Kinetics of the emission of subterahertz acoustic phonons from the phonon hot spot in GaAs crystals

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In the initial stage of the existence of the phonon "hot spot," which is produced near the surface of a GaAs crystal by laser pulses with an intensity 1 mJ/mm^2, the flux density of acoustic phonons with frequencies in the range 0.3–0.6 THz out of the hot spot increases slowly over time. Possible reasons for the effect are discussed.

Intense optical interband pumping of semiconductors at a low temperature, which is always accompanied by a radiationless relaxation of part of the electronic-excitation energy, gives rise to a region of a so-called phonon hot spot near the surface of the sample. In this hot spot, the phonon occupation numbers are far higher than the equilibrium values.[1] The hot spot persists for a comparatively long time ($\sim 1\ \mu$s) after the optical pumping is ended, and it serves as a source of nonequilibrium acoustic phonons, which are emitted into the cold crystal.

In the present letter we report an experimental study of the kinetics of the emission of subterahertz acoustic phonons (which are "subthermal" with respect to the temperature of the spot) from a hot spot, for the particular example of a hot spot in GaAs. In contrast with other studies of the propagation of phonons produced by photoexcitation of GaAs which have been reported,[2–5] we use a very high level of optical pumping (the energy in the pulse is $\sim$ 1 mJ/mm^2), which undoubtedly produces a hot spot near the surface. A selective detector which detects monochromatic phonons has been used for the first time in research on hot spots, to detect the acoustic phonons emitted by the hot spot.

In the experiments (Fig. 1) we use p-type GaAs ($n_p = 10^{17}$ cm^{-3}) wafers of thickness $d_0 = 0.4$ mm, oriented in the (001) plane and immersed in liquid helium

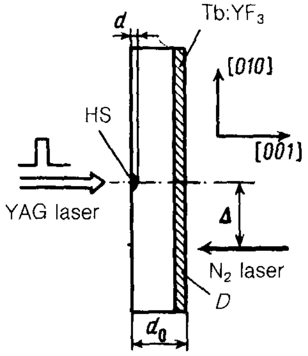

FIG. 1. Experimental arrangement.

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($T_0 = 1.8$ K). The samples are excited by the second-harmonic pulses from a Nd:YAG laser ($\lambda = 530$ nm). The size of the illuminated spot is 0.3 mm^2; the light penetration depth is ~ 1 μm; and the pulse length is 2×10^{-7} s. Under these conditions we would expect[6] the attainment of an extremely high temperature, up to $T \gtrsim 1000$ K, in the hot spot produced by the laser pulse. This expectation is verified directly by the observation of damage to the surface of the GaAs by a series of pulses.

The phonon pulse induced in the sample by the laser pulse is detected at the opposite face of the sample by a contact luminescent phonon spectrometer[7]: a polycrystalline Tb:YF$_3$ film excited locally by a nitrogen laser (the spot area D is 0.1 mm^2). The phonons are detected through measurement of the behavior of the intensity of two spectral lines of the luminescence of the film, $I_1(t)$ and $I_2(t)$, which reflects the time evolution of the density of nonequilibrium phonons of the resonant frequencies $\omega_1 = 0.3$ and $\omega_2 = 0.6$ THz, respectively, in the part of the GaAs sample adjacent to area D.

Figures 2, (a) and (b), shows some typical pulses of the luminescence of the $I_1(t)$ detector, induced by the optical-pumping pulses and measured at the same energy, $W = 1$ mJ/mm^2, but for different positions (Δ) of detector D with respect to the hot spot (Fig. 1). In the case $\Delta = 0$, in which D is positioned "opposite" the hot spot, at the shortest distance D_0 along the [001] line, the $I_1(t)$ pulse has a very steep ($\sim 10^{-7}$ s) leading edge [Fig. 2(a)]. As D is moved, and the line from the hot spot to

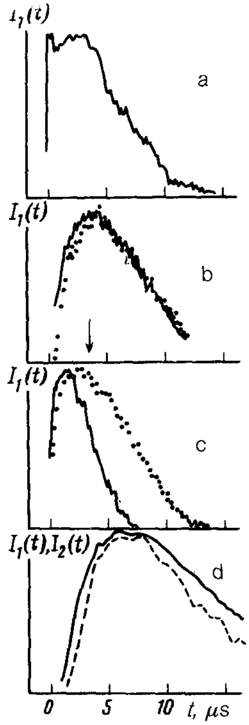

FIG. 2.Normalized luminescence pulses $I_1(t)$ (a, b, c) and $I_2(t)$ (d). a—$W = 1$ mJ/mm^2, $\Delta = 0$; b—$W = 1$ mJ/mm^2, $\Delta = 0.5$ mm (solid line), $\Delta = 2.5$ mm (dotted line); c—$\Delta = 0.5$ mm, $W = 1$ mJ/mm^2 (dotted line), $W = 0.2$ mJ/mm^2 (solid line); d—$\Delta = 2.5$ mm, $W = 2$ mJ/mm^2. Solid lines: $I_1(t)$. Dashed lines: $I_2(t)$. In part b, the time $\bar{t}$ is shown by the vertical arrow.

D deviates from the [001] direction—which is the direction for the focusing of TA phonons in gallium arsenide[3,5]—by 10–15°, the leading edge of $I_1(t)$ turns out to be stretched out markedly over a time $\bar{t}\approx 5$ μs [Fig. 2(b)]. As Δ is increased, the distance $x=\sqrt{\Delta^2+d_0^2}$ from the hot spot to D increases, and the amplitude of $I_1(t)$ decreases, the time $\bar{t}$ remains essentially constant [Fig. 2(b)]. We observe a parallel transport of the front for a time $\delta\ t$, which is approximately equal to the ballistic-transit time $\delta x/U_L$, where δx is the change in x upon a displacement of the detector, and $U_L=5\times10^5$ cm/s is the velocity of the long-wave LA phonons. The leading edge of the $I_2(t)$ pulses is delayed slightly with respect to the $I_1(t)$ front [Fig. 2(d)].

The leading edge of the luminescence pulses $I_1(t)$ and $I_2(t)$ reflects the flux density to the detector of nonequilibrium phonons at 0.3 and 0.6 THz which are produced in the GaAs crystal upon the formation of a hot spot in it. This leading edge is determined by both the dynamics of the phonon emission and the time delay of the phonons over the path from the hot spot to detector D. For directions between the hot spot and D which are not along the [001] direction, the behavior of the $I_1(t)$ leading edge [Fig. 2, (b) and (c)] indicates a substantial prolongation of the arrival of phonons at the detector, far longer than the length (0.2 μs) of the laser pulse which forms the hot spot. The independence of the prolongation time $\bar{t}$ from the distance (x) to D means that we cannot attribute this delay to a diffusion regime of the propagation of phonons from the hot spot to the detector. That regime should give rise to a very strong $t\sim x^2$ dependence of the delay on the distance from the hot spot to D. Consequently, the leading edge of $I_1(t)$ and $I_2(t)$ describes the dynamics of the emission of phonons in the frequency range 0.3–0.6 THz by the hot spot. These phonons then reach D ballistically (as follows directly from the parallel shift of the front with increasing x over the ballistic delay time; Fig. 2(b).[1)]

The observed elongation of the leading edge of $I_1(t)$ and $I_2(t)$ over 3–5 μs is therefore evidence of a slow increase in the flux density of subterahertz phonons, in the range 0.3–0.6 THz, out of the hot spot in the initial stage of its existence. This increase occurs *after* the end of the optical pumping. To explain this unexpected effect—an increase in the flux density of phonons out of the cooling spot—we should take into account the circumstance that the ballistic flux density of subterahertz (ω) phonons out of the hot spot is proportional to the product $\bar{n}_\omega\ V$, where $\bar{n}_\omega$ is the density of phonons, and V is the size of that ("emitting") part of the volume of the hot spot from which phonons can escape into the cold crystal without scattering. Since the phonon density $\bar{n}_\omega$ always decreases as the hot spot cools off, the very observation of an increase in the phonon flux over time is evidence that as the hot spot cools off, its size V increases, and this increase occurs more rapidly than the decrease in the temperature T. If we compare the emitting volume V with the entire volume of the hot spot, we find that the situation which we need can be realized, under certain conditions, in the hot-spot model of Ref. 6, in which a localizing of the spot stems from a strong Rayleigh scattering of phonons by defects, and the cooling of the hot spots stems from its expansion, while the total energy of the phonons in the hot spot is conserved.[2)]

Under our conditions of a very strong heating of the hot spot, an increase in the emitting volume could in principle also occur because of another factor, which is closely related to the role of phonon-phonon scattering. Specifically, this scattering causes the mean free path of the subterahertz phonons, $\bar{l}$, in the hot spot to be very small at high temperatures. An extrapolation of data on the attenuation of ultrasound at 10 GHz at $T = 60$ K on the basis of the formula[8] $\bar{l}^{-1} \sim \omega T^4$ leads to the value $\bar{l} \sim 10^{-5}$ *cm*$^{-1}$ *at* $T = 300$ K for phonons at 0.3 THz. This value is an order of magnitude smaller than the initial thickness of the hot spot, $d \sim 1$ μm. Consequently, phonons with frequencies 0.3–0.6 THz cannot escape ballistically from the central part of the hot spot, heated to $T = 1000$ K. These phonons are emitted predominantly from the peripheral region, with a temperature which falls off gradually with distance from the center. The effective volume (V) of this emitting region generally increases during the cooling of the hot spot because of the increase in $\bar{l}$, which leads to an increase in the phonon flux density. In this model we also see an explanation for the observed features in the increase in the phonon flux density: 1) the decrease in $\bar{t}$ with decreasing pump power [Fig. 2(c)], which may be due to a decrease in the hot-spot cooling time as the initial temperatrue of the hot spot decreases; 2) a retardation of the flux density of phonons at 0.6 THz in comparison with that at 0.3 THz [Fig. 2(d)], which may be due to the frequency dependence $\bar{l}^{-1} \sim \omega$. We might add that the emission of subterahertz phonons would generally be a reason for a decrease in the phonon energy of the hot spot, affecting its evolution.

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1) Further evidence for a ballistic propagation of phonons at 0.3 and 0.6 THz in these samples comes from the observation of a ballistic front of $I_1(t)$ and $I_2(t)$ during the propagation of *TA* phonons along the direction in which they are focused [Fig. 2(a)] and also from the results of experiments with heat pulses, in which the source of phonons at 0.3 and 0.6 THz was a film heater on the surface of GaAs.

2) This circumstance was pointed out by I. B. Levinson.

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