CHANGE IN VELOCITY OF SOUND BETWEEN NORMAL AND SUPERCONDUCTING TIN FOR $ql > 1$

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Measurements of the change in the velocity of sound of shear and compressional waves at the normal-to-superconducting transition have been made in a high-purity tin single crystal. The changes observed are of the order of a few parts in 10^4, are electronic in origin, and agree qualitatively with theory for the shear-wave case.

Dating from the early experiments of Bömmel and Olsen,[1,2] several attempts have been made to determine possible gross changes in the velocity of sound between the normal and superconducting states in high-purity metals. Although the thermodynamical change in the static elastic constants at the superconducting transition is now well established,[2] large changes such as those predicted by Ozaki and Mikoshiba[3] and Stevens and Houghton[4] due to interaction of the sound waves with the conduction electrons have not as yet been observed. The purpose of this Letter is to present preliminary results of measurements of the change in the velocity of shear and compressional waves between normal and superconducting tin in the region $ql > 1$, where l is the electron mean free path and $q = 2\pi/\lambda$, where λ is the acoustic wavelength. Our results with shear waves are in rough qualitative agreement with the theory of Ozaki and Mikoshiba,[3] and the relative velocity is found to decrease by a few parts in 10^4 during the transition from the normal to the superconducting state for 13-MHz sound.

The experimental difficulties which have prevented these velocity-change measurements until the present time can mainly be attributed to the large decrease in ultrasonic attenuation which occurs at the transition from the normal to the superconducting state for $ql > 1$. A technique is required, therefore, that will allow the determination of small velocity changes while the attenuation is rapidly varying. Our experiments were carried out utilizing a technique and data interpretation developed by Yee and Gavenda,[5] which allows such measurements. A similar apparatus was developed independently by Melcher, Bolef, and Merry.[6] A continuous-wave resonance technique using frequency modulation and phase-sensitive detection is employed to give an accuracy of 2 parts in 10^6. In the experiments the frequency of one of the standing-wave mechanical resonance peaks is adjusted to keep the acoustic wavelength constant in the composite resonator made up of the sample, transducer, and bond. Thus, the change in resonance frequency is proportional to the velocity change caused by some external perturbing parameter such as temperature or magnetic field. (Note that the variation in length of the sample due to thermal expansion for the temperature range being studied is negligible.) One of the major experimental difficulties involves the occurrence of electrical coupling between the two ultrasonic transducers which can give rise to a false velocity change. This possible error introduced in the data can be overcome by averaging the data for two adjacent standing-wave resonance peaks.[4] In our experiments, data were considered acceptable only when the effects of electrical coupling were very small, i.e., when measurements on adjacent resonance peaks gave essentially identical results.

The sample holder was similar to that used by Deaton[7] and allowed continuous variation of temperature from about 1 to 20°K. In addition, runs were made with the sample immersed directly in the liquid-helium bath so that temperature accuracy and control of ±0.001°K could be achieved. The magnetic field of the earth was cancelled during temperature runs. The sample was placed between the poles of an electromagnet so that a transverse field could be applied to drive it normal at temperatures below the critical temperature of 3.72°K. The tin single crystal used in the measurements was grown from 99.9999% pure starting material and was cut so that sound could be propagated along either the [001] or [110] directions. High-field magnetoacoustic studies[8] indicate a value $2 \lesssim q \lesssim 4$ for an ultrasonic frequency of 13 MHz for this crystal.

The velocity measurements for sound propagation along [001] as a function of temperature are shown in Fig. 1 for frequencies near 13.4 MHz. In Fig. 1(a) is shown the variation of mechanical resonance frequency (which is proportional to the velocity) with temperature for shear waves with polarization along [110], while Fig. 1(b) displays results for compressional waves. These results are averages of data obtained from two adjacent mechanical resonance peaks, the individual shear

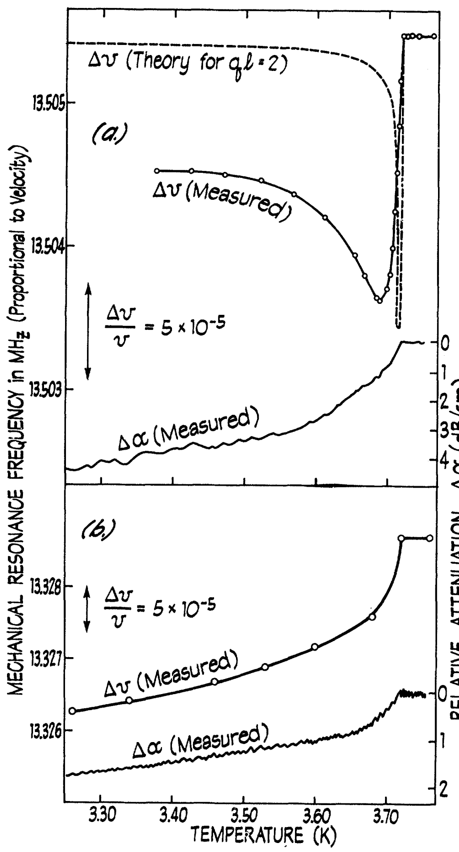

FIG. 1. Relative velocity and attenuation measurements as a function of temperature for 13-MHz sound along [001] in tin. (a) Data for shear waves with polarization along [110], also showing the theoretical curve of Ozaki and Mikoshiba for $ql=2$. (b) Compressional-wave data. Note that $\Delta v/v=(v_n-v_s)/v_n$.

runs deviating from the average by less than 5% and the compressional by less than 15%. The data points were taken after the establishment of equilibrium which required no more than a few minutes. Identical results were obtained for increasing and decreasing temperature runs. Also shown in Fig. 1(a) is a theoretical curve for $ql=2$ using the results of Ozaki and Mikoshiba.[3] It is seen that there is rough qualitative agreement between theory and experiment, at least as far as the relative shape is concerned.

The theory used by Ozaki and Mikoshiba[3] is based on the two-fluid free-electron model used by Claiborne and Morse.[9] A more recent calculation by Stevens and Houghton[4] is based on the BCS model but gives results that differ very little from the two-fluid model. The most striking feature of the theory is the extremely sharp decrease in the velocity of shear waves at the transition temperature T_c. The velocity in the superconducting state v_s reaches a minimum less than 5 mdeg below T_c, and then increases as the temperature is lowered, never reaching the value in the normal state v_n. The experimental curve shows a minimum at about 20 mdeg below T_c, and v_s does not increase as much at lower temperatures as the theory predicts. Careful investigations of the rapid-fall region near T_c seem to indicate that the disagreement is not an experimental artifact, unless an ~15-mdeg thermal gradient could be established with the sample immersed in a helium bath. It should also be noted in Fig. 1(a) that the attenuation of shear waves for 13 MHz falls very sharply at T_c in the first 20 mdeg, then decreases more gradually as the temperature is lowered. This experiment is the first instance, therefore, in which it has been observed that the velocity shift caused by the interaction of the sound wave and the Fermi surface electrons did not follow the attenuation which arises from a similar interaction. In Fig. 1(b) it is seen that the velocity and attenuation changes in the superconducting state are quite similar for compressional waves. The total velocity change for compressional waves is somewhat larger than that observed for shear waves, being $\sim 2\times 10^{-4}$, while the initial drop for shear waves is $\sim 1.4\times 10^{-4}$ at 13.4 MHz. Since there is as yet no theory for the compressional case we are unable to analyze the relative magnitudes in a quantitative manner. It should be noted, however, that the relative compressional velocity below T_c shown in Fig. 1(b) seems to follow a BCS equation[10] of the type $v_s/v_n=2f(\Delta(T))$.

Data on the magnetic field variation of the velocity of compressional waves in tin are shown in Fig. 2 as a function of temperature. The frequency is near 13.325 MHz, with $\vec{q}\parallel[001]$. The velocity is seen to exhibit superconducting-to-normal transitions quite similar to those observed in attenuation studies. The isothermal curves in a magnetic field for shear waves were similar to those shown in Fig. 2.

Experiments have also been conducted at 39 and 65 MHz for both shear and compressional waves. The shape and the magnitude of the resulting curves seem to scale roughly in accordance with theory. The electrical coupling problem mentioned earlier is much more pronounced at higher frequencies, however, and we have so far been unable to obtain the accuracy indicated for the 13 MHz data.

On the basis of our measurements we can draw the following conclusions: (1) The change in ul-

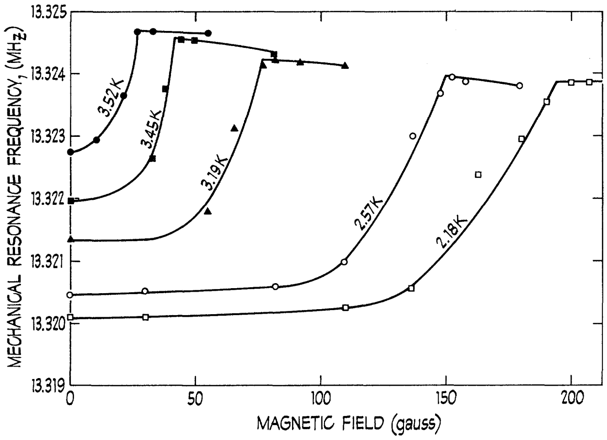

FIG. 2. Magnetic field variation of the mechanical resonance frequency (which is proportional to the sound velocity) of 13.3-MHz compressional waves propagating along [001] in tin for several temperatures below T_c.

trasonic velocity at the normal-to-superconducting transition in tin is easily measurable and is of the same order of magnitude as predicted in the case of shear waves. (2) Our data on shear waves agrees only qualitatively with the theories of Ozaki and Mikoshiba[3] and Stevens and Houghton[4]. The discrepancies in experiment and theory involve the temperature interval of the rapid fall at the transition, and the amount v_s retraces toward v_n. (3) The study of this effect should be as useful as and certainly will complement investigations of superconductivity using ultrasonic attenuation methods. (4) Although preliminary theoretical treatments exist only for the shear case, it is quite possible that information not obtainable from attenuation measurements might be acquired from velocity studies. These studies are currently being extended to indium and lead, the latter to investigate possible variation in behavior which could be caused by the strong coupling nature of lead.

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DYNAMICS OF THE ELECTRIC-FIELD-INDUCED CONDUCTIVITY TRANSITION IN MAGNETITE

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We have studied the dynamics of the voltage-induced conductivity transition in magnetite at 78°K. We observe a time delay between the application of the voltage and the observation of the transition in conductivity, inversely proportional to the power applied to the sample. The mechanism of the transition is interpreted in terms of a Joule self-heating of the sample to the 119°K Verwey transition.

It has been suggested recently[1] that typical Mott insulators may undergo a field-induced transition from the insulator state to the metallic state. A very important feature of the theory is the existence of a critical temperature T_c above which the materials in question are metallic even at zero fields. As an example where the theory of such a transition applies, the behavior of magnetite under high fields has been investigated by Lipsicas and Mattis.[2] They show that Fe_3O_4 at 78°K undergoes a transition from an insulating state to a conducting state when large enough electric fields are applied. The critical temperature T_c for Fe_3O_4 is 119°K.[3]

Magnetite and VO_2 have similar temperature-dependent transitions of their conductivities. It has been demonstrated in the vanadium dioxide case[4] that a fast transition in the conductivity can