Magnetoacoustic Effect in Cesium*

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The magnetoacoustic effect has been observed in high-purity cesium single crystals. Longitudinal waves of frequencies 30–70 MHz were employed. The momentum obtained from the period of the geometric oscillations indicates that the Fermi surface of cesium is distorted from a sphere, bulging about 4% in the [110] direction and depressed approximately 1% in the [100] and [111] directions. The results are compared with the de Haas–van Alphen measurements in cesium, and the positions of the attenuation extrema are compared with the free-electron theory of ultrasonic attenuation.

I. INTRODUCTION

DURING the past seven years a great many experimental investigations have been conducted on the alkali metals in order to determine their Fermi surfaces. Because lithium and sodium undergo martensitic phase transformation at low temperatures, the investigations have been confined mostly to potassium and, to a lesser degree, to rubidium and cesium. In the case of potassium, cyclotron resonance,[1] de Haas–van Alphen,[2] and magnetoacoustic experiments[3–8] indicate that the Fermi surface is very nearly spherical. In addition, the various ultrasonic attenuation experiments are consistent with the free-electron theory of the ultrasonic attenuation due to conduction electrons. Schoenberg and Stiles[2] also employed the de Haas–van Alphen technique to study the Fermi surface of rubidium. Variations in the cross-sectional area were reported and these data were used to calculate the radial anisotropy. The surface was found to bulge in the [110] direction by 0.95% and was depressed in the [100] and [111] directions by 0.23 and 0.46%, respectively. More recently, similar measurements in cesium have been reported by Okumura and Templeton.[9] Their results indicate a probable radial distortion of +3.3% in the [110] direction and −0.9 and −1.4% in the [100] and [111] directions. These distortions are considerably smaller than the theoretical predictions of Ham,[10] but larger than the distortions predicted by Heine and Abarenkov.[11]

Because the magnetoacoustic technique provides a way of directly measuring the extremal calipers of the Fermi surface, a study of the magnetoacoustic effect in cesium was undertaken. This technique essentially makes use of the interaction of the conduction electrons and the electric fields associated with high-frequency ultrasonic waves. In the presence of a magnetic field which is applied perpendicular to the direction of propagation of the ultrasonic wave, the electrons will orbit in real space, and the radius of the orbit in k space is related to the radius of the orbit in real space by the relation $k=(eH/\hbar c)R$. Whenever $R=(n+\phi)\lambda/2$, extrema in the ultrasonic attenuation occur. Minima in the attenuation will occur when n is half an odd integer and maxima when n is an integer. The attenuation is thus oscillatory and periodic in $1/H$. The frequency of the oscillations is proportional to an extremal value of k in the direction $\mathbf{q}\times\mathbf{H}$. The phase of these oscillations is given by ϕ which is independent of $\mathbf{H}$ in the high phase region. In potassium, the experimental values of the positions of the extrema are in excellent agreement with the corresponding values predicted by the Cohen, Harrison, and Harrison[12] theory of the ultrasonic attenuation.

In this paper we compare the results of our magnetoacoustic experiments in cesium with the free-electron theory of ultrasonic attenuation to examine the effects caused by small distortions of the Fermi surface from a spherical shape.

II. EXPERIMENTAL PROCEDURE

The techniques employed for growing the cesium single crystals were similar to those employed in the study of the elastic constants[13] of cesium done at this laboratory. The cesium metal used in the present study was of higher purity, however, and single crystals were slightly more difficult to grow. The cesium had a nominal purity of 99.98+ and was obtained from Pierce Chemical Company in 15-g vials. The crystals were grown by a modified Bridgeman technique in an argon atmosphere. A two-piece furnace was used to facilitate removing the crystals from the furnace. The upper piece consisted of a stainless-steel tube 7 cm in length

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with a 1.5-cm i.d., and the other piece, a mild steel plunger, contained the conical nucleating tip. The stainless-steel tube was wound with resistance wire and alternate layers of asbestos and aluminum foil. Since cesium has a low melting temperature, 28°C, a heat sink was attached to the mild steel plunger to lower the temperature below room temperature in order to obtain a high temperature gradient. The heat sink consisted of a long aluminum rod upon which was wound a small heating coil. The upper end of the rod was attached to the plunger and the lower end was placed in contact with dry ice which was placed in a plastic container and vented to the atmosphere. By adjusting the power to the heaters on the furnace and on the aluminum rod, a high temperature gradient could be maintained during the entire growth process. Briefly, a crystal was grown by first coating the furnace with cured vaseline and preheating the furnace to 80°C. The glass ampoule containing the high-purity cesium was then broken and the cesium transferred into the furnace. The system was then held at 70°C for 1 h before removing power from the heater to start the growth process. Approximately 4 h were needed to complete the growth process.

Upon removal from the furnace, the crystal was immediately placed in a test tube containing mineral oil in which ten drops of isopropyl alcohol were added as an etchant, and the crystal was examined for grain boundaries. The crystal was placed in a two-circle goniometer, submerged in mineral oil in a lucite tank, and optically oriented. The optical data were plotted on a stereogram and the proper rotations made to determine the closest crystallographic direction to the boule axis. The flash planes were found to be (110) planes and usually reflections from six planes were observed.

Once the proper orientation was found, acoustic samples were prepared along the appropriate symmetry direction, both [110] and [100] samples being prepared. Using a string saw, a thin shim was first cut perpendicular to the appropriate symmetry direction and a Laue transmission x ray was then taken of the shim to check the orientation. An XR-7 Polaroid Laue camera made it possible to obtain x-ray patterns with exposure times between 3 and 5 min. Once the orientation was established, the string saw was used for cutting specimens from the boule, which was held firmly on an aluminum table by vaseline and silicon grease at dry-ice temperatures. The string was kept moist with an etching solution of 20 parts of light mineral oil, 20 parts of xylene and 20 parts of isopropyl alcohol. The specimen, held firmly in a lapping ring with a 1:1 mixture of vaseline and Dow Corning 200 fluid (60 000 centistokes), was then lapped to obtain flat and parallel faces suitable for acoustic measurements. The lapping process was carried out on an aluminum plane maintained at dry-ice temperatures and on which an etchant of mineral oil and isopropyl alcohol was placed. The entire preparation of the acoustic specimens was carried out in an argon atmosphere.

After being lapped, the acoustic specimen was cleaned of any mineral oil in a bath of xylene and quickly dried and placed on the cold aluminum plate. A Dow Corning 200 fluid (100 000 centistokes) was then applied to the faces of the specimen. The transducers were in turn carefully positioned on the faces of the crystal with a very slight pressure, to insure a good bond without destroying the faces of the crystal. The specimen was then mounted in a spring-loaded ultrasonic probe and immersed in a cup of liquid nitrogen. Next, the system was removed from the dry box and the coaxial cables on the probe were attached to the coax fittings on the bottom of two coaxial stainless-steel tubes which provided rf leads to the probe and also supported the sample in a glass helium Dewar which was placed between the pole faces of a 12-in. Harvey Wells magnet. The geometry was such that the direction of propagation $\mathbf{q}$ of the ultrasonic wave was perpendicular to the magnetic field $\mathbf{H}$. The magnet could be rotated about an axis parallel to the direction of propagation $\mathbf{q}$ so that $\mathbf{H}$ could be placed along a desired crystallographic axis.

A two-transducer ultrasonic pulse-echo technique was employed to study the variation of the attenuation with magnetic field. Two matched coaxially plated $\frac{1}{2}$-in. x-cut transducers with a fundamental frequency of 10 MHz were used to generate and receive the longitudinal waves in the specimen. An Arenberg ultrasonic pulse generator was used to activate one of the transducers. The acoustic pulses received by the second transducer were passed through a crystal mixer and into a high-gain linear i.f. amplifier. The output of the amplifier was displayed on an oscilloscope and also fed into a boxcar integrator to gate out an echo. The output voltage from the integrator, which is proportional to the echo height, was fed to the y axis of an x-y recorder. The x axis was driven by the voltage from a Hallprobe so that as the field was varied, a direct plot of pulse height versus magnetic field was obtained. An HP 608-D pulse generator was used to calibrate the y axis in decibels and to measure the frequency of the ultrasonic wave. The magnetic field was measured with a 0.1% Rawson-Lush gaussmeter. Once the attenuation measurements were completed, a thin shim was cut parallel to the acoustic face and x rayed to determine the final orientation of the direction of propagation relative to the crystallographic axis. Two of the crystals for which data is reported, crystal I ($\mathbf{q}$ parallel to [001]) and crystal II ($\mathbf{q}$ parallel to [011]), were within 3° of the appropriate crystallographic axes. For these crystals, the longitudinal sound velocity was obtained from the known elastic constants at 4.2°K. These velocities are $V_{110}=1.41\times10^5$ cm/sec and $V_{100}=1.14\times10^5$ cm/sec. For a third crystal, the direction of propagation was approximately 8° from a [001] direction. The attenuation measurements were made at 70 MHz where ql was appreciably greater than unity and a large number of oscillations were observed. The velocity of sound,

$V_{11}'=1.28\times10^5$ cm/sec, was calculated from the elastic-constant and x-ray data.

Although a large number of oscillations were observed only in the three crystals reported above, oscillations were observed in a total of eight single-crystal acoustic specimens. There are many experimental reasons why high ql values are difficult to obtain. The attenuation in cesium is large at low temperatures, and the metal is extremely soft and reactive, thus making it difficult to obtain well-oriented crystals with flat parallel faces. The major difficulty encountered in generating high ultrasonic frequencies, however, was the acoustic bond and/or the breaking of bonds at liquid-helium temperatures. Since recycling a cesium crystal to liquid-helium temperatures results in a shorter electron mean free path, it was necessary to obtain two good bonds on the first attempt if high ql values were to be obtained. In any case, the data obtained from all the crystals is consistent with the measurement reported in Sec. III for crystals I, II, and III.

III. RESULTS

Ultrasonic frequencies between 10 and 70 MHz were employed and the measurements were taken at 1.3 and 4.2°K. At 4.2°K the attenuation was found to decrease monotonically with increasing magnetic field for all frequencies. At 1.3°K, however, the attenuation was oscillatory for all frequencies greater than 10 MHz. Since ql must be greater than 3 to observe geometric oscillations, the data indicates that l, the electron mean free path, is phonon limited at 4.2°K and increases by a factor of 3 or 4 as the temperature is lowered to 1.3°K. This increase in the electron mean free path is consistent with the residual resistance measurements that were made on similar cesium samples using an eddy-current technique.

At 1.3°K, the high-field attenuation is greater than the zero-field attenuation for frequencies 30, 50, and 70 MHz. Since the free-electron theory predicts that the high-field attenuation is equal to the zero-field attenuation when $ql=6.8$, the mean free path can be estimated. The present measurements show that $ql=6.8$ between 20 and 30 MHz for the samples studied and therefore $l=5\times10^{-3}$ cm.

At 30 MHz, the attenuation is oscillatory at low magnetic fields and saturates at high fields at a value slightly higher than the zero-field attenuation. The oscillations are more pronounced and more numerous at the higher ultrasonic frequencies and the high-field attenuation is much larger than the zero-field attenuation. Thirteen easily observable extrema could be detected at 70 MHz and ten at 50 MHz. This dependence of the attenuation on ql is quite similar to the data obtained in potassium and to the results predicted by the free-electron theory. The position of the first minimum in the attenuation which occurs at high fields is also a function of ql. This minimum shifts to lower fields for intermediate values of ql as l is increased. This effect is predicted by the free-electron theory and was also observed in potassium.[4] This latter effect was studied by holding the frequency constant and varying the temperature.

Unlike the results observed in potassium, which has a spherical Fermi surface, the positions of the extrema and the background attenuation are dependent on the direction of **H**. Plots of the relative attenuation versus **H** for crystal I for various orientations of **H** in the (001) plane are shown in Fig. 1. The first two extrema which occur at higher magnetic fields are not shown. The fourfold symmetry exhibited by these curves is indicative that **q** is very close to the [001] direction and these symmetry plots were an invaluable aid in determining the actual orientation of **H** in the (001) plane. It will be noted that when **H** is close to a [110] direction, the oscillations actually decrease in amplitude at large magnetic fields. This effect is most pronounced for the third extremum ($n=\frac{3}{2}$). Figure 2 shows this effect at 70 MHz. The upper curve is for $\mathbf{H}\|[110]$ and the lower curve is for $\mathbf{H}\|[100]$. While this washout is most pronounced when **H** is parallel to a [110] direction, it still exists as **H** is rotated approximately 15° on either side of this direction. Figures 1 and 2 clearly show that the washout is definitely associated with the extremum, $n=\frac{3}{2}$, that is, it occurs at higher magnetic field at high ultrasonic frequencies and lower fields for lower ultrasonic frequencies, and its position is constant in λH. Initially, it was believed that this effect was somehow related to the phase of the oscillations which should be related to the shape of the electron orbit on the Fermi surface and hence to the direction of **H**. However, when crystals were studied with **q** parallel to [011], the same effect was observed, but now with **H** parallel to a [100] direction. In both cases the period of the oscillations yields a caliper in direction $\mathbf{q}\times\mathbf{H}$ or [110] direction. This amplitude effect was observed in all the single crystals studied. In addition, several of these crystals were studied in later experimental runs and while there was a deterioration in the electron mean free path, the washout was observed. In general, the washout occurs whenever **H** is along such a direction that the oscillations are associated with the P_{110} calipers. When **q** is parallel to a [011] direction, however, a slight decrease in amplitude at large fields is also apparent when **H** is parallel to [110]. Okumura and Templeton also reported a variation of signal amplitude with crystallographic direction in the study of the de Haas–van Alphen effect in cesium. For magnetic fields parallel to the [111] direction no signals were observed. However, except for the possible effects of crystal distortion, the two phenomena do not appear to be related.

The present experiments were conducted on crystals which were taken from three different single-crystal boules. Since the attenuation exhibited the symmetry of the crystals, it does not appear that the wash out is associated with local strains in the crystals. In any case,

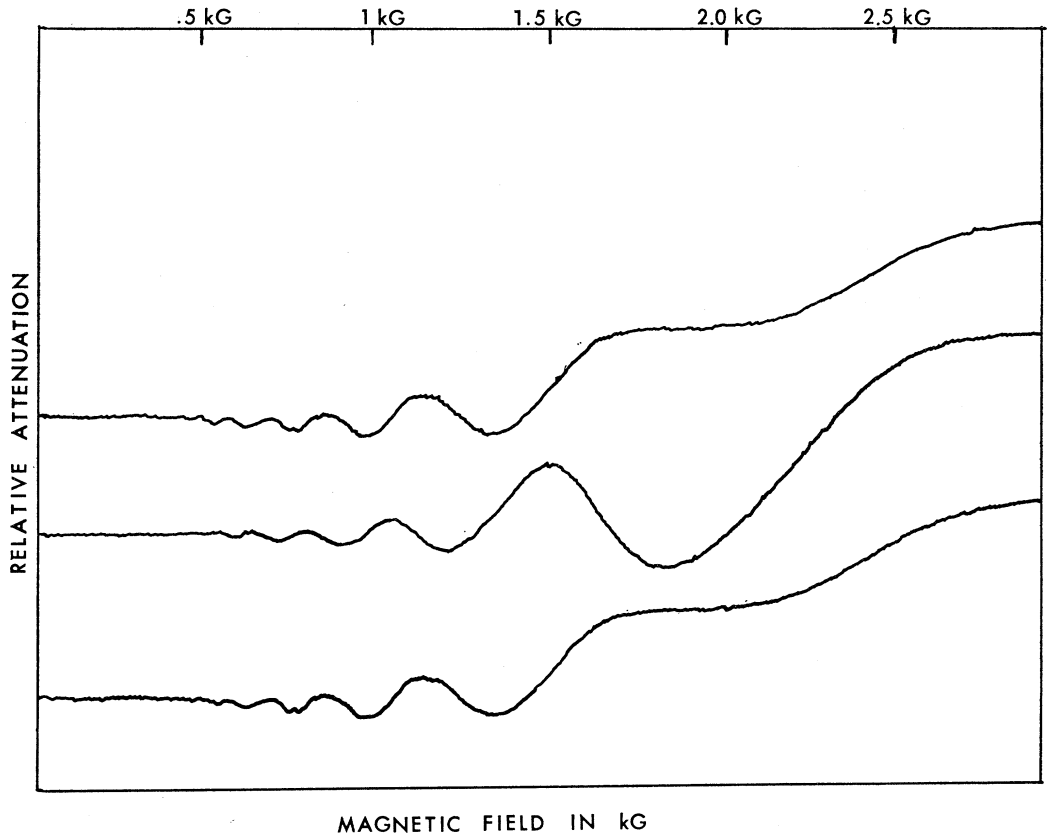

FIG. 1. Relative attenuation of 50-MHz longitudinal waves versus magnetic field for various orientations of **H** in the (001) plane and $\mathbf{q}\|[001]$. Curve 1: $\mathbf{H}\|[110]$; curve 2: $\mathbf{H}\|[100]$; curve 3: $\mathbf{H}\|[1\bar{1}0]$. The curves have been shifted for clarity.

geometric oscillations are periodic in $1/H$ only in the high-phase region, and the first three extrema are not useful in determining the periods of the oscillations. The decrease in amplitude of the geometric oscillation at high fields for particular directions of H and the anisotropy of the limiting attenuation at high fields represent departures from the free-electron theory of ultrasonic attenuation.

In order to compare accurately the frequency of the oscillations for various orientations of **H**, several plots of the pulse amplitude versus magnetic field were made for a given direction of **H**. The magnet was then rotated and several plots were again made on the same graph. This procedure was repeated for various sweep ranges of the magnetic field. The shifts in the extrema could easily be detected and the results were reproducible. As **H** was rotated in the (001) plane from [100] toward [110], the extrema in the attenuation shifted toward higher magnetic fields, as seen in Figs. 1 and 2. The maximum change occured when the magnetic field was rotated from 15° of [110] to the [110] direction. Since the positions of the extrema are proportional to the momentum for a given direction of **H**, this effect was not entirely unexpected. An analysis of all the data for the various crystals indicates that the phase of the oscillations, which also affects the positions of the extrema, depends on the direction of **H**, and the magnitude of the shifts was found to be both a function of the momentum and the phase. For large n, however, the phase is small compared to n so that the positions of these extrema could be used to map out the anisotropy in the momentum, the extrema associated with high momentum occuring at higher fields. When **q** was parallel to [011] and **H** was rotated in the (011) plane from [110] to [100], the shifts were in a direction which indicates that $P_{110}>P_{100}>P_{111}$. It is worthwhile pointing out that no variation of the positions of the extrema could be detected in potassium as **H** was rotated in a plane perpendicular to **q**. The momentum is related to the period of the oscillations which was determined from a least-square fit of the extremum number versus $1/H$ plots. For reasons of internal consistency, the same extrema and an equal number of extrema were used to determine the periods for various orientations of **H**. Usually six to eight extrema were used. The plots were very linear and the probable error in the slopes was between $\frac{1}{2}$ to 1%. The results for three of the crystals studied are shown in Table I. The momenta shown are in directions of maximum distortion of the Fermi surface. For directions of **H** not near these directions,

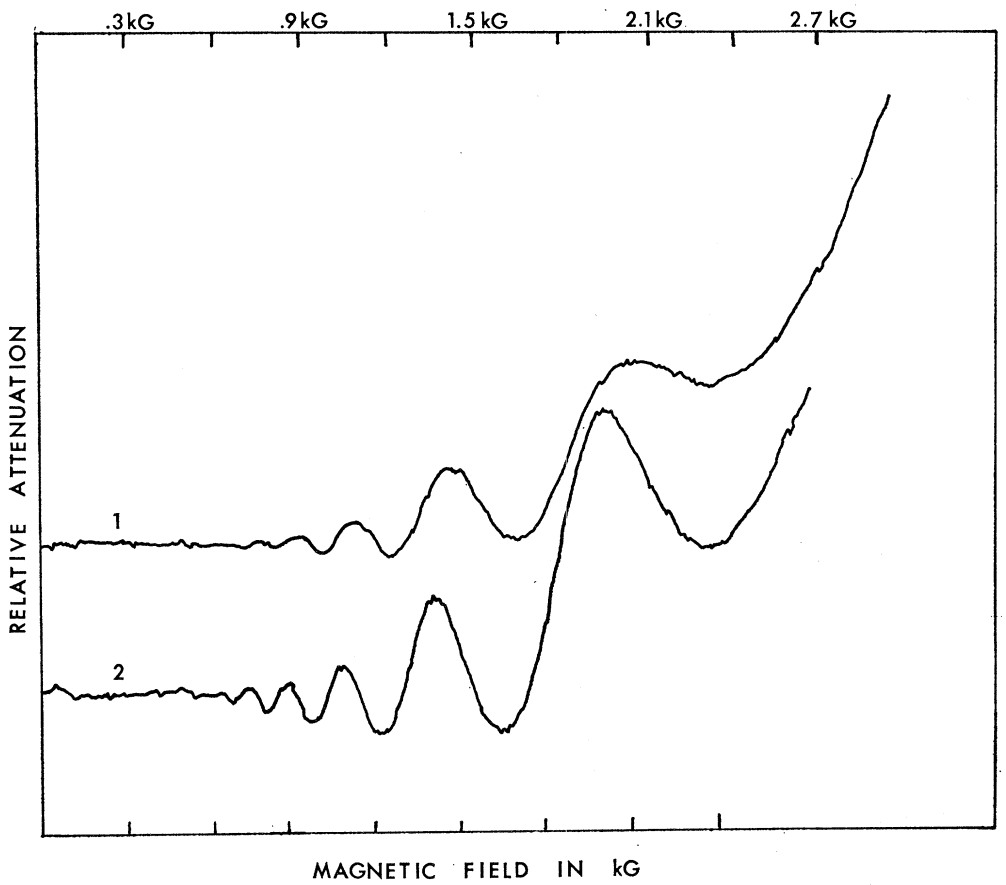

FIG. 2. Relative attenuation of 70-MHz longitudinal waves versus magnetic field with $\mathbf{q}\|[001]$. The upper curve is for $\mathbf{H}\|[110]$ and the lower curve for $\mathbf{H}\|[100]$. The curves have been shifted for clarity.

no noticable shifts in the extrema can be detected, and the periods of the oscillations are essentially the same, except when **H** is within 15° of the [110] direction. The momenta listed are known to about 5% when one takes into account errors in the sound velocity, ultrasonic frequency, periods in $1/H$ and the orientation of **H**. The agreement between the values of the momenta in a given direction for the three crystals is well within the estimated accuracy. The radial anisotropy is known much more accurately than the absolute value of the Fermi momentum since it depends only on the accuracy to which the periods can be measured for a given crystal. The acoustic measurements show that a maximum distortion between 4 and 6% in the Fermi surface exists with a bulge in the [110] direction of approximately 4 or 5% and a 1% depression in the [100] direction and a slightly larger depression in the [111] direction. These results are slightly larger but within experimental error agree with the results of Okumura and Templeton. They are, however, substantially smaller and different from the band calculations of Ham. The radial anisotropy of the Fermi surface of cesium obtained by Okumura and Templeton was represented by contours on a sterogram in the (011) plane. Their results indicate that the bumps in the [110] direction in cesium are more isolated than in rubidium and extend about 20° from the [110] direction. The present acoustic experiments are not nearly precise enough to map out the extension of these bumps, but, as pointed out earlier, the maximum shifts in the extrema of the attenuation occur when **H** is within 15° from a [110] direction.

A comparison of the experimental attenuation curves with the predictions of the free-electron theory can be made by examining the positions of the attenuation extrema. Table II lists the theoretical and experimental values in terms of qR. The theoretical values were taken

TABLE I. Fermi momenta in units of 10^{-20} g cm sec^{-1} for directions of maximum distortion in the Fermi surface. The free-electron value of the momentum is 6.83×10^{-20} g cm sec^{-1}.

Axis	Crystal I	Crystal II	Crystal III
[110]	6.93	6.92	7.05
[100]	6.68	6.52	6.75
[111]	...	...	6.77

TABLE II. Comparison of the theoretical and experimental positions of the extrema associated with various measured calipers in terms of qR.

		qR			
		Crystal I		Crystal III	
n	Theory	P_{110}	P_{100}	P_{110}	P_{111}
$\frac{1}{2}$	2.70	2.54	2.77	2.85	2.48
$\frac{3}{2}$	5.80	5.63	6.18	5.85	5.58
$\frac{5}{2}$	8.95	8.60	9.28	8.68	8.86
$\frac{7}{2}$	12.10	11.73	12.36	11.86	11.88
$\frac{9}{2}$	15.31	14.70	15.33	14.89	14.95
1	4.00	3.84	3.95	4.18	3.89
2	7.25	6.48	7.52	6.94	7.14
3	10.45	10.03	10.69	10.18	10.40
4	13.61	13.29	14.04	13.22	13.81
5	16.77	16.44	17.08		

from the work of Flax and Trivisonno[14] for $ql=13$. The experimental values listed are for data associated with P_{110}, P_{100}, or P_{111} calipers for three crystals. The experimental values of qR for each column were calculated from the measured $1/H\lambda$ values and the appropriate momentum. For $n>3$, the agreement between the theory and experiment is well within the estimated uncertainty of 3 or 4% in the experimental values of these quantities. For $n<3$, the phase of the oscillations becomes an appreciable fraction of n and there is a significant departure from the theory. For directions of **H** not associated with these calipers, fairly good agreement with theory is obtained for all values of n. The portion of the extremal orbit associated with these directions of **H** in the sound-wave front is essentially circular so that this is not entirely unexpected. The limiting phase was also determined from the intercept of the n-versus-qR plots. While these are not known accurately, the phase was found to be dependent on the direction of **H** and was smallest for **H** parallel to the [110] direction. The value of the phase varied between 0.25 (**H** parallel to [110]) and 0.40. The free-electron phase is 0.37. There is also a departure from the theory in the limiting attenuation at high fields. For **q** parallel to [011] the attenuation is the largest when **H** is along a [110] direction and smallest when **H** is along the [100] direction. The frequency at which the limiting attenuation was equal to the zero-field attenuation was also dependent upon the direction of **H**. The mean free path could be different in these directions or more likely the response of the electrons on different portions of the Fermi surface to the electric fields associated with the strain is different. A quantitative treatment of the magnetic field dependence of the ultrasonic attenuation in cesium will be the subject of another investigation.

In summary, the radial anisotropy obtained in the present experiment is in agreement with the deHaas–van Alphen data. In addition, at low magnetic fields (high-phase region) the positions of the extrema in the attenuation are in agreement with free-electron theory but the distortions of the Fermi surface cause very large anisotropic effects in the ultrasonic attenuation at high magnetic fields.

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