Ballistic propagation of phonons in thin anisotropic plates (anthracene)

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The density of phonons incident on a planar geometry detector is calculated, with account taken of the anisotropy of a crystalline anthracene plate. It is shown that the minimum times of flight satisfactorily agree with the times observed experimentally for different groups of nonequilibrium phonons.

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1. INTRODUCTION. FEATURES OF THE PLANAR GEOMETRY

Several works have appeared recently[1-3] on the propagation of thermal pulses in solids with a planar geometry of the experiment. In the usual situation (Fig. 1a), the dimensions of both the source and the detector are small in comparison with the distance between them, i.e., with the dimensions of the crystal. This means that the direction of propagation of the phonons which are recorded by the detector is known beforehand. In the most general case, there are several groups of such phonons. The situation is entirely different in the case of planar geometry (Fig. 1b). Here, at $D \gg d$, phonons reach the detector from arbitrary directions. Therefore the problem—to predict theoretically the shape of the response for a given shape of the excitation pulse—is much more complicated. It is also not clear initially which phonons first reach the detector and when this happens.

There exist many different types of phonon detectors: thermal bolometers, superconducting films, optical detectors. In a rough approximation, they all respond to the total phonon density in the region of the detector. We shall be concerned with the analysis of this important characteristic in the case of planar geometry and an anisotropic crystal.

2. PHONON DENSITY AT THE DETECTOR

Let a certain quantity of phonons be generated at the left wall of a crystal, which serves as a heater (H), in the form of a pulse that is delta-shaped in time. Let the distribution function of these phonons by $\Phi_j(\mathbf{k})$, where $\mathbf{k}$ is the quasimomentum and j characterizes the phonon branch, the consideration being limited to the three acoustical branches. How does the total phonon density depend on the time in this case at the detector, i.e., at the right wall? For simplification of the problem, we neglect reflection of the phonons from the walls, i.e., we limit ourselves to the first arrival of phonons at the rear wall. Physically, this corresponds to an absolutely matched detector, which absorbs all the phonons incident on it. This part of the phonon density—the response function $G(t)$—is given by the expression

$$G(t)=\sum_j \int d^3k\,\delta(t-\tau_j(\mathbf{k}))\Phi_j(\mathbf{k}). \tag{1}$$

Here we have introduced the function $\tau_j(\mathbf{k})$—the time of arrival at the rear wall of the phonon of the jth branch with wave vector $\mathbf{k}$. The physical meaning of Eq. (1) is obvious; the phonon makes a contribution to the density at the same instant when it reaches the rear wall.

It is not possible for a specific case to use Eq. (1) to calculate the function $G(t)$. This is first of all connected with the fact that in many experiments we know almost nothing about the initial phonon distribution function $\Phi_j(\mathbf{k})$. Therefore, assuming $\Phi_j(\mathbf{k})$ to be not too different from isotropic and a sufficiently smooth function (in experiments of Refs. 2 and 3, there was no evidence that this was not so), we shall seek the more important features of the function $G(t)$. Here we limit ourselves to small $\mathbf{k}$, i.e., $|\mathbf{k}| \ll k_B$, the Brillouin wave vector. Then, as is well known,[4] the group velocities of the phonons, and consequently, $\tau(\mathbf{k})$ also, depend only on the direction $\mathbf{n}$ of the wave vector, but not on its magnitude. Then only integration over $\mathbf{n}$ remains in (1) and it is seen that the features of $G(t)$ are connected with the points at which the first derivatives of the $\tau_j(\mathbf{n})$ with respect to $\mathbf{n}$ vanish. These are the maxima, minima, and saddle points.

It is not difficult to see that at $t=\tau_0^j$, where t_0^j is any of the minima of the function $\tau_j(\mathbf{n})$, the response $G(t)$ undergoes a finite positive jump. For this, it is sufficient to expand $\tau_j(\mathbf{n})$ in a series in the region of the minimum up to terms of second order and to carry out the integration in (1) over n in explicit form. At points of the maxima, jumps with the opposite sign are encountered similarly. The saddle points lead to logarithmic singularities of the form $-\ln|t-\tau_0^j|$, τ_0^j is the value of $\tau_j(\mathbf{n})$ at the saddle point.

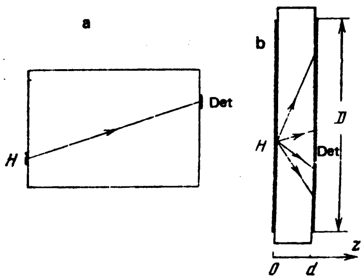

FIG. 1. The usual (a) and plane (b) geometry of the experiments. Det—detector, H—heater (source).

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In the case of a pulse with the form $g(t)$ of finite duration t_p, we have, in place of (1),

$$S(t)=\sum_j \int d^3k\, g(t-\tau_j(\mathbf{k}))\,\Phi_j(\mathbf{k}). \qquad (2)$$

We now denote by Δ the absolute value of the jump at the minima and maxima. We denote by this same letter the coefficient of the logarithm at the saddle points. Let $G^{*\prime}(t)$ be the derivative of the smooth part of the function $G(t)$ in the region of the singularity. At not too large t_p, and in particular at $t_p \lesssim \Delta/|G^{*\prime}|$, the function $S(t)$ will have inflection points near the extremum points of $\tau_j(\mathbf{n})$, and maxima near the saddle points. In order that the singularities of $S(t)$ be observed separately, it is necessary that $t_p \lesssim \Delta\tau$, where $\Delta\tau$ is the distance between the corresponding singularities of $\tau_j(\mathbf{n})$. To find the functions $\tau_j(\mathbf{n})$ in which we are interested, it is natural to use the well known[4] procedure of calculation of the group velocities from the elastic constants of the crystal.

3. THE ANTHRACENE CRYSTAL

Further calculations were carried out for the anthracene crystal in the geometry of the experiments of Refs. 2 and 3. In this geometry, our choice of Oz perpendicular to the plane of the plate (Fig. 1b) corresponds to the standard choice of axes in the anthracene crystal (Fig. 2).

To determine $\tau_j(\mathbf{n})$ with a computer (M-222) we used the elastic constants of anthracene from the work of Lutz.[5] To put the results of the calculations in convenient form, we choose the usual spherical set of coordinates for $\mathbf{n}$: θ is the angle between Ox_3 and $\mathbf{n}$, and φ is the angle between Ox_1 and the projection of $\mathbf{n}$ on the plane Ox_1x_2 and is measured in the direction toward Ox_2. Each vector $\mathbf{n}$ of the hemisphere $\theta > 0$ can be represented by a point on such a diagram (Fig. 3): the angle φ is plotted in explicit fashion; in place of θ we introduce the radius $r = 2\sin^2(\theta/2)$. Such a diagram is convenient in that it preserves the area of the hemisphere.

The angles θ and φ were scanned with variable intervals such that the points filled the area of the diagram uniformly. Owing to the presence of a plane of symmetry perpendicular to Ox_2, it suffices to investigate the semicircle $0 \leqslant \varphi \leqslant 180°$. The original picture contains 630 points on the semicircle (the computer calculation time of a single point amounted to 10 sec). The

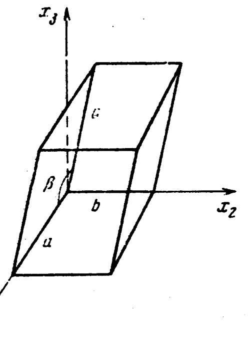

FIG. 2. Unit cell of anthracene (crystal of monoclinic syngony). Parameters: $\beta = 124.7°$, $a = 8.56$ Å, $b = 6.94$ Å, $c = 11.6$ Å. The plane ab is the cleavage plane.

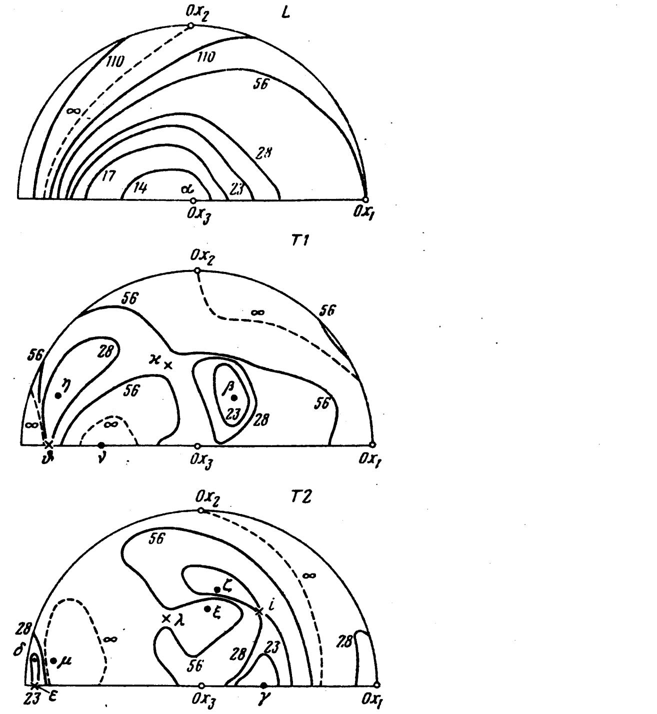

FIG. 3. Map of the time of arrival for the quasi-longitudinal (L) and two quasi-transverse ($T1$, $T2$) branches. The continuous curves connect points with the same times of arrival (in nanoseconds, for a crystal of thickness 45μ), denoted by the numbers at the lines. The dashed curve indicates points $\tau_j(n) = \infty$. The maxima and minima are denoted by the boldface points, the saddle points by the crosses. All the singularities are identified by Greek symbols. This notation is repeated in Fig. 4 and the table.

regions of the singularities were investigated in more detail, with the intervals 2–3 times smaller.

The values obtained for τ_0^j are given in the table. For illustration, Fig. 3, shows the diagrams for all three acoustic branches, as well as the lines of equal arrival times and the locations of the singular points.

4. COMPARISON WITH EXPERIMENT

Figure 4 shows the experimental curve from Ref. 3. It shows also the values τ_0^j of the singular points of the function $\tau_j(\mathbf{n})$ as calculated in the present work. It is seen that the minimum α on the quasi-longitudinal branch

TABLE I. Singular points* of the function $\tau_j(\mathbf{n})$

Designation of the singular point	Phonon branch	Value of τ_0^j nsec for crystal with $d=45\mu$	Character of the singular point
α	L	12.8	Minimum
β	T1	20.2	»
γ	T2	20.2	»
δ	T2	22.2	»
ε	T2	22.5	Saddle
ζ	T2	23.2	Minimum
η	T1	23.3	»
ϑ	T1	27	Saddle
ι	T2	27	»
κ	T1	46	»
λ	T2	58	»
μ	T2	105	Minimum
ν	T1	124	»
ξ	T2	147	Maximum

*The designations of the singular points α, β etc. correspond to those of Figs. 3 and 4.

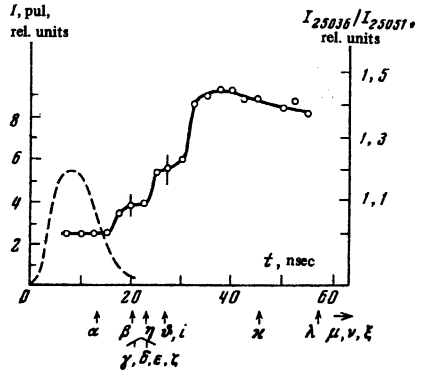

FIG. 4. Experimental curve of the temperature-sensitive ratio of the intensities of two lines in the anthracene spectrum.[3] The time t is measured from the beginning of the excitation pulse. Below the arrows, the times corresponding to the found singular point $\tau_j(n)$ are marked by Greek symbols. The values of $\tau_0{}^j$ for the singular points μ, ν, ξ are beyond the limits of the experiment (the thickness of the crystal $d=45\mu$); the dashed curve shows the shape of the pump pulse.

corresponds to the first arrival of phonons at the detector. A small delay (≈3 nanosec) of the experimental step relative to the calculated value τ_0^j, corresponding to the point α, can be attributed to the duration of the process of generation of longwave phonons from excitons via shortwave phonons.[3] With account of this delay, the second group of arriving phonons (the second step) can be connected with the series of singular points from β to η (minima and one saddle). The points of this series are not resolved experimentally because of the long duration of the excitation pulse.

The third step is close in time (with account of the delay) to the saddles ϑ and i. However, it is scarcely connected with them. The point is that the experimentally measured signal is proportional to the temperature, i.e., the fourth root of the phonon concentration. Therefore the last step corresponds to the arrival of a substantially larger number of phonons than the first two. Yet the singularities of ϑ and i are not separated in the calculation against the background of the other singularities.

There exist two possibilities of explanation of the last arrival. The first is as follows. This arrival is connected with the hydrodynamic propagation,[3] and the first two with the acoustic precursors, i.e., with the phonons which avoid phonon-phonon collisions. The number of such phonons is $\sim\exp\{-d/l\}$, where l is the free path length relative to phonon-phonon processes; it is entirely possible that this number is not very small (the exact value of l is unknown). The second possibility can be connected with the fact that the thickness of the crystal is not sufficient to let the bulk of the decay products reach the acoustic limit in the process of spontaneous decay of the shortwave phonons.[3] Then, because of the dispersion of the group velocity, the bulk of the phonons arrive with effective velocities slower than acoustic. However, in the decay process, a small (power-law) number of longwave phonons is produced and these play the role of precursors and cause the first two steps of the experimental curve.

It is not possible to make a direct choice between the two proposed variants. In any case, however, the first two steps are well explained by the acoustical precursors.

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