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Magnetic Field Dependence of the Velocity of Sound in Ultrapure Cd and Cu†

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The change in the velocity of sound in very pure Cd and Cu as a function of the intensity of a perpendicular magnetic field has been measured for $ql \gg 1$. The velocity oscillates in phase with the oscillations in the attenuation. The amplitude of the relative changes in the velocity is about 10^{-4}. A standing-wave resonance technique which eliminates the effects of stray electrical signals is described. It takes advantage of the fact that transmitted signals for even and odd numbers of half-wavelengths within the sample differ in phase by 180°. The technique is readily adaptable to automatic recording systems.

1. INTRODUCTION

In 1954, Alpher and Rubin[1] showed that the velocity of sound in metals ought to have a quadratic dependence on the magnetic field intensity under the condition that $l \ll \lambda$, where l is the electron mean free path and λ is the wavelength of sound. This effect has been verified experimentally by Galkin and Koroliuk[2] in polycrystalline Sn and Al, by Beattie, Silsbee, and Uehling[3] in polycrystalline Al and Mg, and by Alers and Fleury[4] in single-crystal Au, Ag, Cu, Al, Ta, and V.

In 1963, using a free-electron model, Rodriguez[5] predicted the oscillation of the velocity of sound in metals as a function of magnetic field intensity for field values below the quantum region. These oscillations are related to the oscillations in the attenuation of ultrasound caused by geometric resonance. As in the case of the attenuation, the two conditions that are required for the observation of the oscillations in the velocity are $ql > 1$ and $\omega_c \tau > 1$, where $q = 2\pi/\lambda$, ω_c is the electron cyclotron frequency, and τ is the electron relaxation time. From the results obtained by Rodriguez, one can show that the relative velocity change $\Delta V/V$ is related to the attenuation α by

$$\Delta V/V = (\omega\tau/2)(\alpha/q) \tag{1}$$

for compressional waves with the applied magnetic field $\mathbf{B}_0 \perp \mathbf{q}$ in the limit $ql \gg 1$, $\omega\tau < 1$. Thus one expects oscillations in the velocity of sound analogous to the magnetoacoustic oscillations in the attenuation.

The observation of these oscillations in the velocity was reported by Beattie and Uehling[6] in 1966. Their experiment employed a 100-MHz longitudinal wave propagated in single-crystal Al at 4.2°K. However, because of experimental difficulties, the periods of the oscillations were well defined but not the amplitude nor the phase. They found occasional unexplained reversals in the sign of the velocity shift. That is, in one experiment they found the velocity to be a maximum for a given magnetic field value, while in another experiment they found the velocity to be a minimum for the same value of magnetic field, and the amplitude of the velocity shift differed by as much as several hundred percent from one experiment to another. However, the periods of the oscillations did indicate values of electron momentum which agreed with those obtained from the periods of the oscillations in the attenuation.

The purpose of this paper is to describe an experimental technique and a scheme for interpreting the results which greatly improve the repeatability of measurements of changes in the velocity of sound. The technique has been applied to very pure single crystals of Cu and Cd with results which are reproducible within 17% in the worst case.

2. EXPERIMENTAL METHODS

The choice of experimental technique to measure the velocity shifts as a function of the magnetic field intensity is restricted by the fact that the attenuation of compressional waves generally increases as l increases. The resistivity ratio $(\rho_{300°K}/\rho_{4.2°K})$ of the Cu sample[7] used here was found to be about 35 000 by the eddy-current decay method. The Cd sample came from the same stock as used by Gavenda and Chang,[8] and its resistivity ratio was found to be in excess of 30 000 by a standard four-probe resistance-measuring technique. Both the Cd and Cu samples are estimated to have $ql \cong 100$ at a frequency of 33 MHz. Consequently, at this and higher frequencies, the sound wave is heavily damped at large values of B_0.

The technique chosen for these experimental conditions was a continuous-wave resonance technique. The primary limitation on the accuracy is the stability of the continuous-wave oscillator used [about 6 parts per million (ppm) at 33 MHz, and 3 ppm at 55 MHz]. The principle behind this technique is that the sound wave

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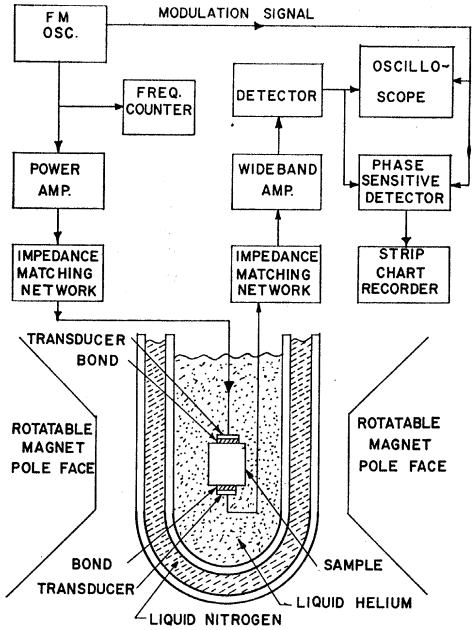

FIG. 1. Schematic representation of apparatus for velocity of sound measurements.

forms standing-wave patterns in the composite system of transducers, bonding material, and sample. (Relatively unimportant effects of phase shifts at boundaries will be ignored here.) When the frequency is such that there is an integral number of half-wavelengths of sound in the composite system, the system is in mechanical resonance. For a given total length of the system L and a resonance frequency f_0 at zero magnetic field, the condition for resonance is

$$L=n(\lambda_0/2)\,, \tag{2}$$

where $\lambda_0=V_0/f_0$, V_0 is the velocity of sound at zero field, f_0 is the resonance frequency, and n is an integer. If an external magnetic field is applied to the sample so that the velocity is changed, the system is no longer in resonance because the wavelength is now different. However, the system can be brought back into resonance by changing the frequency to some value f_1 such that the wavelength becomes equal to λ_0 again. That is, f_1 is adjusted so that

$$V_0+\Delta V=\lambda_0 f_1$$

or

$$V_0+\Delta V=\lambda_0(f_0+\Delta f)\,,$$

where $f_1=\Delta f+f_0$. Since $\lambda_0 f_0=V_0$, this expression reduces to

$$\Delta V=\lambda_0\Delta f. \tag{3}$$

Thus the velocity change can be measured by measuring the change in frequency needed to bring the system back into resonance.

Figure 1 shows the block diagram of the experimental setup used in the measurement of the velocity changes. Before discussing the details of the system it will be helpful to look at Fig. 2, which shows the output of the detector plotted as a function of frequency for a continuous-wave signal. A series of resonance peaks are superimposed on the response curve determined by the finite bandwidth (approximately 1.5 MHz) of the transducers and associated tuned circuits. The frequency separation δf between the peaks for $L\gg\lambda$ is found from (2) to be

$$\delta f=V/2L. \tag{4}$$

If the oscillator is given a small amount of frequency modulation, the detector output will be proportional to the amplitude modulation caused by the slope of the response curve. That is, the output of the detector will have an ac component at the modulation frequency with amplitude and phase related to the slope of the response function. At a resonance peak the slope is zero, so the output is zero. The output is measured with a phase-

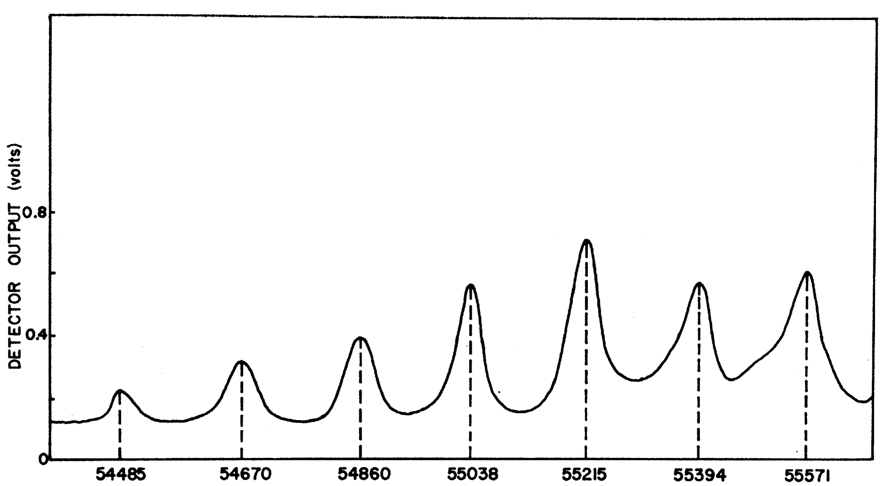

FIG. 2. Detector output voltage versus frequency for compressional waves in Cd at 77°K. Voltage maxima occur near standing-wave resonances.

sensitive detector to enhance the signal-to-noise ratio for the system.

The data presented below were obtained in the following fashion: With zero applied field, the center frequency of the FM oscillator was adjusted until the output of the phase-sensitive detector read zero, then the reading of the frequency counter was recorded. A small magnetic field was then applied, the oscillator readjusted for a null, and the new frequency recorded. This procedure was repeated for increasing values of the field, using steps sufficiently small that there was no danger of getting over onto another resonance peak.

The construction of the sample holder is found to be critical in the velocity measurements. In order to use a continuous-wave signal, the receiving end of the holder must be electrically isolated from the transmitting end. If it is not, the electrically coupled signal E_c, when mixed with the acoustic signal, i.e., the electrical signal E_a that is converted from the sound waves by the quartz transducers, produces an apparent velocity change, for reasons to be explained in Sec. 3.

Figure 3 shows the details of the sample holder that was used in the measurements. The upper and lower housings, as well as the collar, which was machined to fit the sample, and the spacer are made of brass. The position of the upper housing is held fixed by the thin-walled stainless steel tubes which serve as grounds for the two shielded signal leads. The inner conductor of the central lead is also made of thin-walled stainless steel tubing, while the inner conductor of the side lead is a #24 copper wire which is insulated from ground by means of rubber spaghetti. A cylindrical brass sleeve (not shown) encloses the entire assembly.

The edges of the coaxially plated transducers are soldered to a grounded cylindrical probe with indium solder in order to reduce the direct-coupled electrical signal. The electrical coupling was further reduced by electrically grounding the sample to the collar with silver paint.

3. EXPERIMENTAL RESULTS AND INTERPRETATION

A. Effect of Electrically Coupled Signals

The Cd data were obtained with compressional waves propagated in the [1$\bar{2}$10] direction, which was perpendicular to the direction of the magnetic field $\mathbf{B}_0$. The length of the Cd sample along the [1$\bar{2}$10] direction is 1.003 cm. The Cu data were obtained with compressional waves propagated in the [110] direction, which was also perpendicular to $\mathbf{B}_0$. The Cu sample length along the [110] direction is 1.29 cm.

A modulation frequency of 88 Hz and deviations of 4 to 6 kHz were used in determining the frequencies at which resonance peaks occurred. An example of the results obtained when a relatively large electrically coupled signal was present is shown in Fig. 4. The curve obtained for one resonance peak, indicated by +, follows the attenuation curve, while the curve obtained from an adjacent resonance shows a drastically different behavior with peaks and dips interchanged in some cases. This is reminiscent of the results of Beattie and Uehling.[6]

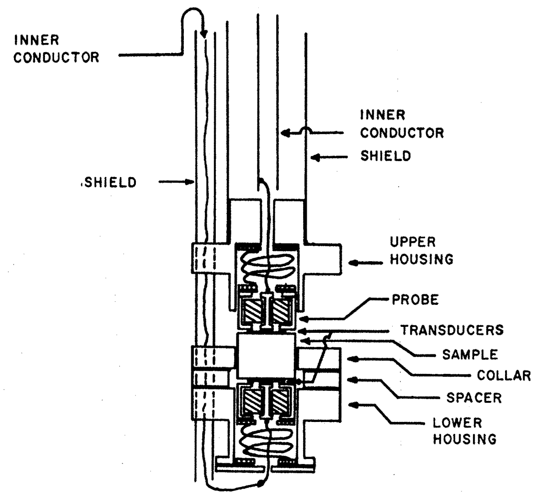

FIG. 3. Sample holder and supporting assembly.

The relative amount of electrical coupling R, measured by comparing the amplitude of the direct electrical pulse with that of the first acoustic echo using a pulse technique, was about 20% of the acoustic signal at zero field for this run. If the electrical coupling is reduced by more careful attention to grounding, or by balancing out some of the electrical signal with a bridge circuit, the velocity shifts measured for two adjacent peaks turn out to be much more consistent, as data presented below will show.

The explanation for this behavior lies in the fact that the amplitude as well as the velocity of the acoustic signal changes as a function of magnetic field. Since the detector output is the resultant of the acoustic and electrical signals, it depends on the phase and amplitude of each. The reason that adjacent resonance peaks show different behavior is that they occur for odd and even numbers of half-wavelengths within the sample, so that the acoustic signal reverses phase for alternate peaks. These effects will now be shown explicitly.

One can express the voltage at the receiving transducer as the resultant of two signals: an electrically coupled signal $E_c e^{i\phi}$, where ϕ is the phase of this signal referred to the voltage at the transmitting transducer, and the acoustic signal E_a, where

$$E_a = E_0 \sum_{m=0}^{\infty} \left[e^{(iq-\alpha/2)L}\right]^{(2m+1)}.$$

α is the energy attenuation coefficient and m is the number of round trips of the acoustic wave through the sample following successive reflections (assumed per-

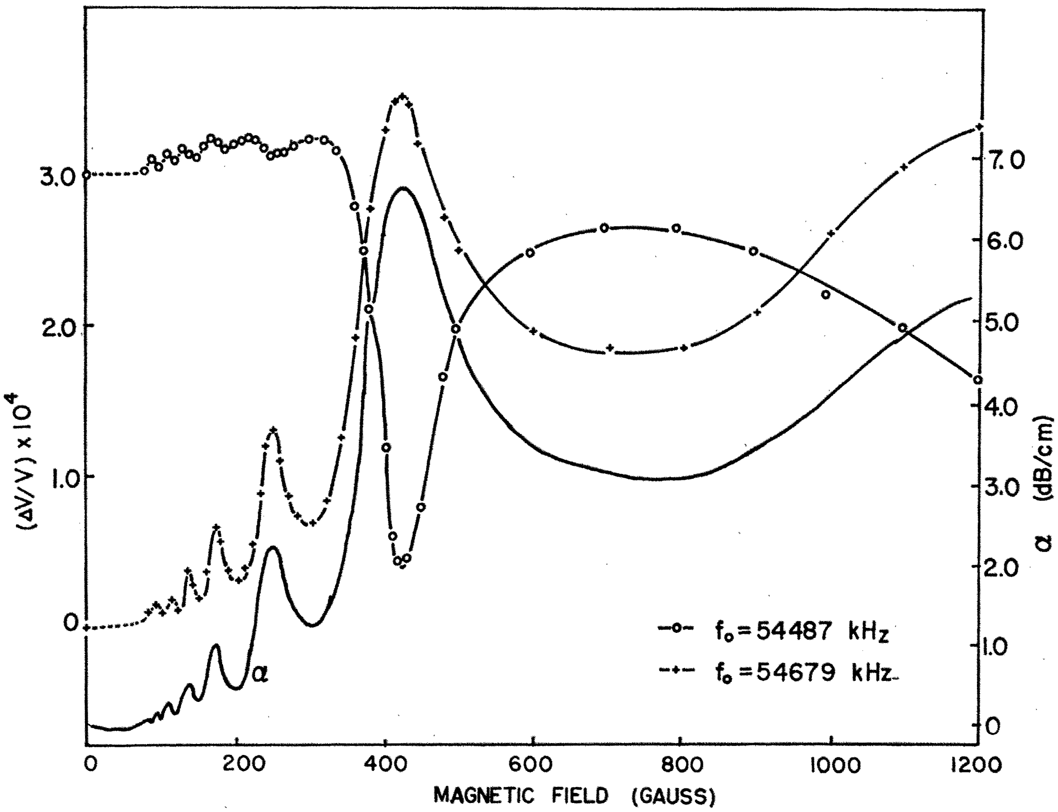

FIG. 4. Measured velocity change $\Delta V/V$ and attenuation α of 55 MHz compressional waves in Cd at 4.2°K versus magnetic field intensity; $\mathbf{q}\|[1\bar{2}10]$, $\mathbf{B}_0$ is 30° from $[10\bar{1}0]$ in the $(1\bar{2}10)$ plane. Measurements of $\Delta V/V$ are shown for two adjacent standing-wave resonances with the 54 487-kHz data shifted upward 3×10^{-4} for clarity.

fect) at the surfaces. This sum can be reduced to

$$E_a=E_0(\sinh\tfrac{1}{2}\alpha L\cos qL+i\cosh\tfrac{1}{2}\alpha L\sin qL)/2(\cosh\alpha L-\cos 2qL).$$

The resultant signal is

$$E_R=E_ce^{i\phi}+E_a.$$

The output power from the detector will be proportional to $|E_R|^2$, which reduces to

$$|E_R|^2=E_c^2+\frac{E_0^2+2E_cE_0[e^{\alpha L/2}\cos(qL-\phi)-e^{-\alpha L/2}\cos(qL+\phi)]}{4(\cosh\alpha L-\cos 2qL)}.$$

Clearly, if $E_c=0$, the resonances will occur for

$$\cos 2qL=1,$$

or

$$qL=n\pi,$$

which corresponds to $L=n\lambda/2$, as expected. The shift in the resonance value of qL attributable to the electrically coupled signal will be designated as δ, where $\delta=qL-n\pi$. Then

$$\frac{|E_R|^2}{E_0^2}=\frac{E_c^2}{E_0^2}+\frac{1\pm(2E_c/E_0e^{-\alpha L/2})[\cos(\delta-\phi)-e^{-\alpha L}\cos(\delta+\phi)]}{4(\cosh\alpha L-\cos 2\delta)},$$

where the upper sign is for even n and the lower sign for odd n. Note that the amplitudes of alternate resonances differ by an amount proportional to E_c, which gives a means for judging the amount of electrical coupling present in an experiment.

The locations of the maxima are found by taking the derivative of the expression above with respect to δ and setting it equal to zero. This yields

$$\{1\pm(E_c/E_0e^{-\alpha L/2})[\cos(\delta-\phi)-e^{-\alpha L}\cos(\delta+\phi)]\}\sin 2\delta+[\sin(\delta-\phi)+e^{-\alpha L}\sin(\delta+\phi)]\sinh\alpha L=0 \quad (5)$$

(under the assumption that E_c and α are slowly varying functions of frequency, so that their derivatives may be neglected).

Now it is apparent that the value of δ which satisfies Eq. (5) will be a function of the attenuation α, which is a function of B_0 unless ϕ is 0 or π. In order to permit one to judge the apparent velocity shift $(\Delta V/V)_A=\delta/n\pi$ resulting from the dependence of the resonance frequencies on α, a family of curves showing $(\Delta V/V)_A$ as a function of α for different values of ϕ is plotted in Fig. 5. One can see that the effect of the field dependence of α can be reduced by keeping the electrical coupling to a minimum, and keeping its phase near zero or 180°. Whether this has been achieved can be determined by making measurements with two adjacent resonances and comparing them, since the results will differ only if electrical coupling is effective. Furthermore, as Fig. 6 shows, for reasonably small $R=E_c/E_0e^{-\alpha L/2}$ the apparent shifts for successive maxima will be of approximately equal magnitudes but in opposite directions; thus a rather good value for the true velocity shift can be obtained by averaging the measurements for successive resonance peaks. The apparent shifts will be exactly equal and opposite for $\phi=90°$.

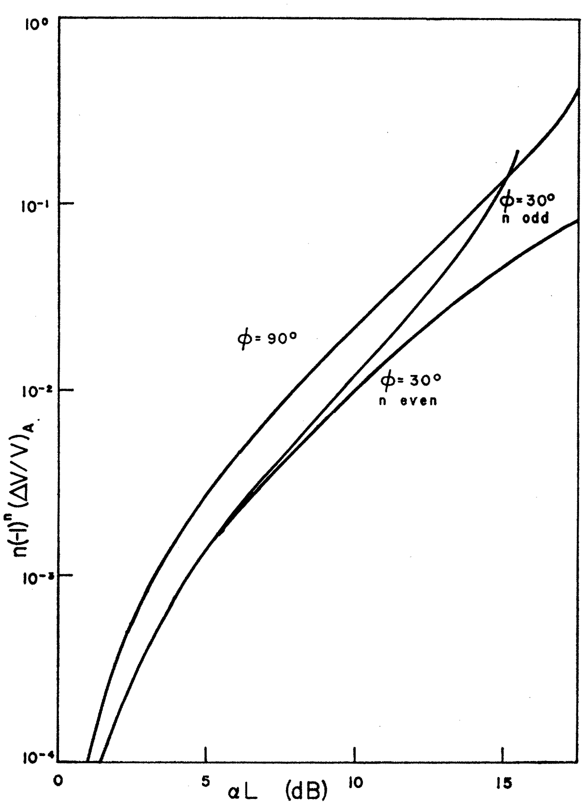

FIG. 5. Apparent velocity shift $(\Delta V/V)_A$ resulting from electrically coupled signal with $R\equiv E_c/e^{-\alpha L/2}=0.01$ as a function of total attenuation αL, where L is the sample length, n is the number of half-wavelengths of sound in the sample, and ϕ is the phase of E_c. The effect for other values of R can be found approximately by adding $\frac{1}{3}[20\log_{10}(R/0.01)]$ to αL.

We can now attribute the large discrepancy between the two velocity curves in Fig. 4 to the presence of an appreciable electrically coupled signal in that experiment. Presumably, similar effects were present in the work of Beattie and Uehling.[6]

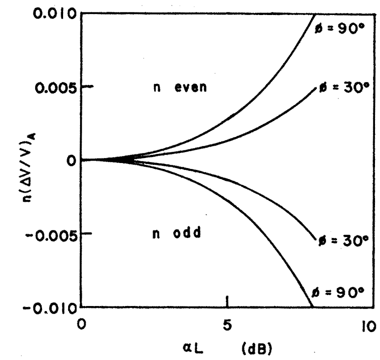

FIG. 6. Apparent velocity shift caused by electrical coupling versus total attenuation for $R=0.01$. For these small values of αL, the shifts are essentially equal in magnitude but opposite in sign for alternate resonances.

In the work reported here, attempts were made to reduce direct electrical coupling to the point where results obtained using adjacent resonances were in good agreement, and then these results were averaged to obtain better values for the actual velocity shifts.

B. Results for Cd

After the electrically coupled signal was reduced so that $R\approx 0.02$ at 33 MHz, the results shown in Fig. 7 were obtained. Here the magnetic field is perpendicular to [0001], so that an open-orbit resonance appears near 650 G. The curves for the two different resonance peaks are quite similar to one another and to the attenuation curve.

In Fig. 8, the averages for the two resonance peaks used in Fig. 7 are indicated by the +'s, while the results from a different run with different transducer bonds, etc., are indicated by the O's. It is clear that we have achieved reproducibility of both phase and amplitude of the velocity oscillations.

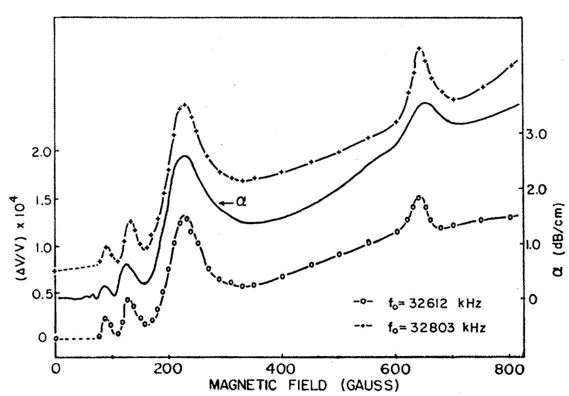

FIG. 7. Relative velocity shift in Cd versus magnetic field as measured for two adjacent resonances with $\mathbf{q}\|[1\bar{2}10]$, $\mathbf{B}_0[10\bar{1}0]$. The upper curve is shifted upward for clarity. The attenuation α is also shown for comparison.

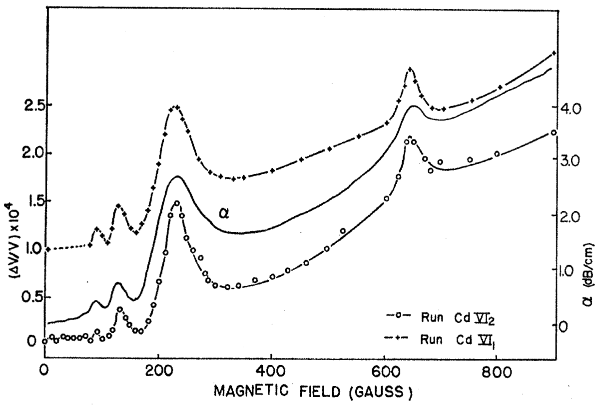

FIG. 8. Relative velocity changes in Cd for two different experimental runs compared with the attenuation α as a function of magnetic field. Run Cd VI_1 is the average of the curves in Fig. 7 shifted upward 10^{-4} for clarity.

C. Results for Cu

All of the measurements reported here in Cu were made with longitudinal sound propagating along [110] and $\mathbf{B}_0\|[001]$. Figure 9 shows the velocity shift for two adjacent resonances at 33 MHz with $R=0.04$. The attenuation is also shown for comparison. Another run where the coupling was reduced so that $R=0.02$ is shown in Fig. 10. Averages of the data for the two runs are plotted against f/B_0 in Fig. 11. The period of the oscillations corresponds to a momentum of 1.39×10^{-19} g cm/sec, which agrees within the experimental uncertainty with the value obtained by Bohm and Easterling[9]: 1.374×10^{-19} g cm/sec.

4. SUGGESTIONS FOR FURTHER IMPROVEMENTS

Our results show that, with proper care, it is possible to measure changes in the sound velocity with sufficient precision to study magnetoacoustic effects in pure metals. In order to make this technique as convenient as that using the attenuation, however, a straightforward method for recording data as a function of field intensity must be developed.

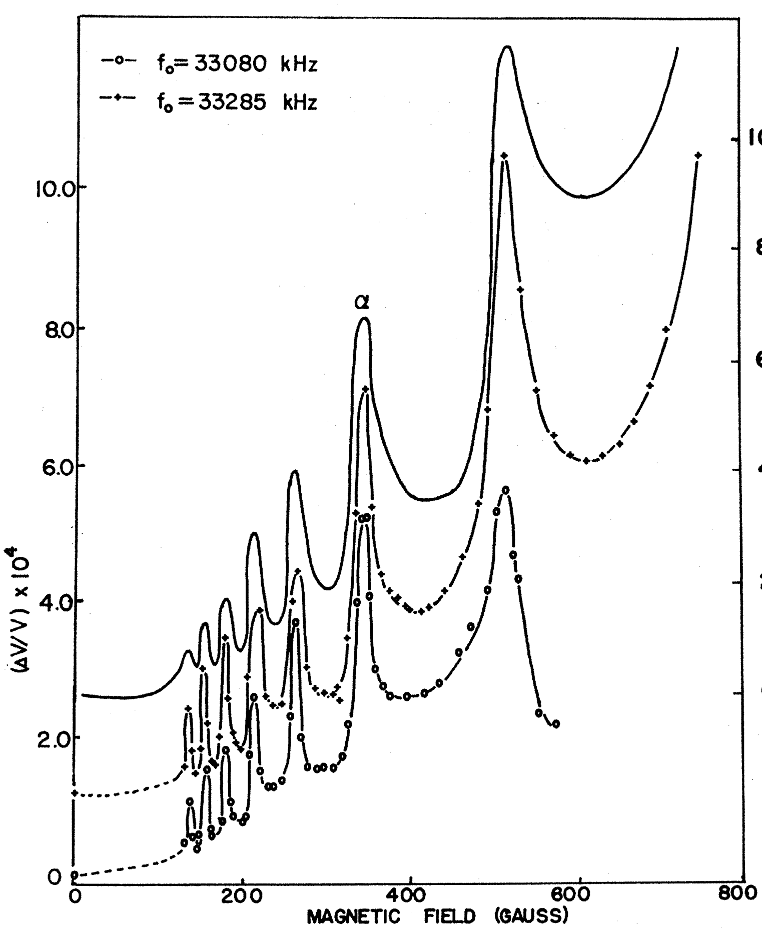

FIG. 9. Relative velocity shifts for 33-MHz compressional waves in Cu at 4.2°K compared with the attenuation: $\mathbf{q}\|[110]$, $\mathbf{B}_0\|[001]$, $R=0.04$. The curve for 33 285 kHz has been shifted upward for clarity.

[9] H. V. Bohm and V. J. Easterling, Phys. Rev. **128**, 1021 (1962).

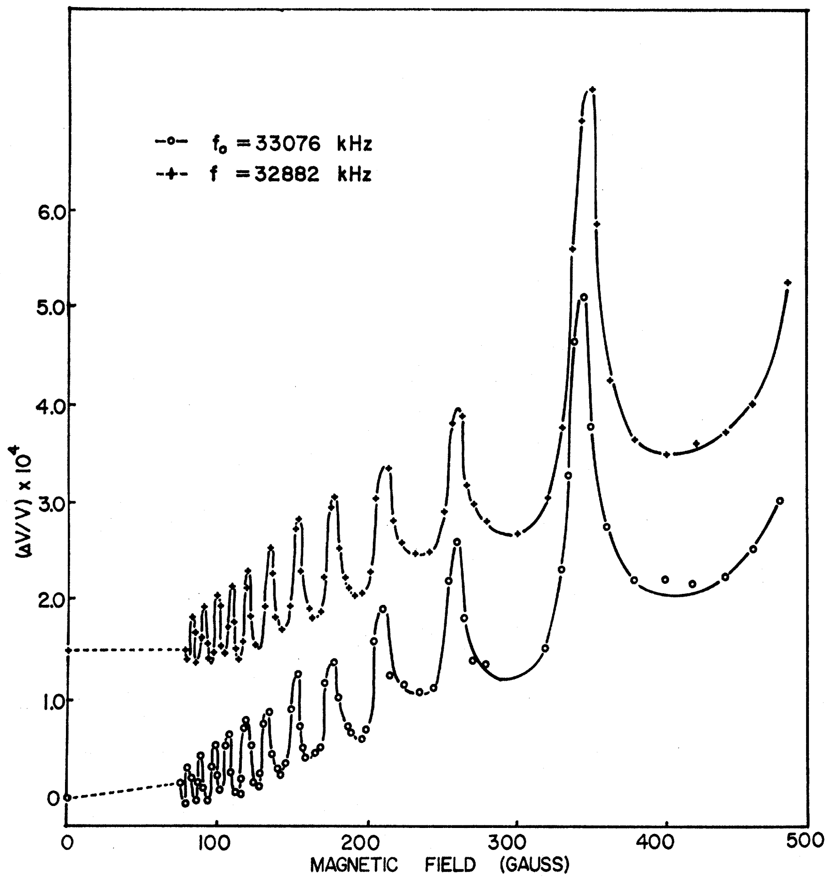

FIG. 10. Relative velocity shifts measured for the same geometry as in Fig. 9, but with electrical coupling reduced to $R=0.02$. The upper curve is displaced 1.5×10^{-4} upward for clarity.

We have made some progress in this direction by using the output of the phase-sensitive detector to control the center frequency of the FM oscillator. There is a tendency for the system to oscillate as a result of phase shift in the feedback loop, but this can be eliminated by proper shaping of the system response function. One can then sweep the magnetic field and record the velocity either in digital fashion by counting the output frequency of the oscillator, or in analog fashion by attaching a frequency-to-voltage converter to the oscillator and recording its output.

Another variation of this technique which is being explored eliminates the frequency modulation of the oscillator and relies on phase-sensitive detection at the oscillator frequency. Again the output of the phase-sensitive detector can be used to control the oscillator frequency in order to keep the system in resonance when the sound velocity changes. If one also measures the amplitudes of the signals for adjacent resonances (which, of course, include some signal due to electrical coupling), it should be possible to determine both velocity and attenuation using a continuous-wave scheme. One would then know the frequency of the ultrasonic wave much more precisely than is possible with a pulsed signal, with consequent improvement in the precision of magnetoacoustic data, particularly that involving sharp resonance effects.

Simultaneous measurements of velocity and attenuation may also shed further light on relaxation phenomena associated with magnetoacoustic effects. The real and imaginary parts of the sound propagation vector $\mathbf{q}$ are related through $\omega\tau$, at least for $\omega\tau\ll1$ and $ql\gg1$ as shown in Eq. (1). If the effects of nonspherical Fermi surfaces can be properly accounted for, it may be possible to measure ultrasonically the effective relaxation times of various groups of electrons in metals.

5. CONCLUSIONS

The change in the velocity of sound in Cd and Cu as a function of magnetic field intensity has been measured under conditions where $ql\gg1$. The velocity oscillates in the same manner as the attenuation under similar conditions; therefore the periods of these oscillations can be used to determine Fermi-surface extremal dimensions.

The magnitude of the velocity shifts (about one part in 10^4) is easily observable if proper precautions are taken regarding the effects of stray electrical signals. A standing-wave resonance technique which takes ad-

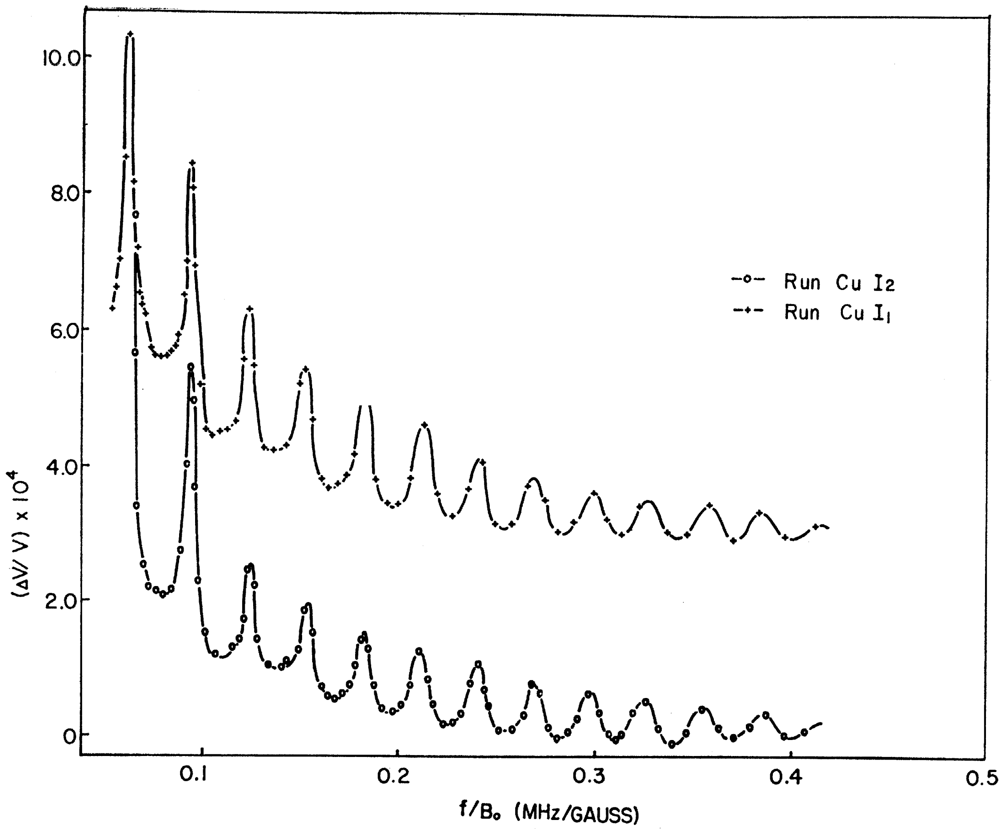

FIG. 11. Average of the velocity shifts in Fig. 9 (run Cu I_1) and Fig. 10 (run Cu I_2) plotted against frequency divided by magnetic field intensity. The upper curve has been displaced upward for clarity.

vantage of the phase reversal between resonances having even and odd numbers of half-wavelengths within the specimen is shown to be successful. A method for adapting the technique for automatic data recording is presented, along with suggestions for simultaneous measurements of velocity and attenuation of sound waves.

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