NONLINEAR PHONON GENERATION*

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Recently, the existence of high-energy acoustic phonons possessing anomalously long lifetimes $\tau_{\vec{k}}$ at low temperatures has been inferred by Geschwind et al.[1] A theoretical explanation of this property for transverse phonons at low temperatures was subsequently proposed by Orbach and Vredevoe.[2] The presence of these long-lived vibrations allows the possibility of "Suhl"-like processes[3] involving the $\vec{k}=0$ optical phonon of energy $\hbar\omega_0$ and its decay into two acoustic phonons with equal and opposite wave vectors $\vec{k}_1$ and equal energies $\hbar\omega_0/2$ (see Fig. 1). This decay can, in principle, be used as a parametric amplifier of high-energy lattice vibrations. In this Letter we investigate the use of this mechanism as a generator of high-energy acoustic phonons. Though we do not believe it possible to achieve breakdown in the usual sense, we shall be able to show that the occupation number of the driven acoustic phonons will be strongly enhanced over the $\vec{k}=0$ occupation number. This enhancement obtains because of the strong coupling to the $\vec{k}=0$ mode and the long acoustic-phonon lifetime.

To be more specific, we shall assume an anharmonic interaction Hamiltonian of the form[4]

$$\mathcal{H}_3=\sum_{\vec{k}}A(\vec{k})c_0c_{\vec{k}}^+c_{-\vec{k}}^+ + \text{c.c.}, \tag{1}$$

where $c_{\vec{0}}$ and $c_{\vec{k}}$ are the destruction operators for the $\vec{k}=0$ optical phonon and the acoustic phonon of wave vector $\vec{k}$, respectively. The rate equation for the occupation numbers of acoustic phonons of wave vectors $\vec{k}_1$ and $-\vec{k}_1$ with energy $\hbar\omega_0/2$ is simply[5]

$$\dot{n}_{\vec{k}_1}+\dot{n}_{-\vec{k}_1}=[4|A(\vec{k}_1)|^2/\hbar\Gamma_{\vec{k}_1}][(n_{\vec{k}_1}+n_{-\vec{k}_1}+1)n_0 - n_{\vec{k}_1}n_{-\vec{k}_1}]-(n_{\vec{k}_1}+n_{-\vec{k}_1})/(\hbar/\Gamma_{\vec{k}_1}), \tag{2}$$

where $\Gamma_{\vec{k}_1}=\Gamma_{-\vec{k}_1}=\hbar/\tau_{\vec{k}_1}$ represents the energy half-width of the acoustic phonons. It is clear that if the occupation number of the $\vec{k}=0$ phonon can be made sufficiently large (viz., $n_0 \gg n_{\vec{k}_1} \gg 1$), the right-hand side of (2) will become positive indicating an exponential growth of the populations of the $\pm\vec{k}_1$ phonon states. This is analogous to the so-called "first-order" Suhl instability in ferromagnetic resonance. The large energy of the $\vec{k}=0$ optical phonon precludes this possibility unless suitably intense optical sources at the infrared-absorption frequency can be found. In the absence of large values of n_0, there are other steady-state solutions of (2) which are of some interest. In particular, consider the situation when $n_{\vec{k}_1} \ll 1$, and $n_{\vec{k}_1}n_{-\vec{k}_1} \ll n_0$. The former condition is easily achieved at low temperatures ($T \ll \hbar\omega_0/2k_B$), the latter at reasonable optical phonon-excitation levels. Then the steady-state solution of (2) results in

$$n_{\vec{k}_1}=[4|A(\vec{k}_1)|^2/\Gamma_{\vec{k}_1}{}^2]n_0. \tag{3}$$

This relation implies a large enhancement of $n_{\vec{k}_1}$ over its thermal equilibrium value when the optical phonon at $\vec{k}=0$ is pumped sufficiently. Orbach and Vredevoe[2] find high-energy transverse phonon lifetimes at low temperatures due to anharmonic processes to be greater than those introduced by boundary scattering (where mode conversion to the rapidly decaying longitudinal phonons occurs). Thus, for a crystal 1 mm on a side, the transverse phonon lifetime $\tau_{\vec{k}_1} \sim 10^{-6}$ sec. The coefficient $A(\vec{k})$ is of the order of the Debye energy $\hbar\omega_0$ resulting in an enhancement factor [the coefficient of n_0 in (3)] of the order of $(\omega_D\tau_{\vec{k}_1})^2 \sim 10^{14}$. This enhancement, coupled with the availability of infrared gas lasers, should

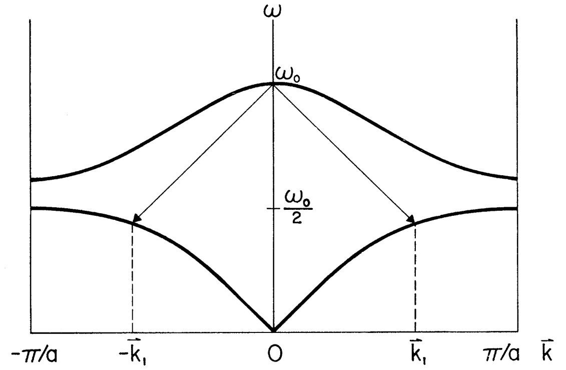

FIG. 1. A schematic for the decay of an optical $\vec{k}=0$ phonon with energy $\hbar\omega_0$ into two acoustic phonons with equal and opposite wave vectors $\vec{k}_1$ and with equal energies $\hbar\omega_0/2$.

make it possible to pump paramagnetic salts at low temperatures at the optically active phonon frequency, producing acoustic phonons at half the pump energy with anomalously large amplitudes. These phonons could then be detected in absorption as vibronic sidebands on the low-energy side of the no-phonon line.

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[5] See, for example, the article by H. Callen in Fluctuation, Relaxation, and Resonance in Magnetic Systems, edited by D. ter Haar (Oliver and Boyd, London, 1962), pp. 69-86.

GEOMETRICAL RESONANCE AND BOUNDARY EFFECTS IN TUNNELING FROM SUPERCONDUCTING In†

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A new type of structure in the tunneling characteristics of Al-AlO$_x$-Pb film diodes (Al and Pb both superconducting) has been reported recently for diodes employing very thick Pb films (2.9-9.7 μ).[1] The structure consisted of damped oscillations in d^2V/dI^2 vs V which were periodic in V and reflected weak periodic structure in dI/dV. Evidence was presented indicating that the structural period $h\nu$ depended only on Pb film thickness d(Pb). Specifically, $h\nu$ was found to be proportional to $1/d$ over the range investigated. When interpreted in terms of a standing wave phenomenon, the slope of $h\nu$ vs $1/d$ yielded a characteristic velocity comparable to the Fermi velocity.

Similar oscillations have now been observed with thick In films (5.8-32.2 μ), demonstrating that the effect is not restricted to strong-coupling superconductors and hence may occur rather generally. Furthermore, preliminary measurements indicate that the structural amplitude can be greatly enhanced (factors of five or more in d^2V/dI^2) by deposition of a rather thin "overlay" of silver (~0.2 μ) on the exposed In or Pb film surface, even though the silver may be tens of microns away from the AlO$_x$ barrier at which tunneling occurs. Preliminary indications are that $h\nu$ is insensitive to the presence of the silver overlay for the relatively thin silver films employed so far. Finally, measurements on very thick films (33 μ for In and 26 μ for Pb) suggest the absence of an energy gap in the excitation spectrum in the long-wavelength limit.

Diodes were prepared by standard methods[2] and exhibited room-temperature resistances in the range 2-100 Ω. Thin Al films (~300 Å) allowed the influence of longitudinal magnetic fields to be studied with both Al and In superconducting. Diode pairs (sisters) fabricated simultaneously, and differing only by one having a silver overlay, were employed in structural enhancement studies. All films were deposited on glass substrates nominally at room temperature. Preliminary x-ray studies have disclosed that the resulting polycrystalline In films exhibit a strong (101) texture, i.e., (101) crystallographic planes tend to be aligned parallel to the glass substrate. [Lead films behave similarly but with (111) planes involved.] Weighing techniques were utilized to determine In film thicknesses. Conventional modulation methods were employed to measure dV/dI and d^2V/dI^2.

Plots of dV/dI and d^2V/dI^2 for an In film 8.5 μ thick are presented in Fig. 1. These data are for a diode unit with a silver overlay. The structural amplitude (d^2V/dI^2) is approximately five times greater than for a sister unit without silver. To within experimental accuracy, $h\nu$ was the same for both units. The corresponding structure in dV/dI is much stronger than previously reported for Pb (without silver),[1] but is comparable to that observed with Al-AlO$_x$-Pb diodes having a silver overlay.

Matters now proceed much as for the case of Pb. Structural features in d^2V/dI^2 have been indexed by integers η with $\eta = 1$ corresponding to the first and strongest peak of the series.