pling. Although the intrinsic efficiency of such processes is much smaller than that due to "exchange pumping," the application of a strong magnetic field can bring the frequencies into a tractable range.

In principle, more highly involved cases of exchange pumping are possible with magnetic sublattices of higher multiplicity. For example, since it is known that the pump frequency in a four-frequency parametric amplifier need not be the highest, it is conceivable that in such cases of higher multiplicity, exchange modes might be excited with sources of lower frequency.

An analysis of magnetoelastic coupling between the magnon spectra discussed in this Letter and the acoustic and optical branches of the phonon spectra has been carried out by the present author.<sup>12</sup> One finds a great deal of similarity between the exchange branch of antiferromagnetic magnetoelastic waves and those associated with the microwave branch except that a <u>different</u> combination of magnetoelastic energy splitting of both branches should allow (in principle) decomposition of these coefficients into their sublattice components. The parametric excitation of magnetoelastic modes<sup>12</sup> may provide a means of measuring

## some of these splittings.

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## OSCILLATIONS IN THE VELOCITY OF SOUND IN GOLD AT HIGH MAGNETIC FIELDS

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This Letter reports the observation of de Haas – van Alphen type oscillations in the velocity of sound in gold. Since such oscillations are observed in a variety of physical properties, it is not surprising that they can also be seen in the velocity of sound.<sup>1</sup> However, their magnitude and the conditions under which they can be observed in a noble metal is important because of the current interest in the effect of magnetic fields on sound waves in ordinary metals.<sup>2-4</sup>

It has been well established<sup>4,5</sup> that when the sound wavelength is much greater than the electron mean free path  $(\lambda \gg l)$  and when the cyclotron frequency is much less than the collision frequency  $(\omega_c \tau \ll 1)$ , the velocity of sound in-

creases in proportion to the square of the applied magnetic field for longitudinal sound waves propagating at right angles to the magnetic field. In order to determine if the same relation holds in the highest attainable fields and under conditions in which  $\omega_c \tau > 1$ , a series of exploratory experiments were performed on a variety of metal single crystals in magnetic fields up to 100 000 G. The magnetic fields were provided by the  $2\frac{1}{2}$ -in. inside diameter Bitter-type solenoid at the U. S. Naval Research Laboratory. The changes in the velocity of sound produced by these fields were measured using the ultrasonic "sing around" system developed by Forgacs.<sup>6</sup> This system can detect a change of one part in  $10^7$  in



FIG. 1. Relative increase in the velocity of a longitudinal sound wave in gold produced by the application of a magnetic field in various crystallographic directions at right angles to the [110] propagation direction. The data for each field direction have been arbitrarily displaced vertically to avoid confusion of the data points.

the transit time of a ten-megacycle/second ultrasonic pulse traveling through an approximately 2-cm long sample.

Figure 1 shows the results of measurements on a single crystal of gold at 4.2°K in which the sound was propagated down a [110] crystallographic axis and the magnetic field was placed in a variety of directions in the plane perpendicular to this [110] direction. The data for each direction are arbitrarily displaced upward to avoid confusion of data points. The abscissa is the square of the magnetic field and the slope of the straight lines drawn through the data points were determined from the theory<sup>4</sup> (slope =  $1/8\pi\rho v_I^2$  where  $\rho$  is the density of gold and  $v_l$  the longitudinal wave velocity). It can be seen that for all directions studied except the one at 67° to the [001] direction, the data fall on the straight line predicted by the theory. Detailed examination of what appears as a terrible scatter of the data in the  $67^{\circ}$ direction showed that the velocity of sound was actually an oscillatory function of magnetic field with a very short period. Figure 2 shows the

FIG. 2. Oscillatory variation of the velocity of a longitudinal sound wave in gold at  $4.2^{\circ}$ K with the magnetic field The magnetic field lies in a plane normal to the [110] crystal axis and at 67° to the [001] direction. The sound-wave propagation direction is along the [110] direction.



Table I. Comparison of the separation in magnetic field of successive oscillations in the velocity of sound with the period of magnetic susceptibility oscillations observed by Shoenberg.

Magnetic	Sound velocity	Magnetic susceptibility
field	$\Delta H$	$\Delta H$
(kG)	(kG)	(kG)
99.5 88.5 77.5	$0.59 \\ 0.42 \\ 0.37$	0.61 0.48 0.37

variation of the sound velocity with magnetic field over a two-kilogauss interval near 100 kG [Fig. 2(a)], 90 kG [Fig. 2(b)], and 80 kG [Fig. 2(c)]. It can be seen that the period of oscillation and the amplitude both decrease as the magnetic field is lowered.

The observed behavior is in qualitative agreement with the de Haas-van Alphen effect. For quantitative comparison, the field separation of two minima in the sound velocity,  $\Delta H$ , should be compared with the field separation of successive oscillations in the magnetic susceptibility as observed by Shoenberg<sup>7</sup> in gold with the magnetic field in the same orientation. Table I presents this comparison and shows the quantitative agreement obtained. The magnetic field dependence of the amplitude of the oscillations in the velocity of sound can be described by an exponential function of the form  $\exp(-5.1 \times 10^5/H)$ , where H is measured in gauss. Such an exponential amplitude dependence is to be expected for the de Haasvan Alphen effect since the factor  $\exp(-2\pi^2 k T/\hbar\omega_c)$ plays an important role. As a matter of fact, the value of  $2\pi^2 k T/\hbar\omega_c$  is  $6.2 \times 10^5/H$  (for  $T = 4.2^{\circ}$ K and  $m/m_0 = 1$ ) which is in excellent agreement with the measurements when cognizance is taken of the fact that the temperature T and the effective mass m are not particularly well-defined numbers in this case.7

In view of the excellent agreement between the period and amplitude of the oscillations observed in the velocity of sound with the de Haas-van Alphen oscillations observed by Shoenberg,<sup>7</sup> there is little doubt that the electrons responsible for this effect are those in orbits around the necks of the Fermi surface in the [111] direction. Due to an error in orienting the sample in the magnetic field, no data were obtained with the field exactly along the (111) direction. Oscillations in the sound velocity for quite different field directions should also be observable, but judging from the susceptibility measurements they can be expected to have much shorter periods and

smaller amplitudes. Thus they would be difficult to observe and do not appear in the sparse data taken for the lower curves of Fig. 1.

The electron mean free path in this particular gold sample at 4.2°K may be estimated from a measurement of the change in ultrasonic attenuation accompanying the application of the magnetic field. Using the formulas of Steinberg,<sup>8</sup> the mean free path was found to be  $2 \times 10^{-4}$  cm which yields ql = 0.04 where q is the ten-megacycle/second phonon wave number,  $2\pi/\lambda$ . Using this result and the free-electron Fermi velocity of the electrons in gold  $(1.4 \times 10^8 \text{ cm/sec})$ , the mean time between collisions,  $\tau$ , becomes  $2 \times 10^{-12}$  sec, which in turn implies that  $\omega_c \tau = 1$  at about 30 kG.

Much more extensive data were taken on a copper crystal of considerably higher purity (ql=0.4), but no oscillations in the sound velocity greater than 4 parts per million at 100 kG were observed even when the magnetic field was oriented parallel to the (111) direction. This negative result is consistent with the observations of Shoenberg<sup>7</sup> who found that the amplitude of the susceptibility oscillations in copper were considerably smaller than in gold.

In connection with the general problem of modification of the sound velocity with a magnetic field, the present measurements at very high fields showed no deviation from the simple theory.<sup>4</sup> Crystals of aluminum, copper, gold, and tantalum were studied. In the case of the high-purity copper, an attempt was made to verify the previous result<sup>5</sup> that the velocity of sound is a linear function of magnetic field for field orientations in which the electrons describe "open orbits." This linear dependence was not observed. A possible reason for this is that the experimental arrangement did not allow the crystal to be oriented in the magnet with the precision necessary to observe open-orbit effects.

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