Magnetoacoustic Oscillations in the Velocity of Sound in Aluminum*†

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Small oscillations in the longitudinal velocity of sound have been observed as a function of magnetic field strength in aluminum at 100 Mc/sec. These oscillations are shown to arise from geometric resonances and are analogous to the oscillations in the attenuation arising from the same mechanism. The dimensions of the Fermi surface of aluminum calculated from these oscillations agree with previously published results. A comparison of the oscillations in the velocity with those in the attenuation indicates that the shift in the velocity is more sensitive to the geometric resonances than is the attenuation.

I. INTRODUCTION

THE dependence of acoustic wave propagation in metals on the strength and orientation of an applied magnetic field has been observed and studied rather extensively since the first unambiguous observations[1] and interpretations[2–4] of magnetoacoustic effects were made. There are several types of behavior of which the most interesting and significant would appear to be those which are oscillatory in character. Under appropriate conditions, both the sound velocity and attenuation oscillate in magnitude as the applied magnetic field is varied. These oscillations are periodic in the reciprocal of the field.

One distinguishes here three types of oscillation: (a) geometric resonances involving the ratio of cyclotron orbit diameter and acoustic wavelength; (b) cyclotron resonances involving the ratio of acoustic frequency and cyclotron frequency; and (c) de Haas–van Alphen-type oscillations which are a consequence of quantization of electron orbits. In each case the period of oscillation is determined by properties of the Fermi surface. The property most directly involved in each of the three types of oscillation are respectively: (a) a linear dimension of the Fermi surface; (b) the effective electron mass of an orbit on this surface; and (c) a cross-sectional area.

Since the attenuation is most easily measured, nearly all of the attention so far has been on this aspect of acoustic wave propagation. All three types of oscillations have been observed, and in one case[5] two types of oscillations were observed in the same experiment. The oscillations arising from the geometric resonances have been observed in many metals and have contributed greatly to the knowledge of their Fermi surfaces.

Measurements on the velocity of sound are in a somewhat different category. Only the de Haas–van Alphen oscillations[6] have been observed and these only in a few cases. Since there is no frequency requirement for the observation of this type of oscillation, the effect can be studied at frequencies sufficiently low for the acoustic attenuation to be small. This allows the employment of conventional methods for measurement of the velocity of sound such as the sing-around system or other pulse-echo techniques. In order to observe geometric resonances, much higher frequencies are required since a basic condition for observation of such oscillations is $ql \gg 1$, where q is the acoustic wave number and l is the electron mean free path. However, this is also the condition for large contributions to the acoustic attenuation by the electrons. This large attenuation greatly adds to the experimental difficulties in observing the effect of the geometric resonances upon the velocity of sound and precludes the use of pulse echo techniques.

The purpose of this paper is to report what is perhaps the first observation of oscillations in the acoustic velocity due to geometric resonances. The metal selected for this purpose was aluminum, a choice based partly on the fact that a single crystal of sufficiently long relaxation times was available, and partly on the fact that the results of previous Fermi-surface studies, both acoustic[7] and electromagnetic absorption,[8] were available for comparison. Insofar as the dimensions of the Fermi surface are concerned, the present results agree with previous results for all electron orbits observed. However, no additional orbits were observed and no further mapping of the Fermi surface was attempted.

Experimental difficulties precluded the determination of the exact shape of the velocity shift curves as a function of magnetic field. However it was possible to reproduce the oscillatory portions of the curves with sufficient accuracy to allow a comparison with the oscil-

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[1] R. W. Morse and J. D. Gavenda, Phys. Rev. Letters **2**, 250 (1959); Darrell H. Reneker, Phys. Rev. **115**, 303 (1959).
[2] A. B. Pippard, Proc. Roy. Soc. (London) **A257**, 165 (1960).
[3] T. Kjeldaas, Jr., and T. Holstein, Phys. Rev. Letters **2**, 340 (1959).
[4] M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960).
[5] B. W. Roberts, Phys. Rev. Letters **6**, 453 (1961).
[6] J. G. Mavroides, B. Lax, K. J. Button, and Y. Shapira, Phys. Rev. Letters **9**, 451 (1962); G. A. Alers and R. T. Swim, *ibid.* **11**, 72 (1963).
[7] B. W. Roberts, Phys. Rev. **119**, 1889 (1960); G. N. Kamm and H. V. Bohm, *ibid.* **131**, 111 (1963); N. A. Bezugly, A. A. Galkin, A. I. Pushkin, and A. I. Khomchenko, Zh. Eksperim. i Teor. Fiz. **44**, 71, (1963) [English transl.: Soviet Phys.—JETP **17**, 50 (1963)]; K. Fossheim and T. Olsen, Phys. Status Solidi **6**, 867 (1964).
[8] F. W. Spong and A. F. Kip, Phys. Rev. **137**, A431 (1965).

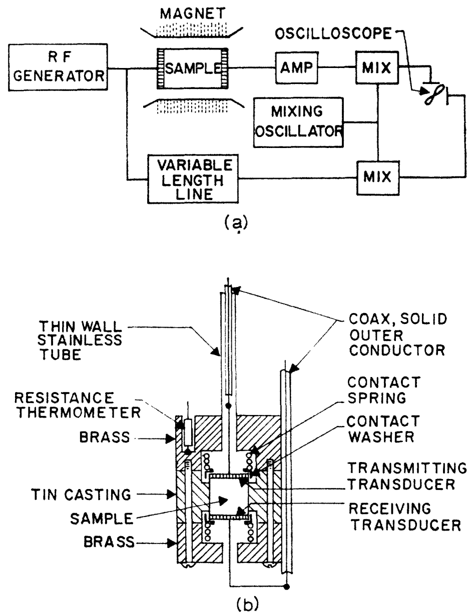

FIG. 1. (a) Block diagram of the experimental apparatus, and, (b) details of the sample holder assembly.

lations in the attenuation curves. This comparison indicates that the velocity shift is more sensitive to the geometric resonances than is the attenuation, a result which does not appear to be contained in the theory.[4,9]

II. EXPERIMENTAL METHOD

The choice of an experimental technique to search for velocity shifts in a magnetic field was dependent upon two considerations. First, the electron contributions to the acoustic attenuation at helium temperatures in pure metals at frequencies of the order of 100 Mc/sec is extremely high. Second, the theory shows that the velocity shifts can be very small. In order of magnitude the electronic contribution to the attenuation is 40 dB/cm and the velocity shift is a few parts per million. Pulse techniques do not appear to be suitable under these circumstances. Consequently, a continuous wave phase comparison technique was employed.

A block diagram of the apparatus is given in Fig. 1. An rf signal is applied to a quartz transducer bonded to the sample. The ultrasonic signal produced is transmitted through the sample where it is detected by a receiving transducer. The output of this transducer is then amplified and mixed with the signal from a mixing oscillator. The 50-kc/sec difference signal is applied to the Y axis of an X-Y oscilloscope. The signal for the reference phase is run through a variable-length constant-impedance coaxial line and mixed with the output of the mixing oscillator. This signal is applied to the X axis of the oscilloscope. In was found that the sensitivity of the apparatus could be increased by slightly overdriving the reference phase mixer. This produced a difference signal with a small amount of second-harmonic content. The addition of this second harmonic changed the zero phase difference Lissajous figure from a straight line to a thin figure eight. The relative proportions of the lobes of the figure eight, and thus the position of the crossing point, are very sensitive to small changes in phase, a 10° phase shift being enough to change the figure eight into a slightly distorted ellipse. Thus by amplifying the figure eight and looking at the change in position of the crossing point, great sensitivity to small changes in phase is obtained.

To measure the phase shifts, the crossing point is set at the center of the oscilloscope face, a magnetic field is applied to the sample, and the crossing point returned to the center of the picture by changing the length of the variable length line. The magnitude of this length change in the reference line then gives the fractional velocity change in the sample. It is

$$\Delta_v/v = v\Delta x/cL\,,$$

where Δx is the change in the line length and L is the length of the sample. The sensitivity of the system is such that a change in the length of the reference line of 0.02 cm can be seen on the oscilloscope. This corresponds to a fractional shift in the velocity of 5×10^{-7} for a sample 1 cm long with an acoustic velocity of 6.6×10^5 cm/sec.

To measure the attenuation the amplitude of the signal was read directly from the face of the oscilloscope. The amplitude response of the system was checked with precision attenuators and found to be linear over the range of interest. The magnetic field was measured with a Rawson-Lush rotating-coil gaussmeter whose accuracy was checked periodically by means of a NMR proton probe.

This phase comparison technique is possibly the only method capable of measuring velocity shifts with the required accuracy in the presence of the attenuations encountered. However, in order to measure these velocity shifts, one of two conditions is required. Either the attenuation must not change when the velocity changes or all of the signal on the line coming from the receiving transducer must be associated with the ultrasonic path. Any signal of another phase on this line will result in the reflection of the changes in the attenuation into the measured phase shift. Since large changes in the attenuation are present, the problem becomes one of isolating the transmitting and receiving lines and transducers from each other.

This problem of isolation is not trivial. Because of the high attenuations involved, the necessary rf isolation for good data is around 120 dB over the one centimeter length of the sample. The main paths for the signal leakage proved to be from the transmitting to the receiving transducer, both around the sample and through the sample. The latter was by capacitative coupling and

[9] Sergio Rodriguez, Phys. Rev. **130**, 1778 (1963); **132**, 535 (1963).

was apparently allowed by the oxide coating on the aluminum sample preventing it from being perfectly grounded. In order to remove both leakage paths as much as possible, the cylindrical surface of the sample was scored and the sample cast into a block of tin. This sample assembly is shown in Fig. 1. Tin was used as the shielding material because its thermal expansion coefficient is slightly higher than that of aluminum, its tensile strength slightly lower, and it is not superconducting at 4.2°K. Because of the stretching of the tin with temperature cycling, a new casting was made every time the sample was cooled to helium temperature. Even so, some leakage signal always appeared to be present.

The sample was a single crystal of aluminum in the form of a cylinder about 1 cm in diameter and 1 cm long. It was oriented with a [110] crystal axis along the axis of the cylinder. The crystal orientation in the plane perpendicular to the cylinder axis was determined by means of x-ray diffraction. This orientation was correlated with the angular dependence of the attenuation at 12 kG. It was then possible to use this angular dependence on subsequent runs to determine the orientation to within $\frac{1}{2}°$.

The geometry of the experiment was set up so that the propagation vector $\mathbf{q}$ of the longitudinal sound wave was along the cylinder axis of the sample, which is a [110] crystal axis. The magnetic field lies in a ⟨110⟩ crystal plane perpendicular to the cylinder axis. The orthogonal crystal axes in this plane are a $[\bar{1}10]$ and a [001] axis. The orientation of the crystal about the cylinder axis is specified by giving the angle between the field and the $[\bar{1}10]$ axis. This is also the angle between the [001] axis and the line defined by the intersection of the plane of the electron orbit and the ⟨110⟩ plane.

The sample was grown from 99.9999% pure aluminum and the original residual resistivity ratio, as measured by the eddy current decay method,[10] was $\rho_{300}/\rho_{4.2}=5200$. During the course of the experiment the ratio decreased to a final value of 2300. We believe this to be due to small stresses on the sample caused by the contraction of the tin shielding upon cooling. In the course of some 30 experimental runs it is thought that these stresses caused enough new dislocations in the lattice to account for the reduction in the resistivity ratio.

The experiment was performed at frequencies between 99.5 and 100.5 Mc/sec and at a temperature of 4.2°K. The transducers were X-cut quartz, coaxially plated, with a fundamental frequency of 20 Mc/sec. They were bonded to the sample with a Dow Corning 200 fluid having a viscosity of 200 000 centistokes. This bonding material was preferred to Nonaq stopcock grease because the bond did not break as often during cooling. No other differences were observed between the two bonding agents.

[10] C. P. Bean, R. W. De Blois, and L. B. Nesbitt, J. Appl. Phys. **30**, 1976 (1959).

The electronics were of standard design. A pair of stub tuners were used in both the receiving and transmitting lines to match impedances. Because this was a continuous-wave experiment, the rf power had to be kept below a value of the order of 2W to keep the rate of helium evaporation at a level which was not excessive.

III. EXPERIMENTAL RESULTS

The experimental curves for the measured phase shifts as a function of magnetic field showed large variations. Not only do the over-all shapes of the curves depend on the frequency at which the measurement is made but occasionally there is a large change between one run and the next. A major factor responsible for these variations is the presence of a small amount of leakage signal. The experimental problem is complicated by the fact that the zero field attenuation of the direct signal as it passes through the sample under test is of the order of 35 dB in these experiments and that it varies by almost this amount with changes in the magnetic field. Consequently, the presence of a leakage signal will lead to measured phase changes which are due in part to the changes in the attenuation of the acoustic signal and in part to changes in the velocity of sound. Analysis of this effect shows that in the presence of a leakage signal as small as 5% of the direct acoustic signal, phase shifts will occur because of changes in attenuation which are larger than those due to changes in velocity. However, this analysis also shows that the undesired effect can be minimized, and in principle reduced to zero, by choosing the relative phase of the leakage and acoustic signals to be close to 0° or 180°. The condition of optimum relative phase is obtained by varying the frequency, successive frequencies of 0° and 180° phase difference occurring with each increase of a half-wavelength of sound in the sample, or a $\Delta\nu$ of about 330 kc/sec in this experiment. At these frequencies of optimum phase difference, the previously observed uncontrolled variations in the shape of the curves appear to be minimized. At the same time an oscillatory behavior as a function of magnetic field is observable. All further data were taken at these optimum frequencies.

This is not to say however that all of the experimental problems of phase shift measurement have been solved. There are still variations in the over-all shapes of the phase shift curves, even at the optimum frequencies. While the analysis predcits some variation in curve shape as a consequence of uncontrolled changes in the magnitude of the leakage signal, the changes observed appear to be larger than the analysis would predict. Also, there is occasionally an apparent reversal in the sign of the velocity shift which is unexplained. This sign was constant for a given experimental run, depending neither on the particular optimum frequency chosen nor on small frequency variations around this optimum frequency. Because of these presently unexplained aspects of the experimental data, only the oscillatory

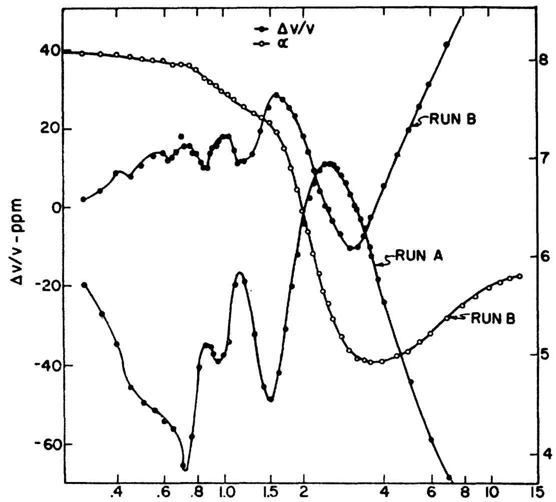

FIG. 2. Examples of curves for the apparent fractional velocity shift and the attenuation as a function of magnetic field. Two different experimental runs are shown. The attenuation curve for run A was essentially the same as that for run B. These curves were taken at an orientation of 0°.

part of the fractional velocity shift curves can be regarded as having any physical significance. No attempt is made to interpret the over-all behavior of the curves.

Figure 2 is a sampling of the experimental data showing oscillations in the fractional velocity shift. Data are given for two different experimental runs, both at the same sample orientation. As can be seen from the curves, the over-all sign of the fractional velocity shift and the shape of the curves is different for the two runs. However, analysis of the data showed that the period of the oscillations in inverse magnetic field was the same for both runs. Figure 3 is a plot of run B as a function of inverse field and clearly shows the oscillatory behavior of the curves. In Fig. 2 an attenuation curve

TABLE I. Comparison of extremal momenta from velocity-shift measurements with values reported by Kamm and Bohm. The sound propagation vector in both cases is along a [110] direction.

Orbit angle from [100] axis (deg)	Experimental dimensions and estimated errors (10^{-19} g cm/sec) Primary orbit	Secondary orbit	Data from Kamm and Bohm[a] (10^{-19} g cm/sec) Primary orbit	Secondary orbit
0	1.44±0.03	not seen	1.45±0.02	0.67±0.03
10	1.55±0.08	0.75±0.05	1.50±0.02	0.72±0.03
20	1.44±0.05	0.75±0.05	1.45±0.05	0.79±0.03
30	1.19±0.08		1.20±0.03	
40	1.08±0.06		1.07±0.03	
50	not seen	1.13±0.06	1.02±0.06	1.18±0.06
60	1.14±0.06		1.11±0.02	
70	0.56±0.04	not seen	0.54±0.02	1.15±0.04
80	0.49±0.04	not seen	0.50±0.02	1.24±0.04
90	0.47±0.04		0.47±0.02	

[a] Data and errors not reported numerically were taken from Fig. 1 of Kamm and Bohm.

for run B is also shown. The attenuation curves at a given orientation for different experimental runs duplicated each other closely enough to be considered identical.

The one thing that was consistent in the experimental data was the period of the oscillations in inverse field for any given orientation. The magnitude of the period suggests that the oscillations are of the geometric type. If they are in fact of this type, maxima or minima in the velocity shift will occur at integral values of n in the equation[11]

$$\hbar k=(e\lambda/2c)(n+\gamma)B\,,$$

where λ is the acoustic wavelength, B the magnetic field, γ is a phase factor that depends upon the shape of the orbit, and $\hbar k$ is the effective average momentum of the dominating electron orbits, usually the extremal momentum, on the Fermi surface. The direction of $\hbar k$ is perpendicular to the plane containing the sound propagation vector and the magnetic field vector.

Thus, one searches the data for the presence of one or more periods in $1/B$. From these periods one calculates k values corresponding to a given crystal orientation. The results obtained from the velocity data in aluminum are listed in Table I. Each value is an average for all data taken at the given orientation. Also included in the table are values taken from Kamm and Bohm's investigation[7] of the Fermi surface of aluminum by magnetoacoustic attenuation. While agreement between the two sets of data is good, several orbits seen by Kamm and Bohm were not apparent in the velocity shift curves. This was not unexpected as the value of ql in this experiment was around 3 to 6 while Kamm and Bohm had a value around 60.

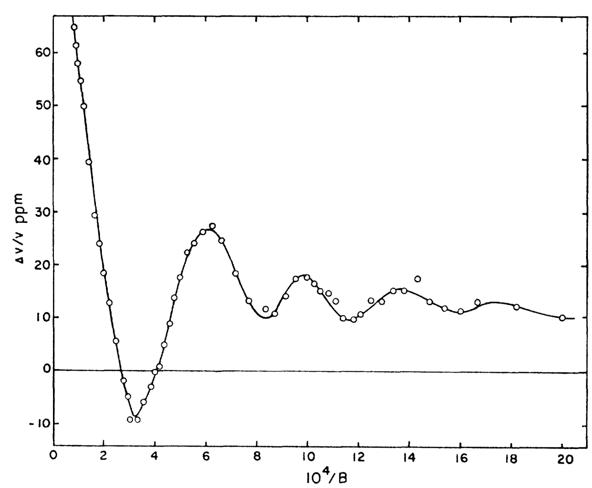

FIG. 3. The apparent fractional velocity shift shown in run B, Fig. 2, replotted as function of inverse magnetic field. For this sample, a phase shift of one degree corresponds to a change in $\Delta V/V$ of 20 ppm.

[11] A. B. Pippard, *Low Temperature Physics*, edited by C. Dewitt, B. Dreyfus, and P. G. deGennes (Gordon and Breach, Science Publishers, Inc., New York, 1962).

Figure 4 is a plot of the oscillation number n as a function of $1/B$ for runs A and B. The slope of these curves is $2c\hbar k/e\lambda$ and therefore depends only upon one variable, the Fermi momentum. One sees that both slopes are the same. However, the values of γ for the two curves are different. These two runs showed the largest variation in γ that occurred between the runs at any one orientation, but all orientations showed some variation. At present this variation in γ is not understood as γ is expected to be a function only of the shape of the orbit.

In Fig. 2, experimental curves are given both the fractional velocity shift and the change in the attenuation for run B. An examination of these curves shows only a suggestion of two peaks in the attenuation curve while the velocity shift has at least four distinct oscillations. A similar behavior is observed for all orientations investigated. Under the present experimental conditions, 100 Mc/sec and a relaxation time of 1 to 4×10^{-11} sec, the value of ql lies between 2 and 8. The theory[3] shows that for values of ql in this range, oscillations in the attenuation should be barely observable. The attenuation curve shown in Fig. 2 is in reasonable agreement with theory.[12]

There has been no theoretical investigation of the fractional velocity shift for small values of ql. For the limit $ql\gg1$, free-electron theory for longitudinal magnetoacoustic effects[4,9] predicts that the fractional velocity shift and the change in the attenuation should have the same functional dependence on the magnetic field. The data taken in this experiment are in definite disagreement with this prediction. At least in aluminum, the fractional velocity shift appears to have a greater sensitivity to the geometric resonances than does the attenuation. At present it is not known whether this increase in sensitivity is in disagreement with the basic theory of magnetoacoustic effects or only in disagreement with evaluations from the theory at the limit of $ql\gg1$.

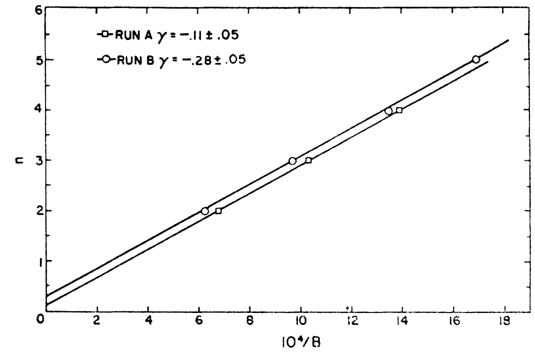

FIG. 4. The oscillation number n for the velocity shift curves shown in Fig. 2 plotted as a function of inverse magnetic field. The phase factor γ is given by the negative value of the curve at infinite field.

IV. CONCLUSIONS

The results presented here indicate a need for further studies of the magnetoacoustic effect. More accurate experimental measurements are needed of the velocity shift arising from the geometric resonances. While one may be limited to the techniques used here, it should be possible to improve the apparatus. It would be desirable to see the velocity shift in several metals and to examine it as a function of frequency and electron relaxation time. There is also a need for further theoretical work. While the Cohen, Harrison, and Harrison formalism predicts the same dependence on magnetic field for both the velocity shift and the change in attenuation in the limit $ql\gg1$, it is not clear that the formalism predicts this relation for smaller values for ql.

ACKNOWLEDGMENT

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[12] The agreement of the attenuation curve with theory is only for the low-field portions of the curve. The attenuation above 3 kG for this orientation disagrees with free-electron theory. The high-field attenuation in aluminum will be discussed in more detail in a future paper.