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Resonant scattering of thermal phonons by dislocations in superconducting aluminum*

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Measurements of the lattice thermal conductivity in superconducting Al demonstrate the existence of a resonant phonon-dislocation interaction near 10^{10} Hz. Three different models of dislocations, each exhibiting an eigenfrequency, may be made to agree with the data. It is suggested that the dynamical behavior of dislocations at thermal amplitudes, as observed in the present measurements, may differ from the dynamical properties appropriate to ultrasonic and other techniques which employ macroscopic strain amplitudes.

I. INTRODUCTION

The string model of a dislocation has been rather useful in explaining the dynamic behavior of dislocations.[1] In this model the dislocation is viewed as an elastic band having mass and tension, each of which may be related quantitatively to the physical properties of the crystal. If the dislocation is held fixed at certain positions by a random array of pinning points along its length, the remaining free segments may vibrate at frequencies which are inversely proportional to the lengths of those segments. The expected frequencies are such that they may influence the thermal properties (specific heat, thermal transport) of a crystal at sufficiently low temperatures. Hence, these properties may also be studied in an attempt to test the model.

Indeed the presence of resonant dislocations has been observed in low-temperature thermal-conductivity measurements on LiF (Ref. 2) and superconducting Nb.[3] However, in neither case was it possible to make a definitive comparison with theory. Either the dislocation loop lengths or the phonon-dislocation scattering cross sections were not experimentally available. In fact, for Nb it was equally possible to explain the experimental results in terms of other models of dislocation motion. It is desirable, therefore, to seek other materials in which dislocations are more amenable to experimental investigation.

Recent measurements on thermal transport in superconducting Al suggested the presence of a resonant phonon-dislocation interaction.[4] The purpose of the present work has been to explore this possibility. To this end we have measured the lattice thermal conductivity of superconducting Al below 1 K using two different techniques. The first was a standard measurement of the thermal conductance of an annealed Al rod which had a section in which dislocations were deliberately introduced. The second technique involved measurements of the thermal conductance of thin Al foils under stress and with heat flow directed perpendicular to the plane of the foil.

Useful information concerning phonon scattering could only be obtained below ≈ 0.2 K with either technique, since at higher temperatures in the superconducting state of Al, the thermal conductivity is dominated by electrons. Hence, although we can state conclusively that a resonant phonon-dislocation interaction does occur in Al, some details of the resonance are not available. Data which support the conclusion that a resonant interaction does occur are presented in Sec. II, and a comparison with theory and with other relevant data is included in Sec. III.

II. EXPERIMENTAL RESULTS

Thermal-conductivity data for an Al rod of 0.6-cm diam were obtained using apparatus which has been described elsewhere.[3] The rod was annealed at 400 °C for 20 h in vacuum, was carefully mounted in the cryostat in an attempt to avoid strain, and then the lower half was bent to introduce dislocations. Data were obtained from both the "unstrained" and the intentionally strained portions. The electrical residual resistivity ratio after mounting was 600. The fractional difference in temperature across the sample was 5% or less during a measurement.

The data obtained from the Al rod are shown in Fig. 1. The solid curve is the electronic thermal conductivity calculated from the BCS theory of superconductivity,[5] assuming a critical temperature of T_c of 1.17 K and an energy gap of 3.52 kT_c.[6] This was subtracted from the data to give the lattice conductivity κ_g. The broken line represents the lattice conductivity if phonons were diffusively scattered by the surface of the rod only. This surface was in fact smooth, so specular reflection of phonons would occur and no correction was made for boundary scattering.[3] The phonon mean free path $l = \kappa_g/\alpha T^3$ with $\alpha = 0.27$ W/cm^{2}K^4. The results are presented in Fig. 2 for the "un-

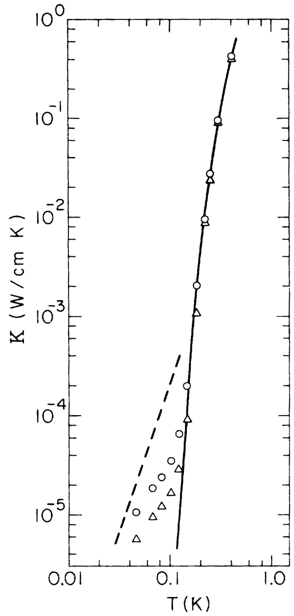

FIG. 1. Thermal conductivity of superconducting Al. ▽, sample intentionally strained; ○, sample not intentionally strained.

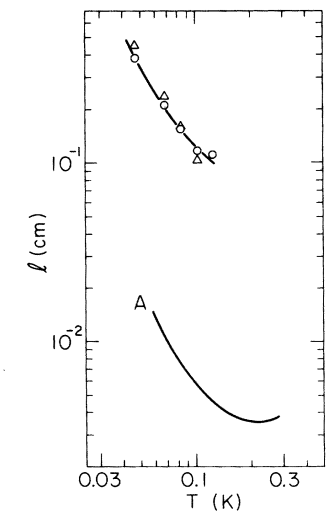

FIG. 2. Phonon mean free path l in superconducting Al. The symbols refer to the same samples as in Fig. 1; see text for details. Curve A is the phonon mean free path deduced from measurements on an Al foil.

strained" sample. Also shown is the mean free path associated with the *additional* phonon scattering caused by strain introduced into the bent section. That the magnitude of the latter is nearly identical to that of the background, which is assumed to be given by the scattering present in the "unstrained" sample, only indicates that we have coincidentally doubled the number of dislocations in the process of bending the rod.

For the measurements of the thermal conductance of Al foils, each foil was sandwiched between two Cu plates of about 1-cm^2 area. The Cu surfaces had been lapped to optical flatness. Mylar of 6×10^{-4}-cm thickness provided electrical isolation between the Cu and Al, and the sandwich was bonded together with epoxy. The thermal impedance R between the copper plates was measured in the presence of a constant heat flux with the Al in both the superconducting (R_{SC}) and normal (R_N) states. Since in the normal state the electronic thermal conductance of the Al foil is large, the difference ($R_{SC}-R_N$) removes the thermal resistance of the Mylar, epoxy, and interfaces and gives just the thermal resistance across the superconducting Al foil. Dislocations were introduced into the annealed Al foil at low temperature by differential thermal contraction. This reduced the chance of recovery of a strained sample or of diffusion of impurities to the dislocations which might take place at room temperature.

Thermal resistance data obtained from a sandwich containing a 0.024-cm thick Al foil is shown in Fig. 3. The foil, *in situ*, had an electrical residual resistivity ratio of 1000 measured perpendicular to the direction of heat flow. Although the thermal boundary resistance of the epoxy-Al interface may differ slightly between the superconducting and normal states,[4] only a small error is introduced if data above ≈0.06 K are used in obtaining the difference ($R_{SC}-R_N$). Also, as mentioned before, data above ≈0.3 K are influenced by electronic thermal conduction and thus are not used to obtain ($R_{SC}-R_N$).

The phonon mean free path obtained from the data of Fig. 3 is included in Fig. 2 as curve A. Data on a much thinner sample, 0.0028 cm thick, gave the same temperature dependence but a factor of 3 shorter mean free path. Much thicker

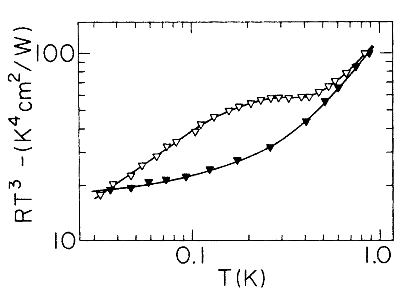

FIG. 3. Thermal impedance R of a Cu-dielectric-Al-dielectric-Cu sandwich, multiplied by the cube of the temperature to remove most of the temperature dependence. ▽, superconducting state of the Al; ▼, normal state.

samples (≈0.1 cm) also gave the same temperature dependence, but no phonon mean free path was computed since it is unlikely that the dislocations were even approximately uniformly distributed through the thickness of these Al samples. It is believed that the strain is greater near the epoxy-Al interface.

We thus found from four independent measurements on foils and two on rods that l in each case had the same temperature dependence, although admittedly the temperature range was rather narrow. We concluded that the same phonon scattering mechanism was present in each sample, and that only the density of scatterers was changed. Also the density of scattering centers was related to the strain. Assuming dislocations were responsible, the rod was sectioned by spark erosion, electropolished to remove surface damage, and dislocation etch-pit counts obtained. In the bent sample the total count was close to 2×10^6 cm^{-2}. The density in the "unstrained" half was therefore $\approx10^6$ cm^{-2}, and the process of bending introduced an additional $\approx10^6$ cm^{-2}. The latter figure may be compared with the estimated density of 0.8×10^6 cm^{-2} obtained by using the radius of curvature of the bend in the sample.[7] If the phonon scattering were proportional to the dislocation density, the foil (curve A of Fig. 2) would contain on the average $\approx2\times10^7$ cm^{-2}.

III. DISCUSSION

The phonon scattering depicted by Fig. 2 is definitely not caused by the static strain field surrounding the dislocations. A dislocation density of order 10^{11} cm^{-2} would be required to produce the measured phonon mean free path in the rod, and $\approx10^{12}$ cm^{-2} for the foil.[2,8,9] This is a factor of over 10^4 larger than the actual dislocation count in the rod, and for the foil, is a larger density than can be expected to exist even in a severely strained sample. Furthermore, the temperature dependence of Fig. 2 is not the T^{-1} expected for static-strain-field scattering.[8,9] One might attempt to match the measured temperature dependence by assuming a special dislocation array,[10,11] but this would necessitate an even greater density of dislocations. It is therefore not possible to explain the measured phonon mean free path in terms of static dislocations.

Theoretically, an eigenfrequency may be associated with dislocations in several ways.[2,3] It is likely that each model could be made to fit our data since the data are limited to a narrow temperature range within which the "resonant" minimum near 0.2 K in Fig. 2 is not fully and conclusively developed. In the following we will assume that a resonance does occur at 0.2 K or at a phonon frequency of $\approx2\times10^{10}$ Hz, and will compare the data with four models involving resonant dislocations.

The Granato-Lücke model,[12] mentioned in Sec. I, treats the dislocation like an elastic band stretched between pinning points a distance L apart, on the average. Hence, the dislocation exhibits a natural resonant frequency inversely proportional to L, and incident phonons of the same frequency may be absorbed and then reradiated in a different direction. On the basis of this model a dislocation density of $\approx4\times10^6$ cm^{-2} would be required to account for the thermal resistance of the rod,[1] a value which is in reasonable agreement with the measured value of $\approx10^6$ cm^{-2}. The foil would require $\approx10^8$ cm^{-2}, which is also a reasonable result. In addition, the observed temperature dependence of l is accounted for. However, the average loop length L would have to be $\approx10^{-6}$ cm, a value much shorter than usually deduced from acoustic measurements.[12,13] This might be explained by a set of "weak" pinning points which would not influence ultrasonic or dc stress-strain measurements, but would dominate the thermal-conductivity measurements. In the latter, the dislocations are in thermal equilibrium with the lattice at a very low temperature and thus vibrating with the smallest possible amplitude.

Alternatively, one could attempt to explain the high resonant frequency by an array or atmosphere of impurities which have diffused to the dislocation.[14] However, the dislocations in the foils were formed at low temperature, so diffusion of impurities was unlikely.

Another possibility is that an eigenfrequency is associated with the intrinsic structure of the dislocation. For example, the dislocation may oscillate within the undulating lattice or Peierls potential.[15] The resonant frequency ν_0 is then

$$\nu_0\approx(\tau_P/4\pi\rho b^2)^{1/2} ,$$

where ρ is the mass density, b is the magnitude of the Burgers vector, and τ_P is the Peierls stress. A resonant frequency of 2×10^{10} Hz leads to a Peierls stress of $\tau_P\approx10^7$ dyn/cm^2, a value which is small compared to typically quoted values of $\approx10^9$ dyn/cm^2.[15] The required dislocation density, however, would be in agreement with the experimental value.

Kronmüller has considered the resonant modes related to the relative motion of the parts of a dissociated dislocation.[16] The explicit case of Al has not been considered, probably because the stacking-fault energy in Al is believed to be large and not conducive to the formation of partials.[17] Nevertheless, for several other fcc metals, Kronmüller obtains $\nu_0\approx10^{10}-10^{11}$ Hz, which is in agreement with the present results on Al. In addition, we believe the temperature dependence and required dislocation density would be in qualitative agreement with the data.

There is some additional evidence in favor of the Kronmüller and Peierls potential models. The temperature dependence of l is essentially the same in six different measurements using Al of different purity from four different sources. It is unlikely that the array of pinning points would reproduce so well that L in the Granato-Lücke theory was always the same. With the Kronmüller or Peierls potential models, on the other hand, one would expect the resonant frequency to remain constant since the resonance is essentially independent of purity. It is quite possible, however, that both the string and the resonant structure models are valid. At large amplitudes with the whole dislocation in motion, as in ultrasonic or dc stress-strain measurements, the string model would be appropriate. At thermal amplitudes, as in the present measurements, the intrinsic structure of the dislocation may dominate.

In comparing the present results with other measurements, we note again that resonant dislocations have been studied in LiF[2] and in bcc Nb in the superconducting state.[3] In both cases, there was evidence that the eigenmode was influenced by the dislocation structure, although in LiF there was also clear evidence in support of the Granato-Lücke string model. It is possible that different dislocation models will be appropriate to different kinds of crystals.

Measurements on phonon-dislocation interaction in normal metals are still in a state of confusion. Often the experimental results are in good agreement with calculations of phonon scattering by the static strain field.[18] In other experiments, however, the phonon mean free path may more readily be explained by assuming a movable or resonant dislocation.[14,19] If, as suggested by the present data, eigenmodes of dislocations do exist in metals, why is there so little evidence of this in the numerous measurements on lattice conduction? A possible answer lies in the damping of the resonance by electrons.[20] Near 1 K a reduction in the amplitude of the resonant phonon scattering by a factor of $\approx 10^2$ would reduce the phonon scattering sufficiently that the static strain field mechanism would become dominant.[21] Since the static strain field scattering increases with frequency, a smaller factor would be sufficient at higher temperatures. In the Granato-Lücke theory, however, the influence of electronic damping on the dislocation resonance is related to the average loop length L.[22] The damping effect decreases for smaller L. A reduction of $\approx 10^2$ in the amplitude of the resonant phonon scattering would *not* be realized for the observed magnitude of electronic damping[20] if $L \approx 10^{-6}$ cm as required by the present data. In other models, such as that of Kronmüller, no estimate has yet been made as to the quantitative effect of electron damping on the equilibrium thermal behavior of a dislocation.

IV. SUMMARY

Experimental measurements of the lattice thermal conductivity in superconducting Al have demonstrated the presence of resonant scattering of phonons by dislocations. Both the temperature dependence and the phonon-scattering cross section agree with several theoretical models of a dislocation which display an eigenfrequency near 10^{10} Hz. All of these models have one or more adjustable parameters which have not as yet been determined experimentally. It is thus not possible from the present data to ascertain in general which model is more realistic. Indeed, different models may apply to different materials, and the appropriate model may depend on the magnitude of dislocation motion encountered.

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[1]A. V. Granato, Phys. Rev. B 4, 2196 (1971); and references cited therein.

[2]A. C. Anderson and M. E. Malinowski, Phys. Rev. B 5, 3199 (1972).

[3]A. C. Anderson and S. C. Smith, J. Phys. Chem. Solids 34, 111 (1973).

[4]R. E. Peterson and A. C. Anderson, J. Low Temp. Phys. 11, 639 (1973).

[5]J. Bardeen and J. R. Schrieffer, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1961), Vol. III, p. 170.

[6]G. L. Wells, J. E. Jackson, and E. N. Mitchell, Phys. Rev. B 1, 3636 (1970).

[7]A. H. Cottrell, *Dislocations and Plastic Flow in Crystals* (Oxford U. P., London, 1953), p. 29.

[8]P. G. Klemens, in *Solid State Physics*, edited by D. Turnbull and F. Seitz (Academic, New York, 1958), Vol. 7, p. 1; plus earlier papers cited therein.

[9]P. Carruthers, Rev. Mod. Phys. 33, 92 (1961).

[10]P. Gruner and H. Bross, Phys. Rev. 172, 583 (1968).

[11]M. W. Ackerman and P. G. Klemens, Phys. Rev. B 3, 2375 (1971).

[12]A. V. Granato and K. Lücke, in *Physical Acoustics*, edited by W. P. Mason (Academic, New York, 1966), Vol. 4, p. 225.

[13]$L \approx \frac{1}{4}(1/2\nu)(G/2\rho)^{1/2}$, where G is the shear modulus, ρ the mass density, and ν is the apparent frequency of resonance, 2×10^{10} Hz. The factor of $\approx \frac{1}{4}$ enters for an exponential distribution of loop lengths. J. A. Garber and A. V. Granato, in *Fundamental Aspects of Dislocation Theory*, edited by J. A. Simmons, R. de Witt, and R. Bullough, U.S. Natl. Bur. Stand. Spec. Publ. No. 317 (U. S. GPO, Washington, D. C., 1970), Vol. 1, p. 419.

[14]M. Kusunoki and H. Suzuki, J. Phys. Soc. Jap. 26, 932 (1969).

[15]A. Seeger, Philos. Mag. 1, 651 (1956).
[16]H. Kronmüller, Phys. Status Solidi B 52, 231 (1972).
[17]P. S. Dobson, P. J. Goodhew, and R. E. Smallman, Philos. Mag. 16, 9 (1967).
[18]M. W. Ackerman, Phys. Rev. B 5, 2751 (1972), and papers cited therein.
[19]A. D. W. Leaver and P. Charsley, J. Phys. F 1, 28 (1971).
[20]A. Hikata, R. A. Johnson, and C. Elbaum, Phys. Rev. B 2, 4856 (1970); 4, 674 (1971).
[21]See, for example, Fig. 14 of Ref. 2.
[22]A. V. Granato, in *Dislocation Dynamics*, edited by A. R. Rosenfeld, G. T. Hahn, A. L. Bement, and R. I. Jaffee (McGraw-Hill, New York, 1968), p. 117.