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Attenuation and velocity of sound near the open-orbit resonances in copper

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Simultaneous velocity and attenuation measurements are reported for the [111] and [010] open orbits in copper using 52-MHz compressional waves propagating in the [$\bar{1}$01] direction. The two-phase continuous-wave resonance technique applied to a sharp open-orbit resonance is shown to be useful for measuring one of the physical constants e/ch, the lattice constant, or the velocity of sound if the other two are well known. The behavior of the dispersion near the [111] open-orbit resonance seems to imply that the effective force on the lattice is different for electrons drifting parallel and antiparallel to $\vec{q}$.

I. INTRODUCTION

It has been shown[1] that conduction electrons moving on open trajectories normal to an applied magnetic field can absorb energy resonantly from a sound field, giving rise to resonances in the attenuation. Such open-orbit resonances occur when the period of the real-space orbit projected in the direction of sound propagation equals an integral multiple of the sound wavelength. The field B_n at which the nth-order resonance occurs and the relative width of the resonance are given by

$$B_n = (\hbar f c/nev_s)K \tag{1}$$

and

$$(\Delta B)_n/B_n = 2/ql \quad , \tag{2}$$

respectively. Here K is the period of the open orbit in $\vec{k}$ space, f is the ultrasonic frequency, v_s is the velocity of sound, q is the sound wave number, l is the electron mean free path, and the other symbols have their conventional meanings.

Open-orbit resonances have been found in copper,[2,3] cadmium,[4] and a number of other metals[4,5]; the studies have proved useful in understanding many of the transport properties of metals. For example, Brillouin-zone spacings and electron mean free paths can be deduced from the resonance fields and the relative widths of the resonance peaks, respectively. Such studies have yielded results in close agreement with values determined from other methods.

Conduction electrons can also modify the ultrasonic velocity. Such an effect has been predicted and verified experimentally in a variety of metals.[6–9] As can be seen from Eq. (1), a change in the sound velocity would result in a relative error of $\Delta K/K_0 = \Delta v/v_{s0}$ in the determination of K were the velocity change not taken into account, where v_{s0} is the velocity of sound at zero magnetic field and $K_0 = (nB_n e/c\hbar f)v_{s0}$. It is conceivable that the correction in the velocity can be very important if $\Delta v/v_{s0}$ is large. Kamm[3] has found that neither correct Fermi-surface dimensions nor correct Brillouin-zone spacings of copper could be derived from his measurements unless the velocity of sound along the different symmetry axes was properly adjusted. This indicates that simultaneous velocity and attenuation measurements are essential to the better understanding of the electronic properties of metals.

The purpose of this paper is to report the experimental determination of the attenuation and velocity changes for the [111]- and [010]-directed open orbits in copper, and to demonstrate that, for a sharp resonance like the fundamental of the [111] open orbits, one may find other applications of the ultrasonic technique.

II. EXPERIMENTAL TECHNIQUES

A. General

The measurements were made at a temperature of 4.2 °K using a two-phase continuous-wave resonance technique.[10] Two ultrapure single crystals of copper, designated as Cu-I and Cu-II, cut from a boule[11] having a residual resistivity ratio of 35 000 were used. The longitudinal sound wave was propagated along the [$\bar{1}$01] axis of the crystal and was perpendicular to the applied magnetic field which was provided by a 15-in electromagnet. Cu-I and Cu-II had lengths of 0.323 and 1.01 cm, respectively.

B. Two-phase continuous-wave resonance technique

There are many ways[12] to measure a small change in ultrasonic velocity; one which is widely employed is the continous-wave (cw) resonance technique.[9] The change in the velocity is reflected in the frequency change required to bring the composite oscillator of transducers, bonds, and sample back to mechanical resonance. If one also monitors the amplitude of the mechanical resonance peak, then the attenuation can also be measured. The two-phase cw resonance technique employed in the experiment was designed on this

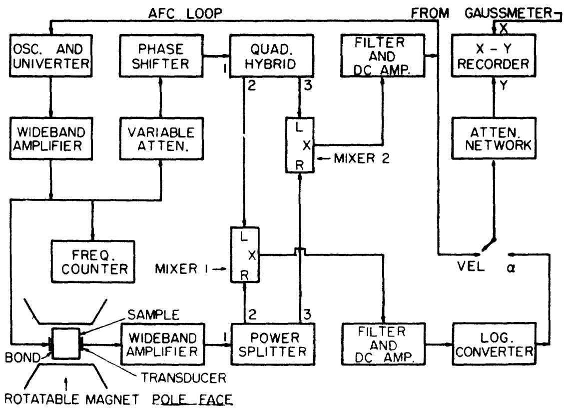

FIG. 1. Schematic diagram of experimental apparatus for ultrasonic attenuation and velocity measurements.

same principle.

Figure 1 shows the block diagram of the experimental apparatus. The heart of the system is the two balanced mixers. The sound wave undergoing a mechanical resonance in the composite oscillator is picked up and reconverted into an electrical signal by the receiving transducer. The recovered signal, after being amplified, is divided by a power splitter into two signals of equal amplitude with phase difference less than 1°. The signals are then fed to the R ports of the balanced mixers. The reference signals at the L ports of the mixers are furnished by a quadrature hybrid. They have the same amplitude but differ in phase by 90°. Because of this phase relation between the reference signals, mixer 1 serves as an in-phase detector while mixer 2 serves as a quadrature detector.

The output of each mixer is first filtered to obtain the dc component and then amplified. The dc output of the in-phase detector, which is proportional to the amplitude of the signal at a mechanical resonance, is logarithmically converted before recording to represent the attenuation. The output of the quadrature detector, which is proportional to the cosine of the relative phase between the signals at the L and R ports of the detector, is fed back to the oscillator frequency-control input to regulate the oscillator frequency so as to maintain the composite-oscillator mechanically resonant when the velocity of sound changes. This frequency change, as in the cw resonance technique, is linearly proportional to the velocity change and, furthermore, it is also linearly proportional to the external dc control voltage. Hence an analog recording of the frequency changes was made by recording the frequency control voltage during the experiment.

The measurements were made in the following fashion: First the output of the quadrature detector was manually adjusted to a null and that of the in-phase detector to a maximum at a mechanical resonance frequency and zero magnetic field using the phase shifter. The field was then slowly swept over the desired range of values and the output of the logarithmic converter and the frequency control voltage were recorded as functions of the applied field. Calibrations of both attenuation and frequency changes were made for each field sweep. The attenuation was calibrated against a built-in voltage offset of the logarithmic converter and the calibration for the frequency changes was made by means of a frequency counter.

C. Electrical coupling and mechanical resonance frequencies

It must be pointed out that the two-phase cw resonance technique, like all cw techniques, is quite susceptible to stray electrically coupled signals which are not associated with the ultrasonic path. The effect of the electrical coupling on velocity measurements has been studied by Yee and Gavenda.[9] A similar analysis[10] made of this two-phase system reveals that the electrical coupling produces both apparent attenuation and velocity changes which are added to or subtracted from the true changes in the fashion indicated by Yee and Gavenda when the electrical coupling is small and the attenuation not too large. This favorable behavior of the coupling effect makes it possible to improve the results by using an averaging method.

Because the electrical coupling plays such a decisive role in the experiments, we must reduce it to a level acceptable within the desired range of attenuation changes. To do this, we have constructed a sample holder that reduces the coupling to 1% or less of the first echo, and used a sample of 0.323-cm length (Cu-I) to reduce the total attenuation. Under favorable conditions the coupling effect is almost nonobservable and the measured velocity changes can be considered as true changes.

Whether the electrical coupling was acceptable or not was judged first by the mechanical resonance spectrum and then by comparisons of data taken with different mechanical resonance frequencies near the fundamental (52 MHz) of the transducers. The composite oscillator is mechanically resonant when there is an integral multiple of sound half-wavelengths inside the oscillator. This corresponds to a condition for which the magnitude of the voltage at the receiving transducer is a maximum when the electrical coupling is zero. Consequently, a series of maxima and minima is expected when the frequency is swept. The maxima corresponding to mechanical resonance peaks, according to our analysis, should be evenly separated and have the same height. However, when the coupling is nonzero the locations as well as the

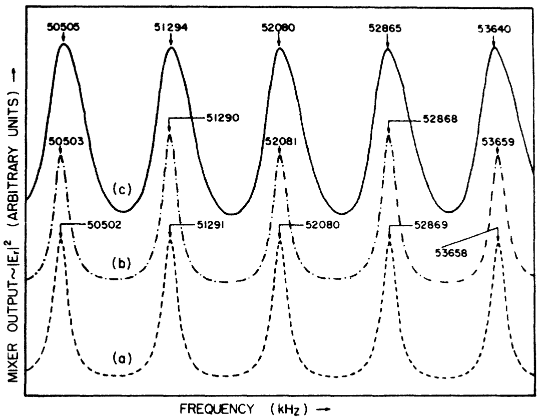

FIG. 2. Computed and experimentally determined mechanical resonance spectra for Cu-I. Curves (a) and (b) are the computed curves obtained under the conditions: (a) $E_c = 0$ and $\phi = 0°$; and (b) $E_c = E_0/10$ and $\phi = 150°$. Curve (c) is the experimental curve obtained at $T = 4.2\,°K$ and $B_0 = 0$ with $\vec{q} \parallel [\bar{1}01]$.

heights of the peaks will be shifted.

Nonparallel sample faces would tend to broaden the mechanical resonances. Small departures from parallelism would have no appreciable effect on the measured changes in velocity and attenuation, however, since our use of the resonances is primarily to enhance the signals and give us an indication of the relative amount of electrically coupled signal present.

Curve (c) in Fig. 2 is the output of a square-law detector versus the frequency made with $\vec{q} \parallel [\bar{1}01]$ at $T = 4.2\,°K$ and zero field. Curves (a) and (b) were computed from an equation given by Yee and Gavenda[9] under the conditions (a) $E_c = 0$ and $\phi = 0^0$, and (b) $E_c = E_0/10$ and $\phi = 150°$, where E_0 is the amplitude of the voltage at the transmitting transducer and E_c and ϕ are the amplitude and relative phase of the electrically coupled signal. In the calculations the zero-field attenuation $\alpha_0 L$ was assumed to be 2.295 dB, a value close to the measured value for Cu-I, and the velocity of sound at zero field was taken to be 5.076×10^5 cm/sec.[13] As compared to curve (a), we considered curve (c) to be satisfactory. This was fully justified by the data taken with the 51 295-, 52 080-, and 52 865-kHz peaks which are presented in Sec. III.

When the electrical coupling is small, the true attenuation change $\Delta\alpha$ can be calculated from the measured change $\Delta\alpha_M$ and the zero-field attenuation α_0:

$$\Delta\alpha = \Delta\alpha_M - (20/L)\log_{10}[(1 - 10^{-(\alpha_0+\Delta\alpha)L/10})/(1 - 10^{-\alpha_0 L/10})] \quad . \tag{3}$$

All attenuations are expressed in dB/cm. One can see that the standing waves enhance the measured values of attenuation by an amount which depends on the total attenuation $(\alpha_0 + \Delta\alpha)L$. Near the open-orbit resonances which we discuss below $\Delta\alpha_M$ varies from about 30 to 40 dB/cm. Iterative solutions of Eq. (3) show that these measured values correspond to true attenuations of 13.0 and 20.0 dB/cm, respectively, for $\alpha_0 L = 2.3$ dB. Thus, what appears to be a 10-dB/cm change is actually a 7-dB/cm change. The attenuation curves which we present below have not been corrected in this fashion since our primary concern is to compare the general features, rather than the absolute magnitudes, of the attenuation and velocity curves.

III. EXPERIMENTAL RESULTS

To ensure that the electrical coupling had been satisfactorily reduced, we measured both the attenuation and velocity changes at three successive mechanical resonance frequencies centered around 52 MHz. Typical results near an open-orbit resonance are shown in Fig. 3. The solid curves are the attenuation and the dashed ones are the fractional velocity shifts. For clarity these curves have been shifted up or down arbitrarily. As can be seen, the magnitudes of the corresponding curves for two adjacent peaks differ by no more than 3%, with larger values for the higher frequency. This small discrepancy is easily explained by the 1% frequency difference between two

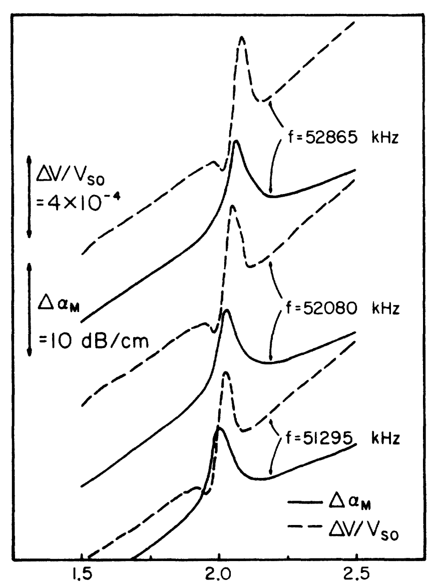

FIG. 3. Relative attenuation and velocity changes of the fundamental resonances of the [111]-directed open orbit in Cu-I measured with three successive mechanical resonance frequencies as indicated, where $\vec{q} \parallel [\bar{1}01]$ and $\vec{B}_0 \parallel [121]$. For clarity the curves have been appropriately shifted up or down.

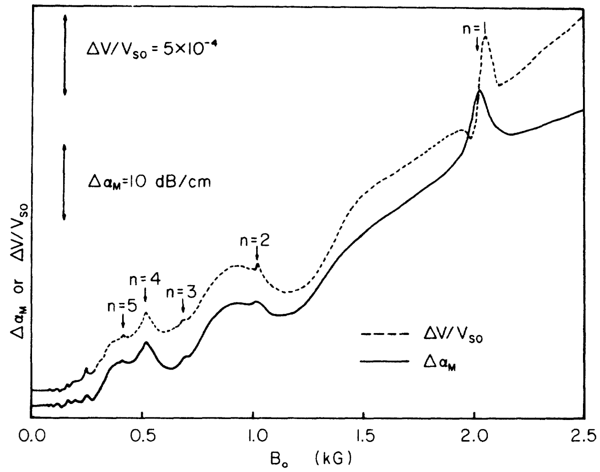

FIG. 4. Relative attenuation and velocity changes in Cu-I measured with $f = 52080$ kHz, $\vec{q} \parallel [\bar{1}01]$, and $\vec{B}_0 \parallel [121]$. Note that the [111]-directed open orbit peaks can be seen up to $n = 5$.

adjacent peaks. In addition we notice that the locations of fundamental resonances of the [111] open-orbit resonances shift toward higher fields for higher frequencies which is fully compatible with Eq. (1). This excellent agreement led us to believe that the electrical coupling was no longer effective in the experiment. The velocity and attenuation of the same open orbits measured over a greater range of field with $f = 52080$ kHz are presented in Fig. 4. Notice that the velocity fluctuates rapidly, especially for the fundamental resonance, in the vicinity of resonances and that the harmonic peaks for both the velocity and attenuation can be identified up to $n = 5$.

Figure 5 shows the splitting of the attenuation and velocity peaks for the [010] open orbits with rotation of the magnetic field in the [$\bar{1}$01] plane. The angle $\Delta\theta_B$ is the angle that the field makes with [101]. Unlike the [111] open orbits, there is no remarkable distinction, other than dislocations of peaks, between the attenuation and velocity. However, the difference becomes clearer when $\Delta\theta_B$ becomes larger. Apparently the velocity changes are much more sensitive to the field direction. For example, a $0.1°$ rotation is not enough for the attenuation peak to split, but it is large enough for the velocity. This seems to be consistent with the velocity changes for the [111] open orbits because the dip on the high-field side of the first peak coinciding with the dip on the low-field side of the second can effectively lower the portion between. Failure to observe a prominent velocity fluctuation could be attributed to the fact that the resonance is much weaker and broader. We have not observed any splitting in either the velocity or the attenuation peaks, as reported[14] for the [111] open orbits.

To be sure that the line shapes of the velocity for the [111] open orbits are genuine and not due to any coupling effect, we modified the two-phase system by inserting a gate and a high-speed switching device between the signal generator and the amplifier to obtain two coherent square pulses, one fixed and one which can be delayed. The fixed pulse is used to excite the sound wave and the other is split to provide the reference signal for the mixers. This gated cw method allows us to eliminate the electrical-coupling error completely by sliding the reference signal to a particular echo so that the detected dc signal from either mixer is made free of electrically coupled signal. It can be shown[10] that the gated cw method measures both true attenuation and velocity changes at frequencies other than the mechanical resonance frequency when the loop gain of the frequency-control feedback and the repetition rate and width of the pulse are appropriately adjusted. However, as with all pulse methods, the sample must be long enough to allow the separation of successive pulses. This prevents its use under conditions when the attenuation is very large.

Shown in Fig. 6 are representative results for Cu-II obtained from the gated cw method with two frequencies near the mechanical resonances. The line shapes of the attenuation as well as of the velocity changes are in complete agreement with the corresponding curves obtained from the resonance method though there is a slight disagreement in magnitudes. We speculate that the discrepancy could be due to the fact that the resistivity ratios for Cu-I and Cu-II may not be equal or that one

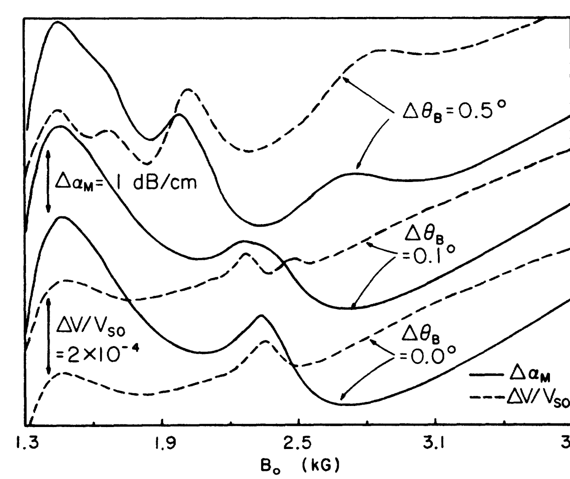

FIG. 5. Splitting of the [010]-directed open-orbit peaks in the relative attenuation and velocity changes in Cu-I with rotation of $\vec{B}_0$ in the ($\bar{1}$01) plane relative to the [101] direction with $\vec{q} \parallel [\bar{1}01]$ and $f = 52080$ kHz. The curves have been shifted arbitrarily.

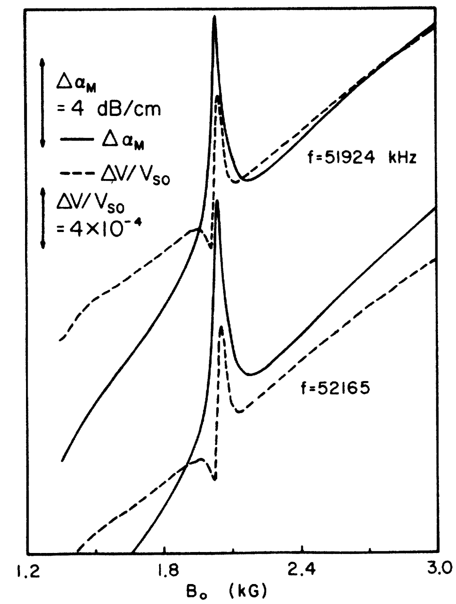

FIG. 6. Fundamentals of the [111]-directed open-orbit resonances in the relative attenuation and velocity changes in Cu-II measured with the gated cw method at two different frequencies with $\vec{q} \parallel [\bar{1}01]$ and $\vec{B}_0 \parallel [121]$. The curves have been shifted arbitrarily.

run was better aligned than the other as far as the perpendicularity of $\vec{q}$ to $\vec{B}_0$ is concerned.

IV. INTERPRETATION OF RESULTS

Mertsching[7] has shown that the attenuation and fractional velocity changes for a longitudinal wave propagating through a pure crystal along an axis of high symmetry are proportional to the real and imaginary parts, respectively, of a function K_L given by

$$K_L = \sum_{n=-\infty} \int \frac{F_n(k_z, \omega_c, \vec{q}, \vec{D})\, dk_z}{\langle \tau^{-1}\rangle + i(\vec{q} \cdot \langle \vec{v}\rangle - \omega - n\omega_c)} \quad . \qquad (4)$$

The function $F_n(k_z, \omega_c, \vec{q}, \vec{D})$, which accounts for the magnetoacoustic oscillations, involves an integration around the orbit in each k_z plane perpendicular to the magnetic field. The resonance effects come from the denominator and occur whenever it has a minimum. This resonance condition leads to Eq. (1) when the frequency ω is neglected.

The experimental results indicate that the open-orbit velocity changes, particularly near the fundamental resonance for the [111] orbits, vary to some extent like the imaginary part of a resonance function. In order to compare the theory with the experiment one must calculate K_L according to Eq. (4). However, such a calculation is not possible for a real metal because the function F_n contains deformation forces which are not calculable at present. Assuming that F_n is a slowly varying function, one can reasonably expect the line shapes of the resonances to be predominantly determined by the denominator of Eq. (4). The fact that the fundamentals of the [111] and [010] open orbits occur in a region where the background changes are monotonic seems to justify this assumption. For simplicity we have assumed F_n to be a constant and plotted the real and imaginary parts of $[1+i \times(\vec{q}\cdot\langle\vec{v}\rangle - \omega - \omega_c)\tau]^{-1}$ for a given k_z as functions of $\omega_c\tau$, as shown in Fig. 7, where $ql \equiv |\vec{q}\cdot\langle\vec{v}\rangle|\tau = 100$ and $\omega\tau = 0.333$. The electrons that drift antiparallel to $\vec{q}$ are taken into account by adding a term containing $[1 - i(\vec{q}\cdot\langle\vec{v}\rangle + \omega - \omega_c)\tau]^{-1}$. When a smoothly increasing background is added and the effect of the electrons drifting opposite to $\vec{q}$ is weighted less, the line shape of the imaginary part can be made to resemble that of the velocity curve for the [111] orbits as determined by experiment. This would mean that the electrons with drift velocity parallel to $\vec{q}$ can affect the elastic constant more effectively than those that drift antiparallel.

Equation (1) can be rewritten as $e/ch = f/B_1 v_s d$, where $d = 2\pi/K$ is the real-space equivalent of the Brillouin-zone spacing in the open-orbit direction and B_1 is the field strength for the fundamental resonance. For a sharp resonance, such as the fundamental for the [111] open orbits, it is possible to measure the resonance field B_1 as well as the appropriate mechanical resonance frequency f rather accurately. Thus, an accurate determination for one of the three physical quantities e/ch, d, or v_s can be made from open-orbit velocity and attenuation experiments when the other two quantities are known. As an example, calculations using Fig. 3 yield $e/ch = (2.415 \pm 0.006)\times 10^6$ esu/erg cm, $a = (3.599 \pm 0.008)\times 10^{-8}$ cm, and $v_s = (5.095 \pm 0.010) \times 10^5$ cm/sec, where a is the lattice constant for

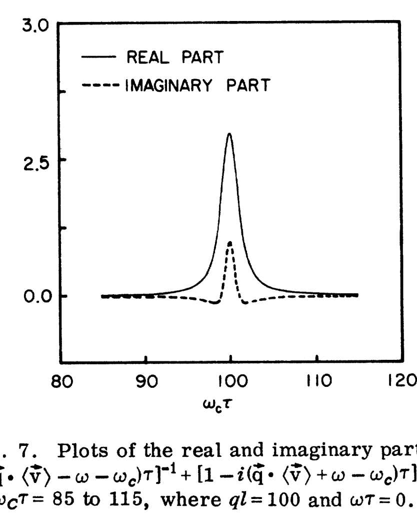

FIG. 7. Plots of the real and imaginary parts of $[1+i(\vec{q}\cdot\langle\vec{v}\rangle - \omega - \omega_c)\tau]^{-1} + [1 - i(\vec{q}\cdot\langle\vec{v}\rangle + \omega - \omega_c)\tau]^{-1}$ vs $\omega_c\tau$ from $\omega_c\tau = 85$ to 115, where $ql = 100$ and $\omega\tau = 0.333$. Note that the resonances for the real and imaginary parts occur near $\omega_c\tau = 100$.

copper. These results agree with values obtained from other methods[3,13,15] to within the experimental uncertainty, which is about 0.2%. The primary source of error in the present experiments was nonlinearity in the amplifier and detector used with the rotating-coil Gaussmeter. Magnetic field values were accurate to only about ± 5G. The precision can be considerably improved by more careful measurements of the field values.

V. SUMMARY AND CONCLUSIONS

The attenuation and velocity of sound in pure copper for the [111] and [010] open-orbit resonances have been investigated. It is found that the velocity of sound changes more rapidly than the attenuation near sharp resonances. The velocity for the [010] open orbit exhibits a more sensitive Doppler splitting. Further studies of the two open orbits in copper with shear waves may shed more light on the open-orbit velocity changes because the resonances are sharper and occur at field values where the background attenuation is relatively small and constant.[14]

Simultaneous measurements of attenuation and velocity changes are important because they give more accurate information about open-orbit electrons than measurements of attenuation alone. For example, if one knows the lattice constant and uses the accepted values for e/ch, the zero-field sound velocity can be determined with high accuracy.

Finally, the experimental results seem to indicate that the electrons with drift velocity parallel to $\vec{q}$ affect the elastic constant more than those that drift opposite to $\vec{q}$ for the [111] open orbits. In fact, very little is known about the deformation-potential force which is purported to be important in magnetoacoustic effects. One might hope to use measurements of the sort reported here to deduce the real and imaginary components of the effective force on open-orbit electrons which leads to the observed attenuation and velocity changes.

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