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QUANTUM OSCILLATIONS OF THE VELOCITY OF SOUND IN ZINC*

by

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ABSTRACT

Large amplitude quantum oscillations of the longitudinal sound velocity, $[\Delta V/V] \sim 10^{-3}$, were observed in single crystal zinc. The effective masses associated with the "cigar" and "butterfly" were determined from the temperature dependence of the amplitudes of the oscillations in the region $1.5 \leq T \leq 4.2$ K . The experimental data are compared with the results of theoretical treatments.

RESUMO

Oscilações quânticas de grande amplitude, $[\Delta V/V] \sim 10^{-3}$, foram observadas na velocidade de ondas sonoras de compressão num monocristal de zinco. As massas efetivas associadas com as regiões da superfície de Fermi conhecidas pelos nomes "charuto" e "borboleta", foram determinadas através da dependência com a temperatura das amplitudes das oscilações. Os dados experimentais são comparados com os resultados de tratamentos teóricos.

QUANTUM OSCILLATIONS OF THE VELOCITY OF SOUND IN ZINC

WE REPORT the first observation of quantum oscillations of the longitudinal sound velocity in zinc.

All measurements were performed on a single crystal of 99.999% purity, obtained from Metals Research (Crystals) Ltd. The experiments were performed with both the applied magnetic field, $\vec{H}$, and the propagation vector, $\vec{q}$, of the longitudinal sound wave along the hexagonal axis, [0001]. Magnetic fields up to 70kOe were provided by a superconducting solenoid. Changes in the sound velocity due to the magnetic field were measured using the technique described in (1). Fractional changes of order 5×10^{-5} in the sound velocity, V, could be resolved. Attenuation measurements were also made using conventional pulse-echo techniques.

In Fig. 1, the magnetic field dependence of the velocity oscillations is shown for a series of seven temperatures, for 10 MHz longitudinal sound waves. In order to determine the de Haas-van Alphen (dHvA) frequencies and the corresponding amplitudes, a Fourier analysis of the data was carried out along the lines of Ferreira and Quadros.(2) For the two higher frequencies (see Table 1), we estimate the amplitudes obtained in this manner to be correct to within 5%. A Fourier analysis of the corresponding attenuation data showed them to be a superposition of the same three dHvA frequencies observed in the velocity, but with amplitudes proportionately quite different from those of the velocity curves.

In Table 1, the dHvA frequencies, labeled according to the notation of Fletcher et al.,[3] are presented. The lowest frequency, f_R, corresponds to a magnetic-breakdown orbit involving the diagonal arms. The frequencies f_C and f_D were assigned by Higgens et al.[4] to butterfly and cigar orbits on the nearly free-electron Fermi surface of Harrison.[5] Although the calculations of Stark and Falicov[6] indicate that there are no carriers at the L point, Fletcher et al. were unable to interpret their extensive data without recourse to the butterflies and cigars, located at the L point.

Theoretical treatments of quantum oscillations in the sound velocity predict that the amplitude of the oscillation varies with T as in the dHvA effect[7-9]. In Fig. 2, a plot of $\ell n \{\frac{A}{T} [1 - \exp(\frac{-4\pi^2 kT}{\hbar\omega_c})]\}$ vs. T is shown for the amplitude A associated with the cigar orbit. The solid line passing through the experimental points represents a least squares fit to the data, and, from its slope, the effective mass is obtained. Since the small correction term $\exp(\frac{-4\pi^2 kT}{\hbar\omega_c})$ contains the effective mass, it was necessary to obtain the final values of the effective masses through a series of iterations. From the plot of Fig. 2, and others of the same nature, the effective masses of Table 1 were obtained. The accuracy of the masses m_C^* and m_D^* is estimated to be 10%, but the uncertainty in m_R^* is $\sim$ 20% because the experimental amplitudes associated with f_R were smaller and were determined with less accuracy. Also shown in Table 1 are masses obtained from a similar analysis of our attenuation data. Our masses m_C^* and m_D^* are in rough agreement with those obtained by Venttsel' [10], 7^o from the [0001] axis.

The existing theoretical treatments of quantum oscillations in the sound velocity can be roughly divided into two groups. In the first group [7,8,11] the detailed interaction between the sound wave and the conduction electrons is considered. These treatments neglect possible complications due to multiple bands or non-parabolic dispersion laws, which exist in the case of the multiband Fermi surface of zinc. We have not attempted a comparison of the experimental amplitudes with the results of these treatments.

Another line of approach, followed by Rodriguez [8] and by Testardi and Condon, [9] involves the use of a thermodynamic argument to obtain the electronic contribution to the elastic constants. The treatment of Rodriguez, which is appropriate for density of states oscillations and which, therefore, should be valid for $q\ell \lesssim 1$, has been used to calculate the order of magnitude of the velocity oscillations in Ga [12], Al [13], and Cu [13]. (In the case of Ga, experimental conditions were such that $q\ell \gg 1$, and so this treatment may not be applicable.) Since we estimate $q\ell \sim 0.1$ in our experiments, we have applied Rodriguez's theory to zinc, despite the fact that it may not be strictly correct [12,13].

Rodriguez assumes the longitudinal sound velocity to be determined by the bulk modulus and that the latter quantity is given by the second derivative of the free energy with respect to volume. Using this approach, and the Lifshitz-Kosevitch expression for the free energy of the electrons in a magnetic field [14], one obtains an expression for $\Delta V/V$. See Ref. 8 Eq. (29), Ref. 12 Eq. (1), and Ref. 13 Eq. (3). (The last reference includes the effects of electron scattering throught the use of a Dingle temperature T_D.) To obtain these expressions of $\Delta V/V$, the derivative of the extremal Fermi surface area with respect to volume, $\frac{\partial S}{\partial v}$, was determined using a free-electron model. However, one can avoid this approximation using the experimental value for $\frac{\partial S}{\partial v}$, obtained from the dependence of S on hydrostatic pressure [15]. Finally, since the acoustic velocities are determined by the elastic constants of the material and the elastic constants arise from the strain derivatives of the free energy, one expect to calculate $\Delta V/V$ more accurately, using the experimental dependence of S on strain [16].

In Table 1, the peak-to-peak amplitudes obtained from a Fourier analysis of the lowest curve of Fig. 1 ($T = 2.23$ K) are compared with the amplitudes calculated from Eq. (3) of Ref. 13, using the free-electron value $E_F = 21.8$ eV, [6] the masses of Table 1, and the mean value of the magnetic field for the data of Fig. 1, $H = 69.2$ kOe. The effects of spin and electron scattering were neglected and the geometrical factor

$[\partial^2 S/\partial k_z^2]$ was taken equal to 2π. With these latter approximations, we have also calculated the amplitude of the R oscillations, using the experimental dependence of S on hydrostatic pressure [15], and the amplitude of the C oscillations, using the experimental dependence of S on strain [16]. We find order of magnitude agreement, for the frequencies C and D, between the experimental amplitudes and those for which $\frac{\partial S}{\partial v}$ has been calculated in the free-electron approximation. There is, however, much better agreement between the experimental and calculated amplitudes in the cases where this approximation can be avoided. We note that including the effects of spin and electron scattering would reduce the calculated amplitudes. However, we were unable to determine T_D from our data and g is known only for the needles in zinc. These, and other real metal effects, can only be taken into account properly through a treatment similar to that of Testardi and Condon [9].

An important consequence of Eq. (3) of Ref. 13 is that the sound velocity oscillations should be independent of the ultrasonic frequency. However, some of our later experiments show that the experimental amplitudes are dependent upon frequency. This apparent frequency dependence is being investigated in greater detail.

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Table 1.

Results of the measurements of quantum oscillations in sound velocity in zinc for $\vec{q} \| \vec{H} \| [0001]$

Identification [a]	R	C	D
dHvA frequency (10^7 Oe.)			
present work	0.50	1.15	1.72
Fletcher _et al._ [a]	0.50	1.12	1.71
Effective masses (m^*/m_o)			
present work (velocity)	0.20	0.32	0.51
present work (attenuation)	0.18	0.34	0.49
susceptibility results [b]	—	0.37	0.46
Amplitudes of velocity oscillations, $\Delta V/V$ (10^{-3})			
experiment	0.2	1.53	1.04
calculated from Eq. (3) of Ref. 13	14.0	20.2	19.6
calculated from hydrostatic pressure data of Ref. 15	0.16	—	—
calculated from strain dependence of S , Ref. 16	—	0.61	—

[a] FLETCHER R., MACKINNON L. and WALLACE W.D., see Ref. 3.

[b] VENTTSEL' V.A., 7^o from the [0001] axis, see Ref. 10.

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FIGURE CAPTIONS

Fig. 1. Magnetic field dependence of the fractional change in velocity as a function of temperature.

Fig.2. Temperature dependence of the amplitude A associ ated which the cigar orbits. The amplitude A was obtained from a Fourier analysis of the curves of Fig. 1 .

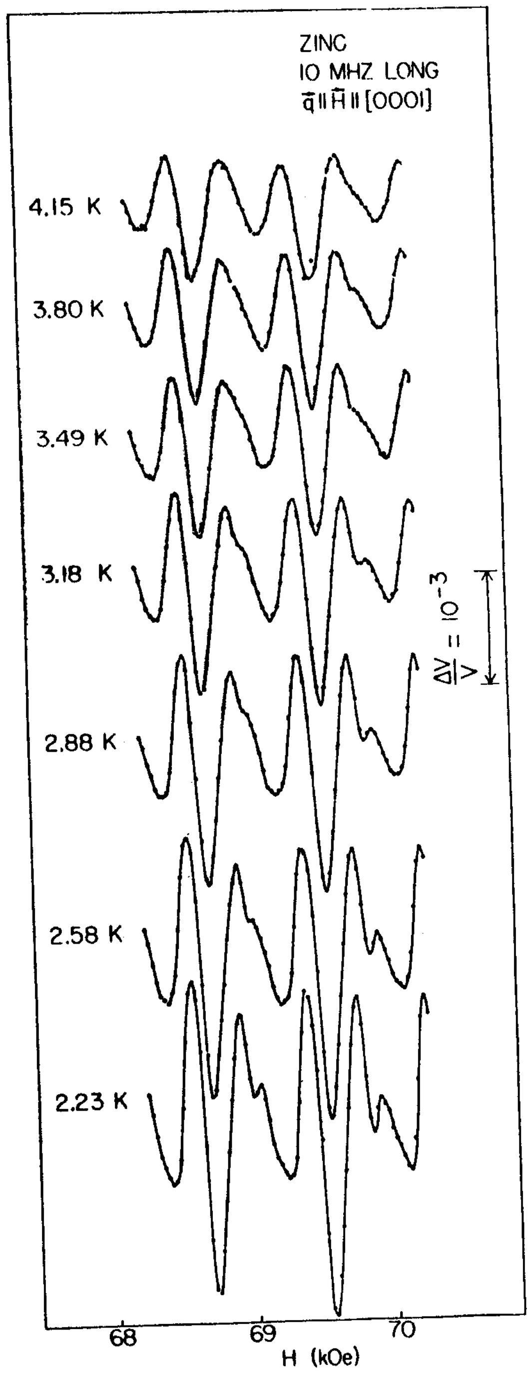
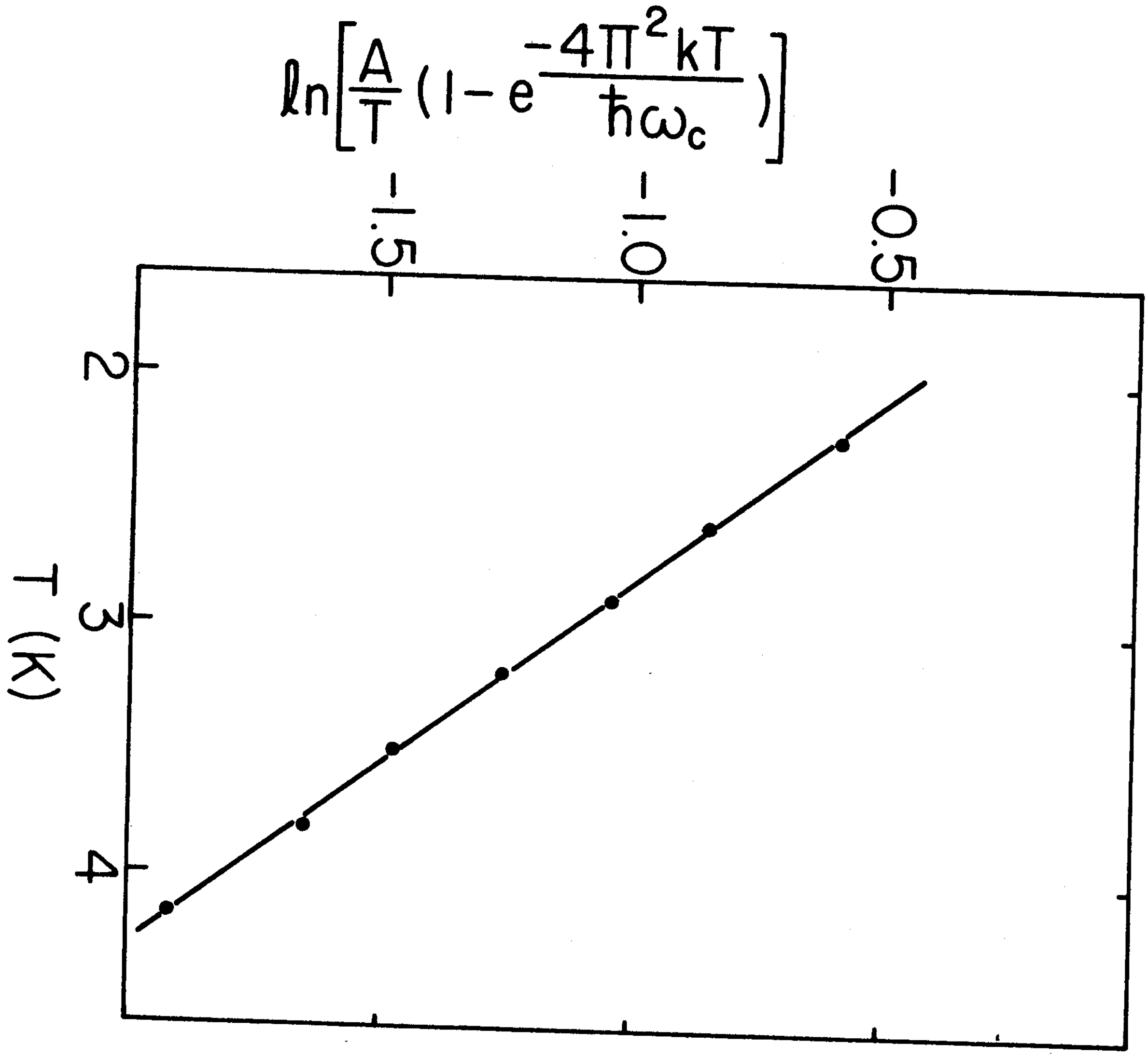

ZINC
10 MHZ LONG
$\vec{q} \parallel \vec{H} \parallel [0001]$
4.15 K
3.80 K
3.49 K
3.18 K
2.88 K
2.58 K
2.23 K
$\frac{\Delta V}{V} = 10^{-3}$
68
69
70
H (kOe)

FIG. 1



$\ell n\left[\frac{A}{T}\left(1-e^{\frac{-4\pi^2 kT}{\hbar\omega_c}}\right)\right]$
-1.5
-1.0
-0.5
T (K)
2
3
4

FIG. 2