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Influence of Dislocation Motion on the Ultrasonic-Velocity Change in Superconducting Indium

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The change in velocity of compressional sound waves propagating in superconducting and normal indium has been measured at 13 MHz as a function of temperature and amplitude of the cw ultrasonic signal. It is found that the velocity of sound is strongly amplitude dependent, especially in the superconducting state. At the highest amplitude utilized, the velocity is observed to have changed by about one part in 10^3. The results can be explained on the basis of a direct interaction between the conduction electrons and oscillating dislocations as proposed by Granato and Lücke and by Kravchenko.

INTRODUCTION

Recent publications by several authors[1,2] have questioned the existence of a direct interaction between the electron gas in metals and oscillating dislocations, a mechanism which has been postulated to explain amplitude-dependent effects observed in ultrasonic attenuation measurements.[3–5] The purpose of this paper is to report that compressional-wave ultrasonic-velocity measurements in normal and superconducting indium strongly confirm the existence of an electron-dislocation interaction, and the experimental observations agree qualitatively with the theory of Granato and Lücke.[6] The results also are in accord with several recent experimental and theoretical investigations of the effect of the conducting electrons on dislocation motion (both vibrational and translational).[7–9] It appears that this technique might be quite useful for investigating dislocation effects in pure metals, especially the phenomena which occur when the electron gas is shorted out by the superconducting electrons.

The model of Granato and Lücke[6] assumes that a network of dislocations of length L_N in a crystal is pinned by impurity points L_C apart and by the intersection of the network dislocation loops. When an external stress wave is applied, the dislocations oscillate in a manner similar to the forced-damped vibration of a string[10] and two loss mechanisms are predicted to change the velocity and the logarithmic decrement (i.e., the attenuation) of the impressed wave. These are (a) a frequency-dependent amplitude-independent effect caused by the interaction of the forced dislocation motion with some damping motion (in this case, the conduction electrons) and (b) an amplitude-dependent loss due to the fact that on the basis of this model, the same stress-strain law is not followed on the loading and unloading cycles. Granato and Lücke derived the following equations for cases (a) and (b), respectively:

$$\left(\frac{\Delta v}{v}\right)_\nu = \frac{C_1}{2\pi}\,\frac{\omega_0^2-\omega^2}{(\omega_0^2-\omega^2)^2+\omega^2 d^2}\,, \tag{1a}$$

$$\Delta_\nu = \frac{C_1 \omega d}{(\omega_0^2 - \omega^2)^2 + \omega^2 d^2}, \qquad (1b)$$

$$\left(\frac{\Delta v}{v}\right)_A = \frac{C_2}{2}\left(\frac{C_3}{\epsilon} - 1\right) e^{-C_3/\epsilon}, \qquad (2a)$$

$$\Delta_A = C_2 (C_3/\epsilon - 1) e^{-C_3/\epsilon}, \qquad (2b)$$

where Δv is the difference between the measured velocity and the true elastic velocity v, Δ is the logarithmic decrement (which is proportional to the attenuation), and the subscript ν denotes the frequency-dependent loss while the subscript A indicates the amplitude-dependent loss. The constants C_1, C_2, and C_3 are related to properties of the material being studied, ω is the acoustic frequency of the external stress wave, ω_0 is the fundamental resonant frequency of the dislocation line, d is proportional to the damping constant B, and ϵ is the strain amplitude.

EXPERIMENTAL

The experiments were performed by using a continuous-wave (cw) resonance technique identical to that described earlier for measuring the electronic change in the velocity of sound at the superconducting transition in tin[11] and lead.[12] The frequency of a standing-wave resonance peak is monitored to keep the acoustic wavelength constant in the composite resonator made up of the sample, transducer, and bond. Under these conditions, the change in resonant frequency is proportional to the velocity shift caused by the variation of an external parameter such as temperature or magnetic field. Errors in measurement caused by stray electrical signals were found to be quite small in the present experiments, so that possible erroneous velocity changes due to electrical coupling could be neglected. The indium single crystals were grown by Semi-Elements, Inc., from 99.99%-pure starting material and were not intentionally deformed in any way. High-field magnetoacoustic studies[13] indicated that $ql \sim 0.6$ at 13 MHz for the samples, where q is the acoustic wave number and l the electron mean free path.

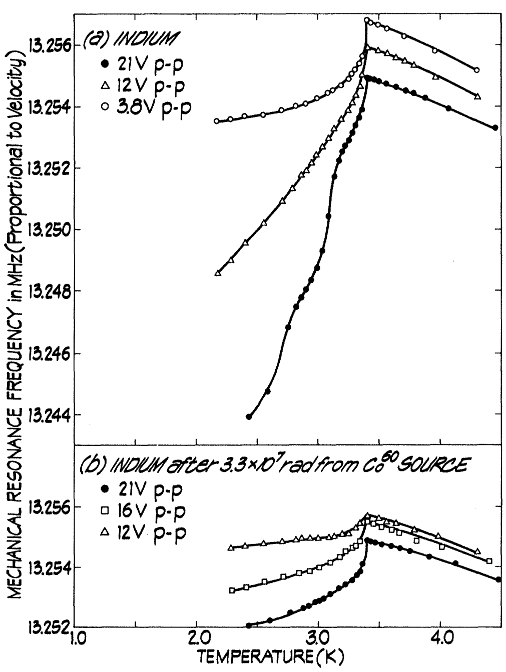

FIG. 1. Relative velocity as a function of temperature for compressional waves along [100] in indium at various sound-wave amplitudes. The amplitudes are expressed in terms of the peak-to-peak voltage of the cw signal. (a) Before irradiation; (b) after 3.3×10^7-rad irradiation from a Co^{60} source.

RESULTS

Velocity measurements for compressional sound waves propagating along [100] in indium as a function of temperature and sound amplitude are shown in Fig. 1(a) for frequencies near 13.25 MHz. It is observed that the change in mechanical resonance frequency, which is proportional to the sound velocity, is strongly dependent on the amplitude of the cw signal, especially below the superconducting transition temperature $T_c = 3.40$ K. Nonlinear effects occur at the highest amplitudes and the total velocity change is largest for the highest amplitude. As the amplitude of the acoustic signal is decreased, the total change in velocity below T_c decreases and tends to approach a limiting value $\Delta v/v \sim 2.3\times10^{-4}$. Data from the same run on the attenuation change are shown in Fig. 2(a), where the attenuation is obtained from the width of the mechanical resonance curves in a manner similar to that of Bolef and de Klerk.[14] The attenuation curves vary with temperature and pulse amplitude in a manner quite similar to previously published pulse measurements.[3,4] The largest total attenuation change occurs now for the lowest sound amplitude, while the least attenuation change occurs for the highest sound amplitude.

In order to establish that the observed amplitude changes were caused by dislocation motion, the indium samples were irradiated with a Co^{60} source (~1-MeV γ irradiation) and received a dose of 3.3×10^7 rad. The ultrasonic velocity and attenuation measurements were then repeated and the results are shown in Figs. 1(b) and 2(b). The irradiation, which produces numerous point defects which pin the dislocations and prevent their motion, greatly inhibits the amplitude effects observed previously. The amount of velocity change observed below T_c is now approximately independent of amplitude, as is the total attenuation change, in good agreement

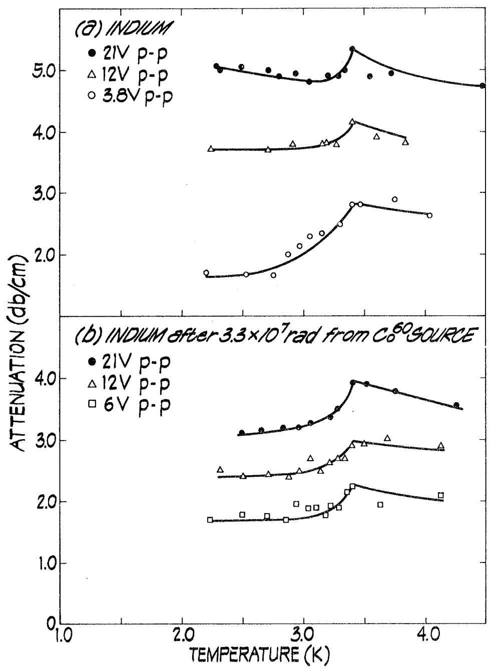

FIG. 2. Relative attenuation as a function of temperature for compressional waves in indium at various sound-wave amplitudes. The amplitudes are expressed in terms of the peak-to-peak voltage of the cw signal. (a) Before irradiation; (b) after 3.3×10^{7}-rad irradiation from a Co^{60} source.

with the neutron irradiation experiments of Thompson and Holmes.[15] Amplitude effects are not completely absent, however, as can be noted from the vertical displacement of the velocity and attenuation curves for various amplitudes.

DATA ANALYSIS

Amplitude-Independent Data

It should be possible to analyze the data in Figs. 1 and 2 on the basis of the Granato and Lücke model and obtain some of the basic parameters in Eqs. (1) and (2). The results should be consistent, for example, with the damping constant B which has been recently measured for aluminum[7] or can be calculated theoretically.[9] The damping constant B can be considered to consist of two parts[7]

$$B = B_e + B_{ph} ,$$

where B_e is the interaction of dislocations with the conduction electrons, and B_{ph} the interaction with phonons. Calculations of B_e have been made by Kravchenko[9] and Holstein[16] and are found to be independent of temperature. Measurements of B_e and B_{ph} in aluminum by Hikata, Johnson, and Elbaum[7] indicate that B_e is temperature independent and agrees well with theoretical calculations, whereas the phonon interaction term B_{ph} decreases with temperature down to about 50 K. The temperature dependence of B_e below the superconducting transition temperature, however, must reflect the vanishing number of "normal" electrons as the temperature approaches zero. The quantities varied in the present experiment were the amplitude and temperature, whereas the ultrasonic frequency was held constant near 13 MHz. The amount of electron damping changes with temperature, reflecting the changing mean free path and the superconducting transition, so that B_e should clearly reflect these changes and decrease sharply as the temperature is lowered in the superconducting state.

By combining Eqs. (1a) and (1b) in the case of no amplitude dependence, it is found that the damping constant B_e can be written as

$$B_e = \frac{2\omega_0^2\, a}{\omega}\left(1-\frac{\omega^2}{\omega_0^2}\right)\frac{\Delta_\nu}{(\Delta v/v)}\,, \qquad (3)$$

where a is the Burger's vector which is 3.25×10^{-8} cm in indium. Since the attenuation and velocity changes were measured experimentally as a function of temperature for various amplitudes, extrapolation to zero amplitude allows determination of Δ_ν and $\Delta v/v$. Unfortunately, these values include the electron-phonon contributions to the attenuation and velocity changes, so that quantitative information on the temperature variation of B_e cannot be obtained from the present measurements. Assuming that the electronic changes in Δ_ν and $\Delta v/v$ follow BCS behavior and are about the same size as the dislocation contributions, a plot of B_e as a function of temperature can be made. It is found that B_e decreases sharply below the transition temperature as is expected. Theoretical values of B_e just above T_c are found to be $\sim 0.4\times10^{-5}$ for edge dislocations and $\sim 1.1\times10^{-5}$ dyn sec/cm for screw dislocations on the basis of a free-electron model assuming three electrons per atom.[9,15] The values of B_e which can be determined from the experimental parameters through Eq. (3) are found to be consistent with the theoretical values given above if $\omega_0 \sim 10^{8}$ sec^{-1} and the average dislocation loop length is $\sim 2\times10^{-3}$ cm.

Amplitude-Dependent Data

The amplitude dependence of the ultrasonic velocity is quite similar to that of the attenuation which has been discussed at length by other investigators.[3,4] In Fig. 3 is shown the amplitude dependence of $(\Delta v/v)_A$ for a number of temperatures. The points shown are measured relative to the velocity which was determined by extrapolation to zero amplitude. The amplitude dependence is most pronounced below T_c, although there is a definite amplitude effect in the normal state. After

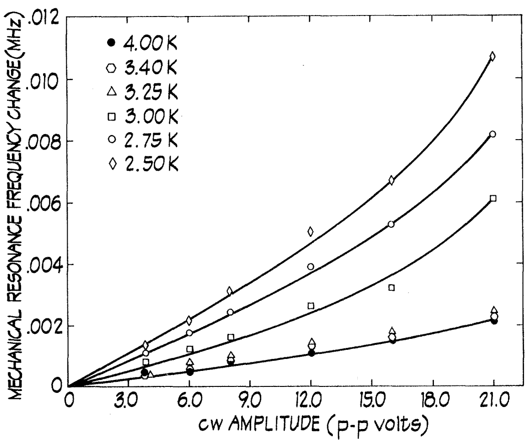

FIG. 3. Amplitude dependence of the relative velocity (in terms of mechanical resonance frequency) at various temperatures around $T_c = 3.40$ K. The points shown were measured relative to the velocity determined by extrapolation to zero amplitude. The amplitudes are expressed in terms of the peak-to-peak voltage of the cw signal.

the sample had been irradiated, the observed amplitude dependence at all temperatures was approximately the same as that observed above T_c before irradiation so that the amplitude effect is greatly inhibited by the pinning action of the radiation on the dislocations. Even after the irradiation, there remains a definite amplitude-dependent effect in which the velocity of sound decreases as the sound amplitude is increased. This effect occurs in the normal and superconducting states and is probably due to the remnant effects of dislocations insufficiently pinned by the radiation.

The dependence of $(\Delta v/v)_A$ on the strain amplitude ϵ is generally discussed in terms of a Granato-Lücke plot[6] which is a plot of the product of the $(\text{amplitude})^{1/2}$ and $(\Delta v/v)_A$ vs $1/\epsilon$. A plot of this type is shown in Fig. 4, where the strain amplitude is plotted in terms of the peak-to-peak voltage of the cw signal propagating through the composite oscillator, since Read[17] has shown that the strain amplitude is proportional to the voltage applied to the quartz transducer. It is observed that the experimental data lie quite accurately on straight lines, as the theory would predict. The slopes of the plots for different temperatures appear to be about the same, but the y intercepts increase as the temperature is lowered. Since the y intercept is determined by the constant C_2, it follows that C_2 increases as the temperature is lowered below T_c. This constant is related to λ, the total length of movable dislocation line, the network length L_N, and the dislocation pinning length L_c by the relation $C_2 \sim \lambda L_N^3/L_c^2$ as well as some other parameters which should remain fixed with temperature. It thus appears that $\lambda L_N^3/L_c^2$ increases in the superconducting state when the electron-damping mechanism is gradually removed (see Note added in proof), although physically, it is difficult to visualize any of these parameters changing appreciably with electron damping.

CONCLUSION

The cw measurements of the change in sound velocity and attenuation in normal and superconducting indium have indicated a pronounced effect on the amplitude of the impressed sound wave. Amplitude effects of this type have been reported for ultrasonic attenuation measurements in superconductors, but the present investigation is the first showing how the sound amplitude affects the velocity above and below T_c. The velocity of sound is found to be shifted to lower values for increased sound amplitudes, the effect being the strongest at temperatures below T_c.

The behavior of the sound velocity can be explained in terms of an interaction between the conduction electrons and dislocations which are vibrating because of the impressed ultrasonic wave. Since a dislocation has associated with it an electrostatic field, at low temperatures in the normal state, the electron cloud tends to damp the motion of the dislocations. In the wave equation of the sound wave, this force of interaction must appear and thus contribute to the stiffness of the lattice and therefore to the sound velocity. In a high-purity metal, where there is an appreciable electron-phonon interaction in addition to the electron-dislocation term, the sum of these two contribu-

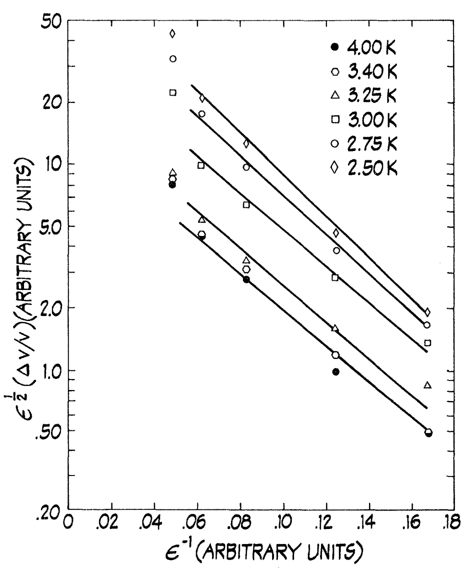

FIG. 4. Granato-Lücke plot of the velocity change for several temperatures in the normal and superconducting states for indium.

tions is of importance in determining the velocity of sound. At the superconducting transition temperature T_c, the electron-phonon and electron-dislocation forces begin to weaken and approach zero as the temperature approaches zero, since the superconducting pairs cannot participate in interactions with the phonons or the dislocations. As the lattice becomes less stiff, the velocity should decrease below T_c as is observed, the amount of fall depending on the initial value of the two interaction forces mentioned above. The total electron-phonon velocity change in the superconducting state is constant and is about 2 parts in 10^4, whereas the change due to the electron-dislocation interaction is amplitude dependent and as large as 1 part in 10^3 for the highest amplitude employed. Nonlinear effects arise at very high amplitudes in the superconducting state because of the possibility of detachment of the dislocations from pinning points.

All the above data can be fit qualitatively to the vibrating-string dislocation model proposed by Granato and Lücke and by Kravchenko. As far as temperature and amplitude dependence of the velocity and the decrement, the present data are in good accord with previous investigations in temperature regions where comparison is possible. Analysis of the amplitude-independent and amplitude-dependent components of the velocity and attenuation provides information about several dislocation parameters of the material being investigated. Although dislocation parameters are in general quite difficult to measure with a large degree of exactness, combination of the ultrasonic velocity and attenuation data, together with simultaneous metallurgical techniques might allow considerable detail to be obtained about dislocations in high-purity single crystals. The ultrasonic velocity measurement, in particular, is extremely sensitive to the presence of dislocations, much more so than the attenuation, and thus serves as an excellent indication of even a small dislocation content.

Note added in proof. This observation might possibly be explained on the basis of the results of A. Hikata, R. A. Johnson, and C. Elbaum, Phys. Rev. B 2, 4856 (1970), who found that L_c decreases as the temperature decreases.

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