unit vector of the direction of the phonon monentum, and $\bar{n}$ is the distribution function averaged over the directions of the vector $\mathbf{e}$. The integral term in Eq. (3) describes the arrival of phonons at the level ω in the decay of phonons of higher frequencies, and has the same form if we make the simplifying assumption that the integral kernel is independent of the directions of the phonon momenta; $\rho(\omega)=3\omega^2/2\pi^2v^3$ is the density of states of the phonons; $P(\omega'\to\omega)$ is the kernel associated with the probability of phonon decay, and can be represented conveniently in the form $P(\omega'\to\omega)=h(\omega/\omega')/\tau_l(\omega')\rho(\omega')\omega'$, where the function $h(x)\sim 1$ for $x\sim 1$. The phonon-decay frequency has the form

$$1/\tau_l(\omega)=\gamma\omega_D(\omega/\omega_D)^5, \tag{4}$$

where γ is a parameter that depends on the rate of anharmonic decay.

We seek the distribution function of the phonons in the following form:

$$n(\omega, \mathbf{r}, z, \mathbf{e}, t)=n_0(\omega, \mathbf{r}, t)+\delta n(\omega, \mathbf{r}, z, \mathbf{e}, t), \tag{5}$$

assuming that its isotropic part n_0 is independent of z because of the uniformity of the heating over the depth of the film and the absence of leakage of phonons through the boundary.

On the function (5) we impose two restrictions. The first is that the energy density of the phonons is completely determined by the isotropic part n_0. For an isotropic spectrum $\omega(\mathbf{q})$, this condition leads to the expression

$$\int do\delta n(\omega, \mathbf{r}, z, \mathbf{e}, t)=0, \tag{6}$$

where do is a solid angle in the direction of $\mathbf{e}$. The second restriction is that the boundary conditions for complete diffuse reflection of phonons from the surface of the film have the form

$$\delta n(z=\mp d/2,\ e_z\gtrless 0)=0, \tag{7}$$

where $z=\pm d/2$ are the film boundaries, $e_z=\cos\theta$, and θ is the angle between the direction of the phonon momentum and the z axis.

Equation (3) can be solved by making a Fourier transformation $\mathbf{r}\to\mathbf{k}$ and a Laplace transformation $t\to\lambda$. We obtain

$$v_z\frac{\partial\delta n}{\partial z}+\left(\lambda+i\mathbf{k}\mathbf{v}_r+\frac{1}{\tau_l}\right)\delta n=-\left(\lambda+i\mathbf{k}\mathbf{v}_r+\frac{1}{\tau_l}\right)n_0 +\int_\omega^\infty d\omega'\rho(\omega')P(\omega'\to\omega)n_0(\omega'), \tag{8}$$

where $\mathbf{v}_r$ is the component of the phonon velocity in the plane of the film. The solution of Eq. (8), with the condition (7) and averaged over z to give $\overline{\delta n}=\int_{-d/2}^{d/2}\delta n dz/d$, has the form

$$\overline{\delta n}=\frac{I(\omega)-(\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r)n_0(\omega)}{\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r}\left\{1-\left[1-\exp\left(-\frac{\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r}{v|\cos\theta|/d}\right)\right]\frac{v|\cos\theta|}{d(\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r)}\right\}, \tag{9}$$

where

$$I(\omega)=\int_\omega^\infty d\omega'\rho(\omega')P(\omega'\to\omega)n_0(\omega'), \qquad \mathbf{k}\mathbf{v}_r=kv\sin\theta\cos\varphi,$$

where φ is the angle between the vectors $\mathbf{k}$ and $\mathbf{v}_r$.

Next, we must substitute the expression (9) into the condition (6) and integrate it over the angles. As a result, we obtain an equation for the isotropic part $n_0(\omega)$ of the distribution function. It is not possible to integrate the expression (9) analytically. Since all angles are important in the integration, we interpolate (9) by the following function, which coincides with (9) at the angles $\theta=0$ and $\theta=\pi/2$:

$$\overline{\delta n_i}=\frac{I(\omega)-(\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r)n_0(\omega)}{2v|\cos\theta|/d+\lambda+1/\tau_l+i\mathbf{k}\mathbf{v}_r}. \tag{10}$$

A detailed numerical analysis of the functions (9) and (10) shows that the maximum deviation of $\overline{\delta n_i}$ from $\overline{\delta n}$ for any angles θ and φ amounts to no more than 10% of the function $\overline{\delta n}$.

Substituting (10) into the condition (6) and taking into account the inequalities assumed above, we obtain an equation for the function $n_0(\omega)$:

$$\frac{\tau_l[I(\omega)-(\lambda+1/\tau_l)n_0]}{1+\lambda\tau_l}\ln\frac{2\gamma_0}{1+\{1+[kv\tau_l/(1+\lambda\tau_l)]^2\}^{1/2}} =n_0\left\{\left[1+\left(\frac{kv\tau_l}{1+\lambda\tau_l}\right)^2\right]^{1/2}-1\right\}, \qquad \gamma_0=\frac{2v}{d(\lambda+1/\tau_l)}. \tag{11}$$

We take into account the explicit form of the expression for $I(\omega)$ and express the kernel $P(\omega'\to\omega)$ in terms of $h(\omega'/\omega)$, and also introduce new variables $x=\omega/\omega'$, $\xi=\hbar v\tau_l(\omega)$, and $\eta=\lambda\tau_l(\omega)$. We seek the self-similar solution of Eq. (11) in the form

$$n_0(\omega, k, \lambda)=A\omega^\alpha F(\xi, \eta), \tag{12}$$

where A and α are arbitrary constants. The equation for the function $F(\xi, \eta)$ has the form

$$\frac{1}{1+\eta}\left[\int_0^1 dx x^{-\alpha-6}h(x)F(\xi x^5, \eta x^5)-(1+\eta)F(\xi, \eta)\right] \times\ln\frac{2\gamma_0}{1+\{1+[\xi/(1+\eta)]^2\}^{1/2}}=F(\xi, \eta)\left\{\left[1+\left(\frac{\xi}{1+\eta}\right)^2\right]^{1/2}-1\right\}. \tag{13}$$

The large parameter γ_0 in (13) is under the logarithm. Thus, Eq. (13) essentially contains no large or small parameters. Consequently, the self-similar function $F(\xi, \eta)\sim 1$ for ξ, $\eta\sim 1$. By making the inverse Fourier and Laplace transformation on (12), we obtain

$$n_0(\omega, r, t)=A\omega^\alpha f\left(\frac{r}{v\tau_l(\omega)}, \frac{t}{\tau_l(\omega)}\right). \tag{14}$$

The quantity $\bar{r}$, defined by the relation $\bar{r}/v\tau_l(\omega)\sim 1$, is the characteristic length scale of the phonon nonequilibrium, while the relation $t/\tau_l(\omega)\sim 1$ determines the characteristic frequency ω of the phonon generation at time t.

By estimating (10) in the range of angles $|\cos\theta|<(\lambda+1/\tau_l)/v/d$ (i.e., near $\theta=\pi/2$), we discover that $\overline{\delta n}\sim n_0$, and this confirms our assumption that it is not possible to use the diffusion equation.

CONCLUSION

Thus, we have ascertained that, if a phonon hot spot is not established, the distribution function of the nonequilibrium phonons has the form (14). By calculating the energy density of the phonons with the aid of (14), one can easily show that the characteristic length scale of the phonon nonequilibrium has the form $\bar{r} = vt$. Over the characteristic lifetime $\tau_l(\omega)$ of the phonon generation, as a result of diffuse scattering at the boundaries, there is an accumulation of a sufficient number of phonons moving ballistically almost parallel to the film surfaces. The number of such phonons does not depend on the thickness of the film. With decrease of d, the solid angle within which the energy-transporting phonons can propagate ballistically decreases, but this decrease is balanced by the increase of the frequency of diffuse scattering at the boundaries, as a result of which phonons can be scattered into this solid angle.

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Translated by P. J. Shepherd