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[1]A. T. Lubchenko and B. P. Pavlik, Fiz. Tverd. Tela 5, 1714 (1963) [Sov. Phys. Solid State 5, 1246 (1964)]; A. K. Ganguly and J. L. Birman, Phys. Rev. 162, 806 (1967).

[2]J. F. Scott, R. C. C. Leite, and T. C. Damen, Phys. Rev. 188, 1285 (1969). See also references quoted herein.

[3]E. Mulazzi, in preparation. The theory is valid for large-radius excitons whose lifetime γ is larger than the phonon relaxation time. The different contributions coming from Raman effect, hot luminescence, and their interference [V. Hizhnyakov and I. Tehver, Phys. Status Solidi 39, 67 (1970)] will be discussed by considering also the change of the phonon frequencies in the exciton state due to the exciton phonon coupling. In a previous publication [see E. Mulazzi, Phys. Rev. Lett. 25, 228 (1970)] the two effects, Raman scattering and hot luminescence, have not been discussed in a separate way.

[4]R. Z. Bachrach and F. C. Brown, Phys. Rev. B 1, 818 (1970).

[5]E. R. Cowley and A. Okazaki, Proc. Roy. Soc., Ser. A 300, 45 (1967).

[6]Mulazzi, second paper of Ref. 3.

[7]Because of the nearness to the fundamental absorption region, the ordinary second order spectra for both TlBr and TlCl were found to have an intensity about one order of magnitude higher than for alkali halides in the same experimental circumstances. This higher intensity could be very profitable in studies in which high precision and high resolution are important, such as the search for critical-point singularities in two-phonon density of states.

NONLINEAR PROPAGATION OF HEAT PULSES IN SOLIDS

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We have observed self-steepening and narrowing of transverse heat pulses in pure NaF at high input-pulse powers when the ambient temperature is in the ballistic regime of the thermal-conductivity curve. The details of such nonlinear propagation of heat are found to be in agreement with the theory proposed by Tappert and Varma, which shows that under certain conditions heat pulses develop an instability.

In comparison with other systems in which nonlinear phenomena have been observed, the nonlinear parameters for the vibrational properties of solids are quite large. For example, for a typical insulating crystal the quantity $\epsilon = a_3 h / a_2$, where a_2 and a_3 are the second (harmonic) and third derivatives, respectively, of the interatomic potential and h is the lattice spacing, is of order unity. However, it is usual to linearize the equations of motion for the atoms in the solid and obtain for example a diffusion equation or a wave equation for propagation of heat in solids. Experimentally, the vibrational properties of solids are studied either at very low frequencies, where the available power input is insufficient to drive the nonlinearities, or by studying propagation of heat by introducing a differential increase of temperature. By the second method the phenomena of thermal conductivity, second sound, and ballistic propagation of heat have been studied. In this paper we describe experiments in which we have observed propagation of heat under conditions in which the nonlinearity and dispersive nature of the vibrational properties of solids is dramatically illustrated.

Recently a great deal of mathematical and numerical-analytic work has been devoted to the study of nonlinear dispersive waves and a variety of interesting effects are predicted. The work relevant to our experiment, and its extension to cover our special experimental situation, is discussed in the succeeding paper.[1]

Our experiment is performed in the ballistic regime of the thermal conductivity curve, and we use the method of heat pulses.[2] In this method an alloyed metal film, serving as a heater, is deposited on one end of the solid to be studied and a superconducting film, serving as a bolometer, on the other. The typical lifetime of a phonon in a metal is 10^{-13} sec and since the typical width of our pulses is 10^{-7} sec, the heat pulse is "thermalized" in the heater, so that a heater temperature T_h can be defined. The vibrations so generated are transmitted to the solid under study. In the ballistic regime the mean free path of phonons is of the order of the size of the sample so that a temperature in the heat pulse cannot be defined. The heat pulse merely serves to launch a packet of phonons at one end of the solid which travel down the crystal; the

energy is stored in the vibrations of the atoms covered by the pulse. The atoms covered by the heat pulse vibrate incoherently with a power spectrum given by[3]

$$g(\omega, T_h, T_a) = (\hbar\omega^3/8\pi^2)[v_l^{-2}(\omega) + 2v_t^{-2}(\omega)]\{[\exp(\hbar\omega/kT_h)-1]^{-1} - [\exp(\hbar\omega/kT_a)-1]^{-1}\}. \quad (1)$$

In Eq. (1) T_a is the ambient temperature of the crystal. For $T_h \gg T_a$, $g(\omega, T_h, T_a)$ has its maximum at $\hbar\omega_{max} \approx 2.8kT_h$. In addition, the mean square displacement of the atoms is approximately given by

$$\langle y^2 \rangle = (9\hbar^2/mk_B\Theta_D)(T_h/\Theta_D)^2 \quad (2)$$

in the Debye approximation. For propagation along a symmetry direction, a part of the vibrations so set up travels approximately at the longitudinal sound velocity and the others at the transverse velocities. For $\hbar\omega \gg kT_a$, the lowest transverse branch has an exponentially decreasing decay rate ($\sim\exp[-\hbar\omega/kT_a]$) into other branches,[4] and this is the one that shows the nonlinear behavior.

The heat pulses were propagated along the (100) direction in pure NaF at an ambient temperature (1.4 to 4.2°K) which is within the boundary-scattering (ballistic) regime of the thermal conductivity curve. The single crystal was a piece of double-grown NaF grown by G. Schmidt of Cornell University and was of dimensions $1.9\times1.9\times1.1$ cm^3. The 50-Ω Constantan heater was varied in size between 5×5 and 9×9 mm^2. The indium bolometer[5] of similar size was biased by means of a magnetic field and currents up to 30 mA. In order to generate the high heater temperatures necessary for the experiment, the output of a Hewlett-Packard 214A pulse generator was amplified such that peak pulse amplitudes up to 500 V could be conveniently obtained.

Figures 1(a)-1(d) show some of the data obtained for different heater powers at an ambient temperature of 2°K. The pictures were taken under identical conditions except for the different amplifier gains employed to compensate for the larger signals at the higher powers. At low powers [Fig. 1(a)] we observe typical ballistic longitudinal and transverse pulses with a width somewhat larger than that of the input pulse.[6]

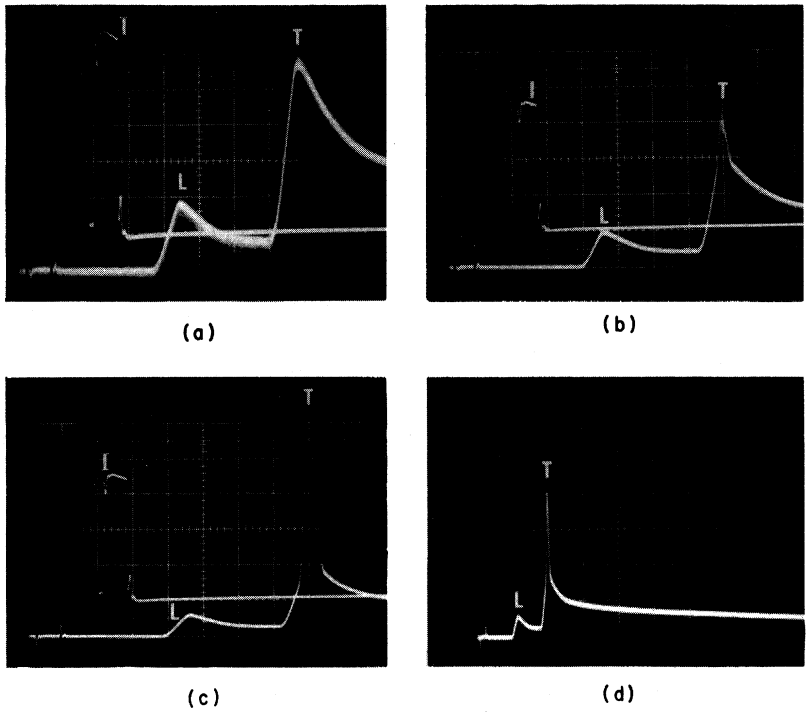

FIG. 1. Power dependence of heat pulses in NaF near threshold conditions. In some of the pictures the input pulse I is superimposed. Power densities (a) 50 W/cm^2, (b) 350 W/cm^2, (c) 1000 W/cm^2, and (d) 600 W/cm^2. Time scale 0.5 μsec/div in (a)-(c) and 2.0 μ sec/div in (d). The ambient temperature of the crystal was 2°K. The amplifier gain is not the same in the various cases. See text.

Above a threshold pulse power [Fig. 1(b)] a sharp spike is seen to develop on the usual transverse pulse. This spike grows dramatically in intensity and narrows in width with increasing pulse power [Fig. 1(c)]. Figure 1(d) was taken at an intermediate power level and on a four-times shorter time scale. It shows both the sharp spike and the background (diffusive) heating of the sample at these high powers. In all cases, the width of the transverse spike at half-height is narrower than the input pulse width. The peak amplitude of the transverse pulse in case (c) is 100 times that in case (a) for a power change of factor 20. The nonlinear growth of the transverse pulse is also demonstrated by comparing the relative amplitudes of the longitudinal pulse (which shows no unusual behavior) and the transverse pulse in cases (a)-(c).

It is clear that we are observing nonlinear propagation of the transverse heat pulse when the amplitude of vibration of atoms in the pulse is high enough. The phenomenon depends upon a competition between dispersion which tends to broaden the pulse and the nonlinearities which tend to narrow it. The theory of such effects is discussed in the following paper.[1] Here we compare quantitatively some of the predictions of the theory with the experimental results. The strain $y(\xi = x - v_g t)$, where v_g is the linear group velocity of the dominant phonons, is predicted to be of the form

$$y(\xi) = A\,\mathrm{sech}(\xi/\Delta), \tag{3}$$

where $\Delta \sim A^{-1}$. Such pulses are referred to as envelope solitons. In the experiment it is the energy density $E \sim y^2$ that is measured. We have found that the observed pulse shapes fit a $\mathrm{sech}^2\xi$ behavior to within the experimental error. Equation (3) predicts that the width of the pulse is proportional to the inverse square root of its height H. In Fig. 2, we plot Δ as a function of $H^{-1/2}$. The theory predicts a lower and an upper limit on T_h based on the consideration that the typical length in which the instability develops must be less than both the length of the crystal and the mean free path due to scattering from defects. The limits are given in terms of the nonlinearity coefficients and the dispersion. Unfortunately these parameters are hard to estimate for NaF. Experimentally the lower and

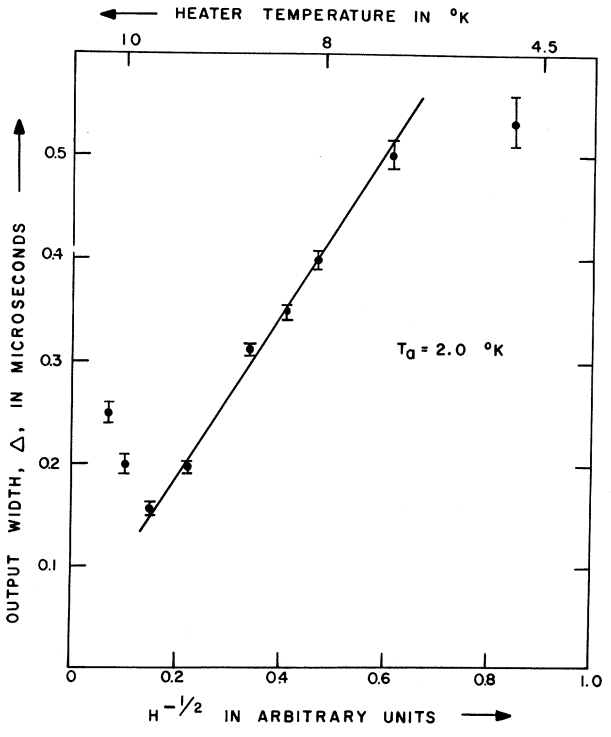

FIG. 2. Plot of the output width Δ versus $H^{-1/2}$ where H is the output-pulse height at fixed input-pulse width. Estimated heater temperatures are also indicated.

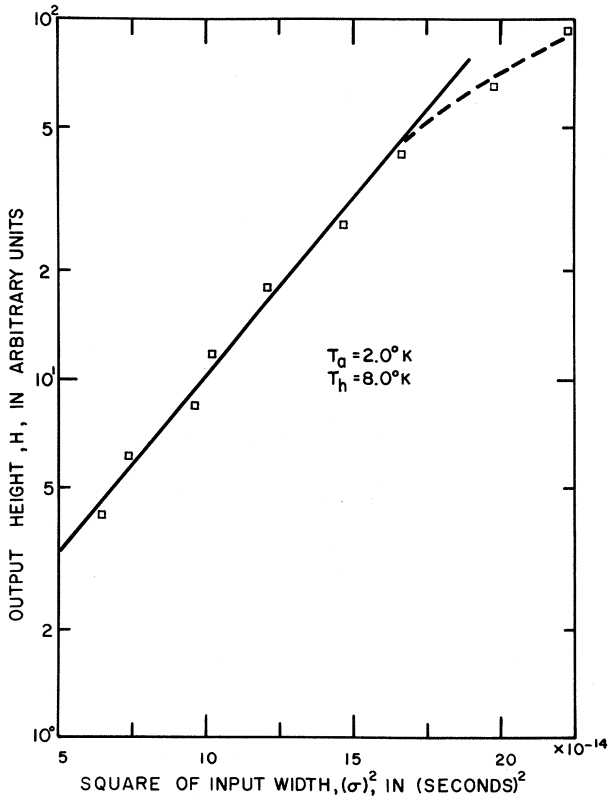

FIG. 3. Semilog plot of output height H versus square of input width σ^2 at a fixed input-pulse height.

upper bounds of T_h, for our sample of NaF, are ~5 and 10°K, respectively, for an ambient temperature of 2°K.

The theory predicts that the typical length (or time) for the development of the instability varies as σ^{-2}, where σ is the input width. In Fig. 3, we have plotted the output height H against σ^2 and find that for small σ, $H \sim \exp(\sigma^2)$ verifying that the time constant for the instability does indeed follow the predicted behavior.

In addition to the experiments described above, measurements were made in other heater-detector geometries. In one such experiment, a large heater (1.2×1.2 cm^2) was deposited on one end and a linear array of three detectors of size 3.5×3.5 mm^2 was deposited at the other end. It was found that each detector received approximately the same amount of energy at all power levels. Thus power-dependent spatial focusing effects are unimportant in our experiment. In another experiment heat pulses were propagated approximately 20° from the (100) direction. No pulse steepening was observed, confirming that propagation along a symmetry direction is necessary to observe the effects discussed here.

In summary, we have observed nonlinear propagation of heat pulses in solids in excellent agreement with the theory of Tappert and Varma.

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[1]F. D. Tappert and C. M. Varma, following Letter [Phys. Rev. Lett. 25, 1108 (1970)].

[2]For a recent review, see R. J. von Gutfeld, in *Physical Acoustics*, edited by W. P. Mason (Academic, New York, 1968), Vol. 5, p. 233.

[3]For simplicity we are neglecting acoustic-mismatch effects. Recent experiments show that for many solids this is approximately justified. See, for example, V. Narayanamurti, Phys. Lett. 30A, 521 (1969), and to be published.

[4]R. Orbach and L. A. Vredevoe, Physics (Long Is. City) 1, 91 (1964).

[5]By a suitable choice of biasing current and magnetic field our 1000-Å thick, slowly evaporated indium bolometers could be operated over a nearly linear transition region greater than 0.5°K in width without saturation at the high powers.

[6]T. F. McNelly, S. J. Rogers, D. J. Channin, R. J. Rollefson, W. M. Goubau, G. E. Schmidt, J. A. Krumhansl, and R. O. Pohl, Phys. Rev. Lett. 24, 100 (1970).

ASYMPTOTIC THEORY OF SELF-TRAPPING OF HEAT PULSES IN SOLIDS

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Using an asymptotic analysis leading to a modified Korteweg-de Vries equation and a nonlinear parabolic equation, it is shown that under certain conditions on phonon dispersion and lattice anharmonicity, self-trapping of heat pulses occurs in solids at low temperatures.

In an insulating crystal at an ambient temperature T_a in the ballistic regime of the thermal conductivity curve, a heat pulse generated at temperature $T_h > T_a$ may be regarded as a collection of large-amplitude waves propagating on a nonlinear lattice. The propagation of heat pulses is determined by the equations of motion,

$$m\ddot{u}_i^{\alpha} = \sum_{j\beta} a_{ij}^{\alpha\beta} u_j^{\beta} + \sum_{j\beta}\sum_{l\nu} a_{ijl}^{\alpha\beta\nu} u_j^{\beta} u_l^{\nu} + \sum_{j\beta}\sum_{l\nu}\sum_{m\delta} a_{ijlm}^{\alpha\beta\nu\delta} u_j^{\beta} u_l^{\nu} u_m^{\delta}, \quad (1)$$

where u_i^{α} is the displacement of the ith atom in the α direction, and the a's are derivatives of the interatomic potentials. Along a symmetry direction the propagation of different polarizations decouple ($a_{ij}^{\alpha\beta} \sim \delta_{\alpha\beta}$) in the harmonic approximation. Nonlinear terms with $\beta, \nu, \cdots = \alpha$ produce self-action effects, whereas nonlinear terms with $\beta, \nu, \cdots \neq \alpha$ produce coupling to other polarizations and therefore loss (or gain) to the pulse. Neglecting the short initial

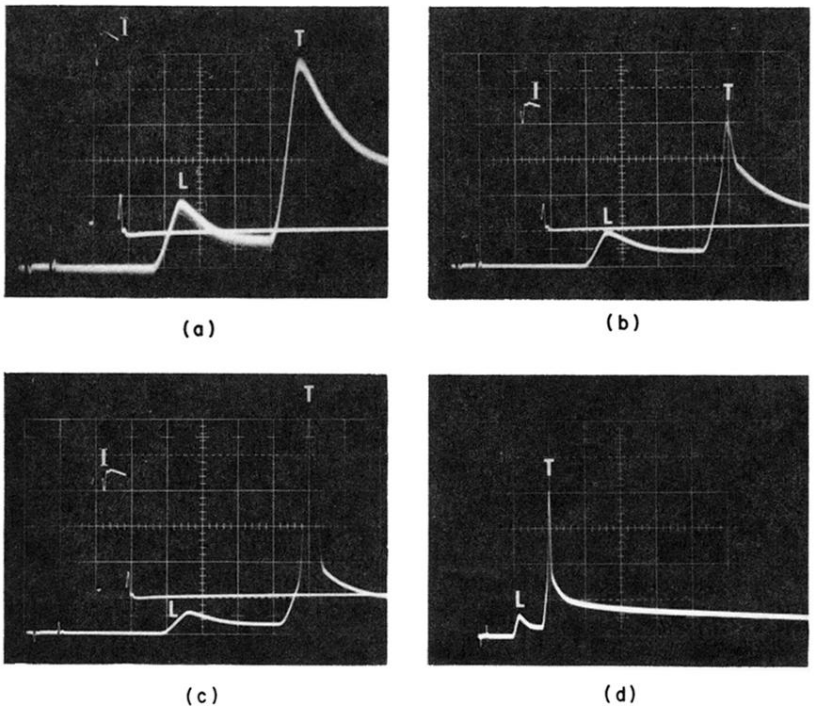

FIG. 1. Power dependence of heat pulses in NaF near threshold conditions. In some of the pictures the input pulse I is superimposed. Power densities (a) 50 W/cm^2, (b) 350 W/cm^2, (c) 1000 W/cm^2, and (d) 600 W/cm^2. Time scale 0.5 μsec/div in (a)-(c) and 2.0 μ sec/div in (d). The ambient temperature of the crystal was 2°K. The amplifier gain is not the same in the various cases. See text.