Magnetoacoustic Effect and Ultrasonic Attenuation in Potassium*

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The magnetic field variation of ultrasonic attenuation has been observed at 4.2°K and 1.2°K in high-purity single crystals of potassium. Longitudinal waves of frequencies 10 to 50 Mc/sec were propagated in four crystals oriented along [100] and along [110], the magnetic field being perpendicular to the direction of propagation. The attenuation was found to be an oscillatory function of the magnetic field strength and periodic in $1/\lambda H$. The Fermi momentum was found to be 7.6×10^{-20} g cm/sec, in reasonable agreement with the free-electron value. No shifts in the positions of the extremal values of the attenuation were observed as the magnetic field was rotated in the (100) or (110) plane, indicating that the Fermi surface of potassium is spherical to better than 1%. The $1/H\lambda$ values for the extrema and the magnetic field dependence of the ultrasonic attenuation were found to be in agreement with the free-electron theories of ultrasonic attenuation.

INTRODUCTION

THEORIES[1–4] of ultrasonic attenuation of longitudinal waves due to conduction electrons have been given for the zero-magnetic-field case and for electrons in the presence of a magnetic field. For longitudinal waves in the absence of a magnetic field the attenuation caused by free electrons is

$$\alpha(0)=\frac{nm}{2\rho v_s\tau}\left[\frac{q^2l^2\tan^{-1}ql}{3(ql-\tan^{-1}ql)}-1\right], \quad (1)$$

where n is the number of free electrons per unit volume, m is the electron mass, v_s is the velocity of sound, ρ is the density, τ is the electron relaxation time, l is the electron mean free path, and q is the magnitude of the sound wave vector.

In the presence of a magnetic field, the attenuation is an oscillatory function of H and as $H\rightarrow\infty$, the attenuation saturates. In the free-electron case, the limiting value of the attenuation is given by

$$\alpha(H)=\frac{nmv_f}{30\rho v_s}q^2l. \quad (2)$$

The difference between (1) and (2) is easily measured and these measurements are reported in a later section.

These theories predict that for sufficiently high ql values, the attenuation is oscillatory and nearly periodic in $1/H\lambda$. The period of these oscillations, $\Delta(1/H\lambda)$, is related to the extremal dimensions of the Fermi surface by the relation[5]

$$P_f=e/2c\Delta(1/\lambda H)\,. \quad (3)$$

During the last few years, various experimental measurements of the Fermi surface of potassium have been made. The de Haas–van Alphen measurements of Schoenberg and Stiles[6] on single crystals of potassium indicate that there is a very small departure of the Fermi surface from a spherical shape. Cyclotron resonance measurements by Grimes and Kip[7] indicate that while the cyclotron mass is greater than the free-electron value, it is isotropic.

Foster, Meijer, and Mielczarek[8] recently reported some magnetoacoustic measurements at 4.2°K on zone-refined single crystals of potassium oriented along the [110] direction. Their measurements indicate that the Fermi momentum, within experimental accuracy, is in agreement with the free-electron value. If the Fermi surface of potassium is spherical, and the present study confirms that of other investigators in this regard, it should be possible to compare directly the observed variation of ultrasonic attenuation in a magnetic field with the free-electron theories of ultrasonic attenuation.

The present study, while supporting much of the work of Foster *et al.*, does extend and differ from it. The present study includes attenuation measurements on single crystals oriented along [110] and [100]. It also includes measurements at 4.2 and 1.2°K. These latter measurements show the variations of the magnetic field dependence of the ultrasonic attenuation with mean free path. The positions of the extrema, the strength of the oscillations, and the magnitude of the change in attenuation with magnetic field are also in accord with the free-electron theories of ultrasonic attenuation. In this regard the results differ from those reported by Foster *et al.*

EXPERIMENTAL PROCEDURE

The techniques for growing the single crystals and the preparation of the acoustic specimens were the same as those employed by this laboratory for the determi-

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nation of the low-temperature single-crystal elastic constants of potassium.[9,10]

The sample, mounted in a spring-loaded sample holder, was suspended between the pole faces of a 12-in. Harvey-Wells magnet in such a manner that the direction of propagation, **q**, of the ultrasonic wave was perpendicular to the magnetic field, **H**. The magnet could be rotated about an axis parallel to the direction of propagation, **q**, so that **H** could be placed along a desired crystallographic axis.

An ultrasonic pulse-echo technique was employed to study the variation of the attenuation with magnetic field. A coaxially plated $\frac{1}{2}$-in. x-cut transducer with a fundamental frequency of 10 Mc/sec was used to generate the longitudinal waves in the specimen. A silicone release agent, Dow Corning 7 Compound, was used to bond the transducer to the potassium sample.

A Matec transmitter-receiver set was employed to activate the transducer and to receive the acoustic pulses. The received signals were rectified and displayed on an HP-175A oscilloscope, and by means of a display scanner, an output voltage proportional to the desired echo height was fed to the y axis of an x-y recorder. In this way, as the field was varied, a direct plot of pulse height versus magnetic field was obtained. A comparison pulse generator with a calibrated attenuator was used to measure the relative attenuation and to calibrate the y axis of the chart in decibels. The pulse-generator was also used to measure the frequency of the ultrasonic wave.

Velocity measurements were made at liquid-helium temperatures during each experimental run. An Arenberg wide-band amplifier was used in place of the Matec receiver so that the unrectified pulse could be observed directly. Round trip transit times were measured at 10 Mc/sec with the delayed sweep of a Tektronix 545 oscilloscope. Velocity measurements were repeated at a later time with the buffer technique that was employed for the low-temperature elastic-constant study in this laboratory. The present velocity measurements agree with those previously reported if one takes into account the deviation of the actual direction of propagation from the crystallographic axis. The direction of propagation for the crystals studied deviated from the actual crystallographic axis from 3° to 5°. The sample lengths were measured at room temperature with a micrometer and the appropriate length correction for 4.2°K was made using the thermal expansion data of Swenson.[11] Measurements were made on two samples, I and II, oriented along [100] and on two samples, III and IV, oriented along [110].

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RESULTS AND DISCUSSION

Measurements were made on each of the crystals at 10, 30, and 50 Mc/sec at 4.2 and 1.2°K. At 10 Mc/sec the attenuation decreases monotonically with increasing magnetic field, saturating at 1 kG. At 30 and 50 Mc/sec the attenuation is oscillatory and is nearly periodic in $1/H\lambda$, with the attenuation increasing and then saturating a high magnetic fields. The oscillations become more numerous and more pronounced at the higher frequencies or ql values, and the limiting attenuation at high fields increases as ql is increased. Ten easily observable extrema were detected at 50 Mc/sec.

The effect of lowering the temperature on the magnetic field dependence of the ultrasonic attenuation is shown in Fig. 1. The frequency is 30 Mc/sec for both curves shown. The upper curve is for 4.2°K and the lower curve is for 1.2°K. It is readily seen that the oscillations are much more pronounced and more numerous at the lower temperature because of the increase in the mean free path. In addition, the value of the attenuation at high magnetic fields increases. These features are consistent with a larger ql value and in agreement with the results obtained when l remains fixed and the frequency is increased. No detectable shift of the extrema is observed with the exception of the first minimum which appears to be a function of ql.

This same effect was observed on all the crystals studied although the shift in the first minimum is much less pronounced at higher ql values. For each plot of attenuation versus field, the value of $1/H\lambda$ was calculated at each extremum point. Table I gives the results, at the highest ql value, for each of the crystals studied in this work; crystal I has the highest ql value and crystal IV the lowest. The $\frac{1}{2}$-integer values of n correspond to minima in attenuation and the integral values to maxima in attenuation. The value of a particular $1/H\lambda$ has a calculated precision of 4%, but the internal consistency appears to be better than this. The theo-

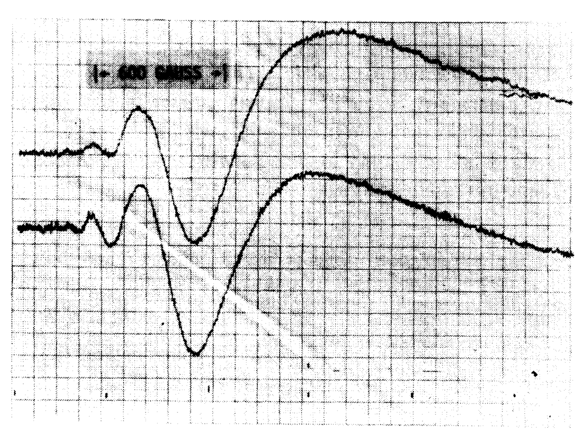

FIG. 1. Recording of signal amplitude versus increasing magnetic field. The upper curve is for 4.2°K and the lower curve for 1.2°K. The frequency is 30 Mc/sec. $\mathbf{H}\perp\mathbf{q}$ and $\mathbf{q}\parallel$ [100].

TABLE I. Comparison of theoretical and experimental values of $1/H\lambda$ for attenuation extrema.

			$(1/H\lambda)\times10^{-1}$ (G cm)$^{-1}$			
n	qR	Theory	Crystal I (100)	Crystal II (100)	Crystal III (110)	Crystal IV (110)
$\frac{1}{2}$	2.94	0.95	0.90	0.95	0.85	0.79
$\frac{3}{2}$	6.04	1.96	1.91	1.96	1.96	1.86
$\frac{5}{2}$	9.16	2.97	2.92	2.99	3.02	2.93
$\frac{7}{2}$	12.28	3.98	3.95	4.06	4.00	3.88
$\frac{9}{2}$	15.41	4.99	5.04	···	···	4.88
1	4.04	1.31	1.32	1.39	1.31	1.34
2	7.27	2.36	2.39	2.45	2.39	2.44
3	10.45	3.39	3.42	3.54	3.40	3.44
4	13.61	4.41	4.47	4.70	4.53	4.55
5	16.77	5.44	5.53	···	···	···

retical values of $1/\lambda H$ are also shown for comparison. The theoretical values were obtained from the qR values, tabulated by Cohen, Harrison, and Harrison (CHH), assuming $P_f=7.86\times10^{-20}$ G cm/sec, that is, the free-electron value of the momentum. It is clear that the theoretical positions of the extrema agree rather well with the corresponding experimental values of these quantities and are within the calculated precision of the experimental points. Foster *et al.* reported that their experimental values of $1/H\lambda$ for the first five minima differ from the theoretical values. In our measurements, the first attenuation minimum alone appears to differ significantly from the theoretical value.

An analysis of all the data for this extremum shows that the $1/H\lambda$ value increases as ql is increased. Figure 1 shows this effect quite directly. In addition, agreement with the theoretical value of CHH improves as ql increases. Kjeldaas and Holstein have calculated the magnetic field dependence of the attenuation of longitudinal waves for intermediate ql values. An examination of their theoretical plots reveals a similar shift in the position of the first extremum as ql increases. No significant differences in the positions of the other extrema with the theory of CHH are present. In this regard the present experiment appears to be consistent with the free electron theories of ultrasonic attenuation. Plots of extremum number versus $1/H\lambda$ are linear with the first extremum exhibiting the largest deviations from the straight line. This extremum was, therefore, given little weight in the momenta plots. This treatment of the data is consistent both with the findings of other investigators of the magnetoacoustic effect and with the results reported above and contrary to the results of Foster *et al.* who weighted this extremum the most in their momenta plots.

TABLE II. Experimental values of velocity of sound and Fermi momentum for the four potassium samples.

Sample	Velocity (10^5 cm/sec)	Frequency Mc/sec	Momentum (10^{-20} G cm/sec) $P_{f(\max)}$	$P_{f(\min)}$	P_f
I$_{(100)}$	2.28	50.0	7.6	7.7	7.7
II$_{(100)}$	2.24	50.0	7.3	7.7	7.5
III$_{(110)}$	2.66	50.0	7.5	7.6	7.6
IV$_{(110)}$	2.57	70.8	7.5	7.8	7.8

Table II shows the momenta calculated from the data of Table I. The column $P_{f(\max)}$ is the value of P obtained by plotting n versus $1/\lambda H$ for the maximum in attenuation and using Eq. (3). $P_{f(\min)}$ gives the corresponding value for the minima in attenuation. For all crystals studied, there is a definite trend in the data which indicates that the $\Delta(1/\lambda H)_{\max}>\Delta(1/\lambda H)_{\min}$. This result is consistent with the theory of CHH if one considers only the first 10 extrema to correspond with the experimental case. It can be seen from Table II that this leads to $P_{f(\max)}<P_{f(\min)}$. The last column lists the P_f obtained using all the extrema, but weighting the first two extrema less. The values listed in Table II are in reasonable agreement with the free-electron value of 7.86×10^{-20} G cm/sec and within the calculated precision of this experiment.

No noticeable shift in positions of the extrema can be detected if the magnet is rotated and a plot of pulse height versus field is made. In addition, no shift in any extremum can be observed if the field appropriate to that extremum is held constant and the magnet is slowly rotated. These results indicate that the Fermi surface of potassium is spherical to better than 1%, a conclusion consistent with that of other investigators.

To check the magnitude of the attenuation and its dependence on magnetic field, the difference in attenuation between $H=0$ and $H=14$ kG was measured as a function of frequency at 4.2 and 1.2°K. These measurements can be compared directly with the difference between Eqs. (1) and (2). A numerical calculation shows that the zero field attenuation, $\alpha(0)$, is equal to the limiting attenuation, $\alpha(H)$ when $ql=6.8$. For ql values <6.8, $\alpha(0)$ is greater than $\alpha(H)$, and for ql values >6.8, $\alpha(0)$ is less than $\alpha(H)$. The measured changes in attenuation were $+1.2$ dB/cm, -1.5 dB/cm, and -10.5 dB/cm at 10, 30, and 50 Mc/sec in sample I at 4.2°K. This implies that for sample I at 10 Mc/sec, $ql<6.8$ and ql is >6.8 at 30 and 50 Mc/sec. The best fit of the data with the difference between Eqs. (1) and (2) shows that $ql=13.5$, 8, and 2.7 at 50, 30, and 10 Mc/sec, respectively, and that l is about 10^{-2} cm at 4.2°K. Similar potassium samples had resistance ratios between room temperature and 4.2°K which ranged between 2500 and 3000. These measurements were made by an eddy current method[12,13] and indicate that the mean free path obtained from the acoustic measurements is very reasonable. The effect of lowering the temperature is to increase the mean free path, and an analysis of the attenuation data for $T=1.2$°K indicates that l and the corresponding ql values are increased about 20% over

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their values at 4.2°K. The number and strength of the oscillations observed for the relatively low frequencies employed in this study and the effect of lowering the temperature on the attenuation are indicative of samples of high mean free path. For these reasons, the electron mean free paths in the samples studied in the present work are believed to be longer than those reported by Foster *et al.*

While it is difficult to compare accurately the attenuation measurements obtained on crystals oriented in different directions, since the mean free path varies from specimen to specimen, we find that the attenuation of longitudinal waves of a given frequency propagated along [110] is not as large as in the [100] direction. This is expected because of the higher sound velocity and corresponding lower value of q in the [110] direction. In this regard, the results are consistent with the theory.

SUMMARY

The value of P_f obtained in this study is consistent with that of other investigators and agrees quite well with the free-electron value. The positions of the extrema in attenuation are in agreement with the corresponding theoretical values, and the relative change in attenuation and strength of the oscillations and their dependence on ql are in accord with the various free-electron theories.

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Calculation of Low-Lying Quantum States of the F_3^+ Color Center

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The low-lying quantum states of the F_3^+ color center have been calculated for the equilateral-triangular configuration in KCl. The calculation has been done under the assumption that the crystal is a dielectric medium with an effective dielectric constant. The possibility that the new optical absorption band appearing at 1.29 eV in KCl is due to the F_3^+ center is also discussed in this paper.

I. INTRODUCTION

EXPERIMENTALLY two new optical absorption bands have been found for KCl. These bands lie at 0.9 eV and 1.29 eV. Schneider and Rabin[1] have suggested that the new band occurring at 0.9 eV is due to a transition of the F_2^+ center, and that the second band appearing at 1.29 eV arises from a transition of the F_3^+ center.

In a previous paper,[2] the author calculated the energy of $1s\sigma$-$2p\sigma$ transition of the F_2^+ center in KCl to be 0.93 eV. This result suggests that this transition of the F_2^+ center may be responsible for the band occurring at 0.9 eV, in agreement with the general suggestion of Schneider and Rabin.

In the present work, the low-lying quantum states of the F_3^+ center have been calculated. The method used in this work is based mainly on a method given in the earlier paper.[2] In that study the negative-ion vacancy was considered to be a spherical region with unit positive charge. Furthermore, it was assumed that the interaction of the electron with phonons could occur only when the electron moved within the region far from the trap center, and that the interaction of the trap center with the phonons occurs regardless of the position of the electron. Obviously, this idea can also be applied to the other F-aggregate centers, such as F_3^+, F_3, and F_4 centers. If we do that, however, we would obtain complicated expressions and integrals. In this calculation, we avoid this trouble by introducing an effective dielectric constant [which is equivalent to a parameter (see part A of Sec. II)] for the ionic crystal under consideration, such that (a) the negative ion vacancies can be regarded as positive point charges immersed in a dielectric medium, and (b) the interaction of the F_3^+ center with the phonons occurs regardless of the position of the electron. The effective dielectric constant can be determined from the known quantum state of the F center.

To test this method, the energy of the $(1s\sigma)^2$-$(1s\sigma 2p\sigma)$ transition of the F_2 center was also calculated for KCl. By comparing the result for the F_2 center with previous calculations,[2,3] we have found that the method used in

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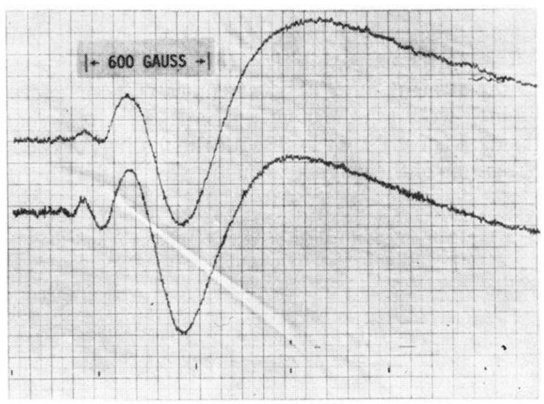

FIG. 1. Recording of signal amplitude versus increasing magnetic field. The upper curve is for 4.2°K and the lower curve for 1.2°K. The frequency is 30 Mc/sec. $\mathbf{H}\perp\mathbf{q}$ and $\mathbf{q}\parallel[100]$.