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Stress Dependence of the Needle-Shaped Electron Pockets in Zinc*

L. M. Reitz
General Electric Lamp Division, Cleveland, Ohio

and

D. M. Sparlin
Department of Physics, University of Missouri-Rolla, Rolla, Missouri 65401

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The stress dependence of the cross-sectional area of the needle-shaped electron pockets in zinc was determined by magnetostriction techniques in which length changes were measured using a three-terminal capacitance method. Magnetization data were obtained by employing a torque balance. The stress coefficients of the needle pockets are

$$\frac{\partial \ln S_m}{\partial \sigma[0001]} = -(6.0 \pm 0.2) \times 10^{-10}\ (\text{dyn/cm}^2)^{-1}, \qquad \frac{\partial \ln S_m}{\partial \sigma[10\bar{1}0]} = +(2.4 \pm 0.2) \times 10^{-10}\ (\text{dyn/cm}^2)^{-1},$$

$$\frac{\partial \ln S_m}{\partial \sigma[11\bar{2}0]} = +(2.5 \pm 0.2) \times 10^{-10}\ (\text{dyn/cm}^2)^{-1}.$$

I. INTRODUCTION

Several of the electronic properties of pure metals at low temperature exhibit oscillatory components in an applied magnetic field. These include the thermal conductivity, thermopower, entropy, reflectance, magnetization, and magnetostriction. The first of these to be discovered was the periodic dependence of the diamagnetic susceptibility on the reciprocal of the magnetic field, the "de Haas–van Alphen" (dHvA) effect. The first theory of the dHvA effect was derived by Landau, who showed that susceptibility is indeed periodic in $1/H$. The contributions of Onsager[1] and of Lifshitz and Kosevich[2] brought the theory to its current form. These dHvA oscillations have now been observed in many metals and several semimetals, including zinc.

Marcus[3] first observed the dHvA effect in zinc. Dimitrenko, Verkin, and Lazarev,[4] Joseph and Gordon,[5] and Higgins, Marcus, and Whitmore[6] contributed important measurements of the dHvA effect in zinc.

In a series of papers, Harrison proposed models for the Fermi surfaces of polyvalent metals[7] based on the single orthogonalized plane wave (OPW) approximation. Harrison applied the single-OPW approximation to a series of polyvalent metals including zinc and achieved remarkable agreement between deduced and observed electronic properties.

Additional information about the Fermi surface of zinc has been obtained from experiments that effectively altered the size of the Fermi surface. Higgins and Marcus[8] varied the number of electrons per atom by alloying with copper and aluminum. By taking changes in the c/a ratio into consideration, the model correctly predicts the direction and magnitude of the changes in the "needle" oscillation frequency and the direction but not the magnitude of the change in the horizontal-arm oscillation frequency. Alloying apparently did not affect the vertical arm.

Verkin and Dimitrenko,[9] along with Lazarev,[4] have investigated the effect of changing lattice dimensions by hydrostatic pressure. O'Sullivan and Schirber[10] have also measured the pressure derivatives of the areas (dHvA frequencies) of the needle, horizontal-arm, and vertical-arm sections. Their results agree qualitatively with the predictions of the nearly free-electron model. For the needle oscillation, the relation of the pressure (c/a) dependence to the alloying (electron density) effect agrees very well with the model. In discussing the magnetostriction studies presented here, reference will again be made to work of O'Sullivan and Schirber.

Chandrasekar[11] showed that the quantization of electron energy levels that gives rise to the dHvA effect ought also to cause an oscillatory component in the magnetostriction. Shortly thereafter Green and Chandrasekar[12] demonstrated this phenomenon in bismuth. Since the initial discovery, oscillatory magnetostriction has also been observed in several metals, including zinc, beryllium, antimony, and n-GaSb.

The first observation[13] of magnetostriction effects in zinc disclosed oscillations associated with orbits on the needles and the horizontal arms. In subsequent work oscillations arising from the vertical arms were also observed. The present study was undertaken to determine the uniaxial stress dependence of the extremal cross section of the needle-shaped pockets. These results are to be compared with hydrostatic pressure coefficients of O'Sullivan and Schirber.

The uniaxial stress coefficients can be evaluated in terms of strain ϵ_i and magnetization M_H (parallel to H) as follows: Consider the Gibbs free energy of Ref. 2 to be of the form $G(\sigma, H) = G_0(\sigma, H) \times \cos(cS_m/ehH)$. Then

$$\epsilon_i = -\left(\frac{\partial G}{\partial \sigma_i}\right)_{H,T} = -\frac{\partial G_0}{\partial \sigma_i} \cos\frac{cS_m}{ehH} - G_0\left(-\sin\frac{cS_m}{ehH}\right)\left(\frac{c}{ehH} \frac{\partial S_m}{\partial \sigma_i}\right) , \quad (1)$$

$$M_H = -\left(\frac{\partial G}{\partial H}\right)_{\sigma,T} = -\frac{\partial G_0}{\partial H} \cos\frac{cS_m}{ehH} - G_0\left(-\sin\frac{cS_m}{ehH}\right)\left(-\frac{cS_m}{ehH^2}\right) . \quad (2)$$

It is assumed that G_0 is a slowly varying function of σ and H so that $\partial G_0/\partial\sigma$ and $\partial G_0/\partial H$ can be neglected. On dividing (1) by (2), we have

$$\frac{\epsilon_i}{HM_H} = \frac{-\partial \ln S_m}{\partial \sigma_i} . \quad (3)$$

The observed amplitudes of ϵ_i, M_H, and H, are combined to yield the uniaxial stress derivative of the extremal area, S_m.

II. EXPERIMENTAL TECHNIQUES

A. Growing Single Crystals

Single crystals of zinc were grown using the Bridgman-Stockbarger technique. Metal shot 99.9999% pure was sealed in an evacuated ampoule which was heated to about 20 °C above the melting point of zinc (419.5 °C) and lowered through a sharp temperature gradient. All samples were cut from a single pencil-shaped slug 0.4-in. diameter and 5-in. long, of which 80% was a single crystal. The residual resistance ratios were in the range (1–2) $\times 10^4$.

B. Preparing Samples

The slug was oriented by standard Laue technique and spark cut into bar samples approximately $2\times2\times10$ mm. The samples were acid etched to remove any slight surface damage that might have been incurred in cutting. The strain is measured within 3° of the desired crystallographic axis in the completed cells.

C. Capacitance-Strain Measurement

The probe was suspended in a stainless-steel liquid-helium Dewar (insulated with a liquid-nitrogen jacket), where temperatures to 1.2 °K were attained by pumping on the bath. The tail of the Dewar was positioned between the 3-in.-diam pole tips of an 11-in. Magnion electromagnet. A Magnion FFC-4 precision magnet power supply provided fields to 26 kG.

The measuring circuit is shown schematically in Fig. 1(a) and the capacitance bridge in simplified form in Fig. 1(b). The two inductive arms of the bridge constitute the secondary winding of a ratio transformer; they are energized through a common primary by the amplifier. The condition of balance (zero current through the detector) is realized when $V_K C_K = V_U C_U$ or $C_U/C_K = V_U/V_K = N_U/N_K$, where N_U and N_K are the numbers of turns in the U and K arms of the secondary. The value of the turns ratio is very precisely known. The element C_K is a known variable capacitance; C_U is the capacitance of the cell in zero applied field. (The cell is a three-terminal capacitor; the third terminal, the cell body, must be well grounded.) As the zinc sample elongates and contracts during a field sweep, the value of C_U oscillates and the voltage amplitude at the detector varies accordingly as the bridge swings back and forth through the balanced condition. The detector used is a phase-sensitive amplifier tuned and locked onto the 5-kHz audio-modulation frequency. The detector produces a dc signal proportional to the amount by which the cell capacitance is different from the value that balances the bridge. This dc voltage is fed to an x-y recorder as the y variable. An F. H. Bell model 701 Hall probe fastened to the magnet pole face provides a dc signal proportional to the magnetic field which drives the x axis of the x-y recorder.

The measuring circuit components are commercially available units. A Princeton Applied Research model JB-4 lock-in amplifier supplied the audio signal and served as the phase-sensitive detector. A Bogen high-fidelity audio amplifier drove the bridge. The bridge was a General Radio type 1615-A capacitance bridge. The data were

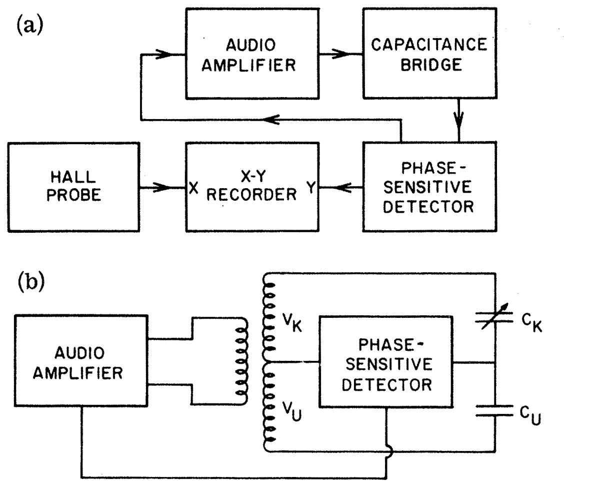

FIG. 1. (a) Interconnection diagram of the experimental apparatus. The Hall probe was a F. H. Bell model FP-701 and was taped to the pole face of the Harvey Wells 11-in. electromagnet. The x–y recorder was a Moseley 7001A, the phase-sensitive detector was a Princeton Applied Research JB-4. The capacitance bridge was a General Radio 1615A, and was driven by a Bogen amplifier with a 100-W capability. (b) Capacitance bridge in simplified form. The principle of operation of the ratio-transformer capacitance bridge (General Radio model 1615A) can be seen in the figure.

recorded on a Moseley model 7001A autograf x-y recorder.

The strain ϵ_t can be calculated from the change in the cell capacitance C as the sample dilates and contracts:

$$C=\frac{KA}{d}, \quad \frac{\Delta d}{L_0}=-\frac{\Delta L}{L_0}=\frac{KA\Delta C}{L_0C^2} \tag{4}$$

(K is the permittivity of space, A the area of the capacitor plates, and L_0 is the length of the sample). Figure 2 is a typical magnetostriction-capacitance recorder trace. For this case, the diameter of the capacitor plate and the length of the sample were both about 1 cm; for $\Delta C=10^{-5}$ pF and $C=2.78$ pF, ϵ is of order 10^{-8}. The construction of the cells follows the general design used by White in measurements of thermal expansion.[14]

D. Measurement of Magnetization

Since zinc is anisotropic, the magnetization M is parallel to the magnetic field only along high-symmetry directions. In other directions there is a transverse as well as a parallel component to M and torque will be generated in the sample. The parallel component can be determined using field-modulation techniques; the transverse components are measurable in terms of the induced torques. The magnetization along the c axis M_c can be calculated from torque data if a geometrical model for the given section of the Fermi surface is available. The needle-shaped electron pocket closely resembles a circular cylinder. With this approximation, M_c is readily derived from the measured torque (about an axis perpendicular to H and c) with the field at some angle θ from c. Since the magnetization of a cylindrical Fermi surface always points along the cylinder axis (in this case the c axis)

$$M_\perp = M_c \sin\theta \tag{5}$$

and

$$\tau = M_c H \sin\theta \tag{6}$$

or

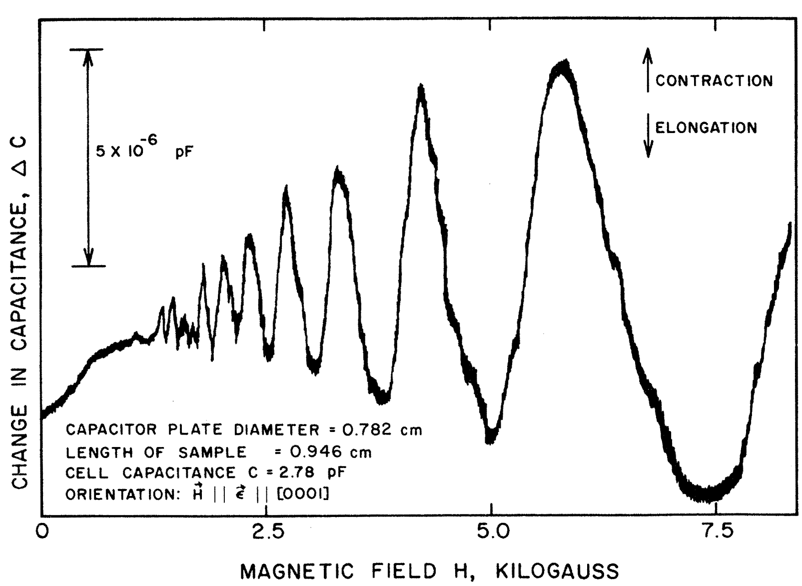

FIG. 2. Plot of the recorded change in capacitance as the magnetic field was swept. Note that the sample is contracting at $H=6000$ G. The oscillatory strain amplitude at this field is -5.23×10^{-8} peak to peak.

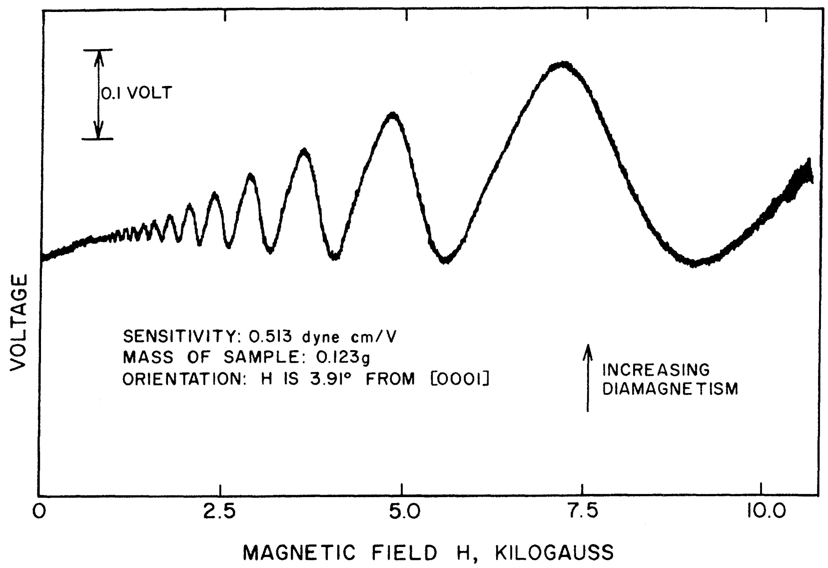

FIG. 3. Plot of the recorded torque as a function of the magnitude of the field applied 3.91° from [0001].

$$HM_c = \tau/\sin\theta \ . \tag{7}$$

The torsion balance[15] was calibrated using a torsion pendulum. The electromagnet, Hall probe, Dewar flask, and x-y recorder are the same as those used for the magnetostriction-capacitance measurements.

Figure 3 is a tracing of a typical torque measurement. At 7500 G the signal amplitude is about 0.187 V; the torque is then (0.187 V) (0.513 dyn cm/V) = 9.59×10^{-2} dyn cm.

The evaluation of the stress coefficients $\partial \ln S_m/\partial\sigma_i$ requires that τ be given as torque per unit volume. Since the values of τ taken directly from the recorder charts refer to the entire mass of the sample, the calculation of the coefficients is

$$\frac{\partial \ln S_m}{\partial \sigma_i} = -\frac{\epsilon_i m}{\tau\rho}\sin\theta \ , \tag{8}$$

where m is the mass of the sample and ρ is the density of zinc. Referring once again the data in Figs. 2 and 3 for $\vec{H} \parallel \epsilon \parallel [0001]$, at 6000 G, we find

$$\frac{\partial \ln S_m}{\partial \sigma_c} = -\frac{\epsilon_c m \sin(3.91°)}{\tau\rho}$$

$$= -\frac{(5.23\times10^{-8})(0.0682)}{9.6\times10^{-2}\ \text{dyn cm}}\ \frac{0.123\ \text{g}}{7.36\ \text{g/cm}^3}$$

$$= -6.2\times10^{-10}\ (\text{dyn/cm}^2)^{-1}$$

$$= -62\times10^{-2}\ \text{kbar}^{-1} \ . \tag{9}$$

The sine value is included according to Eq. (8). The density was temperature corrected using the data of Meyerhoff and Smith.[16]

III. DISCUSSION OF RESULTS

The effect of mechanical damage, dislocations, and impurities can be described in terms of a damping factor $\exp(-2\pi k x/\beta H)$, multiplied times the oscillatory amplitudes calculated in Ref. 2, where x is the so-called Dingle temperature. If the zinc specimens employed in the magnetostriction measurements and those with which the torque data were obtained are identical with respect to content of these kinds of imperfections, then the ratio of normalized strain to normalized torque has the value unity, irrespective of the magnetic field. That is, where H' is the chosen normalization field, the value of $\epsilon(H)\tau(H')/[\epsilon(H')\tau(H)]$ is constant for all values of H. The ratio was evaluated with H along [0001] and ϵ_i parallel to [0001], [10$\bar{1}$0], and [11$\bar{2}$0]. Over the range of fields used in this work, this ratio was constant within experimental error. The data for the longitudinal case deviated least from being constant at unity, those for $\epsilon \parallel$ [11$\bar{2}$0] deviated most. In all cases deleting the low-field readings (which are experimentally most uncertain) brought about a closer approach to constant unit value. It was concluded that the Dingle temperature was in effect the same for all samples.

The values of the stress coefficients obtained from these measurements are given in Table I. The estimated limit of error in these values is approximately 15%. There appears to be a systematic difference between the two transverse coefficients in that at every field given in Table I except the very lowest the [11$\bar{2}$0] value is greater than the [10$\bar{1}$0]. They are taken to be indistinguishable within the experimental uncertainty of these measurements.

The average value of the sum of the three partial derivatives is $-(1.1\pm0.2)\times10^{-10}$ $(\text{dyn/cm}^2)^{-1}$ $= -(11\pm2)\times10^{-2}$ $(\text{kbar})^{-1}$. Assuming that the extremal cross-sectional area is an independent function of the three uniaxial stresses, the sum of the

TABLE I. The stress coefficients $\partial \ln S_m/\partial\sigma_i$ of the needle-shaped electron pockets. Units: 10^{-10}(dyn/cm^2)$^{-1}$.

Field (G)	[0001]	[10$\bar{1}$0]	[11$\bar{2}$0]	Sum
9000	−5.98	2.59	2.82	−0.57
8000	−6.05	2.47	2.74	−0.84
7000	−6.17	2.39	2.74	−1.04
6000	−6.13	2.33	2.70	−1.10
5000	−6.20	2.33	2.73	−1.14
4000	−5.5	2.24	2.35	−0.91
3000	−6.0	2.21	1.70	−2.09
Average	−(6.0 ± 0.2)	(2.4 ± 0.2)	(2.5 ± 0.2)	−(1.1 ± 0.5)

uniaxial stress coefficients is equal to the negative hydrostatic coefficient. O'Sullivan and Schirber[10] used a hydrostatic-pressure technique to determine the pressure coefficient of the area of the cross section of the needle-shaped pocket, with the result

$$\frac{d\ln S_m}{dP} = (32.0 \pm 1.5)\times 10^{-2}\ (\text{kbar})^{-1}\ . \qquad (10)$$

Our value corresponds to

$$\frac{d\ln S_m}{dP} = (11 \pm 4)\times 10^{-2}\ (\text{kbar})^{-1}\ . \qquad (11)$$

The difference of a factor of 3 between the direct pressure results of Schirber and O'Sullivan and that calculated from our values for the uniaxial-stress dependence is well outside the limit of random error. Our best estimate of the effects of constraining the ends of the samples, which are due to soldering to the magnetostriction cell, is that the measured strain will be smaller than the free-sample strain by at most 15%. A smaller effect will operate in the case of strain measured along the c axis. The result will be to increase the computed value of the total pressure derivative of the extremal area.

Gerstein and Elbaum have recently studied the uniaxial-stress dependence of the needle cross section by direct loading of the crystal and studying the phase shift of the oscillatory magneto-acoustic attenuation.[17] They report a value for the uniaxial-stress dependence of the needle with stress along the [0001] axis of approximately one-half the nearly free-electron value of 14.6×10^{-10} (dyn/cm^2)$^{-1}$. Their preliminary value of $-(7.0\pm0.7)\times10^{-10}$ (dyn/cm^2)$^{-1}$ is in fair agreement with our value of $-(0.6\pm0.2)\times10^{-10}$ (dyn/cm^2)$^{-1}$.

The agreement between the two measurements of the uniaxial-stress coefficients performed by different techniques is in contrast with the hydrostatic-pressure result, which is larger by a factor of 3. Perhaps this is because the magnetostriction measurements are performed at zero hydrostatic pressure, while the uniaxial-stress measurements were performed at a modest average applied stress and the hydrostatic-pressure measurements were performed at a considerably larger average stress. If the stress coefficients are stress dependent, then perhaps these three measurements will be found to be in substantial agreement.

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[17]Charles Elbaum (private communication). Their value for $\partial\ln S_m/\partial\sigma_{[0001]} \approx -(7.3\pm0.5)\times10^{-10}$ (dyn/cm^2)$^{-1}$. This is to be compared with our value of $-(6.0\pm0.2)\times10^{-10}$ (dyn/cm^2)$^{-1}$.