# Detection of nonequilibrium phonons by means of resonance Raman scattering (RS) of light

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A method is proposed for optical detection of nonequilibrium acoustic phonons by means of resonance Raman scattering of light. The ballistic-propagation velocities of acoustic phonons along the **a** and **b** axes of an anthracene crystal are determined experimentally.

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Optical methods of nonequilibrium-phonon detection are widely used in experiments on the propagation of high-frequency ($10^{11}$–$10^{12}$ Hz) phonons in dielectric and semiconductor crystals. When high-frequency phonons are generated by thermal pulses, the arrival of different phonon groups can be detected from the variation of the optical properties of impurity centers[1,2] and from the time-resolved, intrinsic-luminescence spectra of crystals.[3] In this paper we report the possibility of using resonance Raman scattering of light (RRSL) as a phonon detector.

The experiments on phonon detection by means of RRSL were performed using thin ($d \approx 5$–$10$ $\mu$m), single-crystal anthracene plates. As shown in Ref. 4, a sharp increase of the intensity ($I$) of the RRSL by the intramolecular vibration with a frequency $\Omega = 1402$ cm$^{-1}$ can be observed as a result of excitation of anthracene crystals near the bottom of the lowest exciton band ($\nu_0 = 25097$ cm$^{-1}$). It was determined that the value of $I$ was very sensitive to the crystal temperature and to optical pumping. These characteristics, which are observed in the stimulated RRSL regime, were used for detecting nonequilibrium phonons.

The experiment was as follows. The front surface of the crystal ([001] plane) was excited by flashes from a nitrogen laser: $\lambda = 3371$ Å, flash duration $\tau_p = 4$ nsec, repetition rate 25 Hz, and optical pumping intensity $P \approx 50$ kW/cm$^2$. The pumping spot on the crystal surface was formed in the shape of a narrow strip with a width of $\sim 10$ $\mu$m and a length of $\sim 2$ mm ($P$, see Fig. 1). The nitrogen-laser radiation was absorbed in a thin surface layer $K^{-1} \approx 0.5$ $\mu$m. The nonequilibrium phonons, which are generated in this region as a result of fast, radiationless processes, are transmitted to the unexcited parts of the crystal.[3]

RRSL was excited by means of a tunable dye laser: tuning region $\nu_{exc} = 24000$-$26000$ cm$^{-1}$, lasing band half-width $\Delta\nu_{exc} = 1$ cm$^{-1}$, flash duration $\tau_p = 3$ nsec, and a repetition rate 25 Hz. The tunable-laser flashes were delayed with respect to the nitrogen-laser pumping flashes by an amount $t_D = 54$ nsec. The illumination spot on the crystal surface ($D$, see Fig. 1) had the same dimensions as the pumping spot, and they were placed parallel to each other. The optical system made it possible to move the pumping spot along the crystal surface, and have to smoothly change the distance $l$

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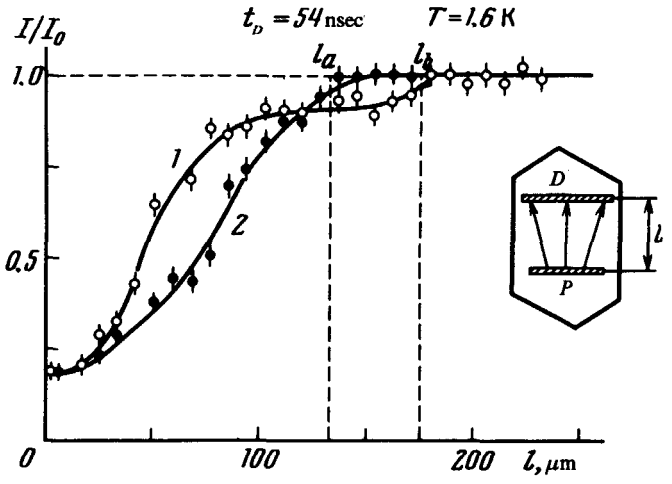


FIG. 1.

between the $P$ and $D$ regions (see Fig. 1). In addition, we could change the position of the $P$ and $D$ spots in the [001] plane relative to the crystallographic directions.

The RRSL intensity at the frequency $\nu = \nu_{exc} - \Omega$ was measured by means of a DFS-12 monochromator with a standard photoelectric recording system. The arrival of nonequilibrium phonons was detected from the change of the RRSL signal.

Figure 1 shows the results of our experiment. The anthracene crystal with a thickness $d \approx 10\,\mu$m was immersed into superfluid helium at a temperature $T = 1.6$ K. The frequency of the exciting RRSL radiation of a tunable laser is $\nu_{exc} = 25085$ cm$^{-1}$ (i.e., $\nu_0 - \mu_{exc} = 12$ cm$^{-1} = 3.6 \times 10^{11}$ Hz) and $I_0$ is the RRSL intensity in the absence of the nitrogen-laser pumping flashes. Curves 1 and 2, which were obtained by orienting the $P$ and $D$ spots along the **a** and **b** crystal axes, respectively, correspond to the relative variation $I/I_0$ of the RRSL signal as the distance $l$ between the heater and the nonequilibrium-phonon detector is varied. As seen in Fig. 1, the arrival of the fastest phonon group can be observed at distances $l_b = 175\,\mu$m and $l_a = 133\,\mu$m for a specified delay time $t_D = 54$ nsec; this corresponds to maximum propagation velocities $v_{\mathbf{b}}^{max} = (0.2 \pm 0.2) \times 10^5$ cm/sec and $v_{\mathbf{a}}^{max} = (2.5 \pm 0.2) \times 10^5$ cm/sec along the **b** and **a** axes.

Calculations similar to those in Ref. 5 were performed by Kazakovetsev for ballistic propagation of acoustic-branch phonons. The results of the calculation show that the phonons of the quasi-longitudinal branches have the maximum propagation velocities along the **a** and **b** axes, and their numerical values $v_{\mathbf{a}}^{max} = 2.45 \times 10^5$ cm/sec and $v_{\mathbf{b}}^{max} = 3.05 \times 10^5$ cm/sec are in good agreement with the experimental data.

Thus, the use of the RRSL method of detecting nonequilibrium phonons makes it possible to study the phonon propagation along the developed surface of thin, single-crystal plates. Such experiments are difficult to perform by using fluorescence detectors. This method has good spatial ($\sim 10\,\mu$m) and time resolution. Moreover, because

of isolation of the RRSL band in the radiation spectrum of the anthracene crystal and because of its high intensity, there is no need to use the photoelectric recording method with high time resolution and sensitivity, which greatly simplifies the experiment.

The operating principle of the RRSL detector is as follows. The arrival of nonequilibrium phonons in the detection region increases the absorption of the RRSL exciting laser radiation with a frequency $\nu_{exc}$. As a result, the RRSL signal decreases markedly because of the critical superlinear dependence of the RRSL intensity on the excitation level in the stimulated light-scattering regime. There is reason to assume that the formation of a long-wave absorption wing near the bottom of the exciton band of an anthracene crystal is primarily due to one-phonon processes. It is easy to see that in this case the phonons detected have frequencies $\omega_{ph} \gtrsim \nu_0 - \nu_{exc}$. By varying the $\nu_{exc}$ value by means of a tunable dye laser, we can record the arrival of phonons with different energies. Thus, the detection of nonequilibrium phonons by means of RRSL can be used, in principle, as a phonon-spectroscopy method in the $10^{11}$ to $10^{12}$-Hz frequency region. Experimental studies in this direction are now underway.

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