spin interaction line shape, while calcium fluoride provides the contrasting situation of a simple cubic lattice where all spins are equivalent. In both materials, when the H_1 field was varied from 3 to 30 gauss for a given pulse length, no shift could be observed in the f.i.d. zeros. The entire visible calcium fluoride f.i.d. shape was also checked for distortion for many pulse-length and field-strength combinations. None was found within the 7% reading accuracy of our data.

The shift of the crossing point vs pulse length was also checked for the proton line shape of a single crystal of ice, and polycrystalline ice. Again a line of slope $\frac{1}{2}$ fitted the data very well. The polycrystalline sample was also examined at -5°C where the internal motion washes out the zeros and substantially changes the f.i.d. shape to that of a Gaussian. Again the line shape was found to be undistorted for pulse lengths of from 2 to 10 μsec, provided the f.i.d. shape was assumed to begin decaying at the center of the rf pulse. The $1/e$ point of the Gaussian was 16 μsec.

From the gypsum and ice data it appears that the two-spin calculation is quantitatively verified, and the calcium fluoride data seems to indicate that to a high degree of accuracy the same result applies to the n-spin case. We thus conclude that while the H_1 field is on, the rate of decrease of magnetization is reduced by a factor of two. These experiments also show that for long rf pulses, the undistorted f.i.d. is observed provided the time origin is taken to be the center of the rf pulse.

A final note: It is easily proven that for the spin system described by $\mathcal{H}_1$, and for $\theta = 90°$,

$$M_x(\tau) = -(1/\omega_1)(\partial/\partial\tau)M_z(\tau).$$

From our measurements $M_x(t)$, we obtained information about $M_x(\tau)$. Thus our measurements are closely related to those of Goldburg and Lee as presented in the previous paper.[3] Since we measured line shapes and not line amplitudes, our measurement technique is not sensitive to the effects of time-dependent terms in $\mathcal{H}_2$.

*This work was supported by grants from the Alfred P. Sloan Foundation, the National Science Foundation, and the U. S. Air Force Office of Scientific Research.

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MAGNETOMORPHIC OSCILLATIONS IN THE HALL EFFECT AND MAGNETORESISTANCE IN CADMIUM*

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Large-amplitude oscillations periodic in the magnetic field have been found in the Hall resistivity ρ_{21} of cadmium single crystals at liquid helium temperatures[1] (see Figs. 1 and 2). Oscillations of the same period and comparable amplitude have been observed also in the transverse magnetoresistivity ρ_{11}. The oscillations differ in phase by about 55° or 130 G. These phenomena appear to be the size-effect oscillations predicted by Sondheimer[2] and first observed by Babiskin and Siebenmann.[3]

Samples and experimental conditions. —Two single crystals, Cd1 and Cd2, were cut in the form of rectangular plates from a zone-refined cadmium bar (Tadanac).[4] The crystals are oriented with the hexagonal axis perpendicular to the large faces and the binary axis at about 17° with respect to the longest dimensions. The dimensions of the samples are, for Cd1, $0.8\times5\times19$ mm; for Cd2, $2.3\times5.5\times15$ mm.

The experimental arrangements were such that the magnetic field was parallel to the hexagonal axis (normal to the large faces of the plate) and the current was in the direction of the longest dimension. Changes in voltage could be measured with a precision of $\pm10^{-9}$ V.

Size dependence. —The period P and the amplitude of the oscillations $|\tilde{\rho}_{21}|$ in ρ_{21} decrease as thickness increases, as seen in Table I and Fig. 3(b). The onset of oscillations occurs at a lower field in the thick sample, Cd2, than in Cd1.

Orientation dependence. —The magnetic field was rotated in the plane normal to the length of the sample. As the angle ϕ between the hexagonal axis and the magnetic field direction is in-

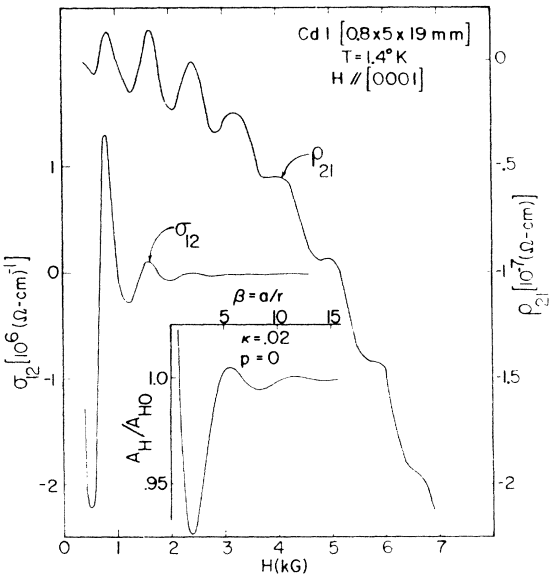

FIG. 1. Magnetomorphic oscillations in the Hall resistivity ρ_{21} and the Hall conductivity σ_{12} for sample Cd1. The insert shows the oscillations in the Hall constant ($A_H = \rho_{21}/H$ and A_{H0} is the Hall constant of the bulk material) as predicted by the Sondheimer[2] theory, where p is a parameter specifying the proportion of specular reflections at the surface and $\kappa = a/\lambda_B$. This compares to Sondheimer's Figs. 4 and 5.

creased from zero, the amplitude of the oscillations observed in Cd1 at 1.4°K decreases; and the positions of the peaks move toward lower fields. The oscillations disappear for $\phi > 25°$.

Temperature dependence. —The oscillations in Cd1 were observed at 4.2°K, 3°K, 2.1°K, and 1.4°K. The oscillations were not found in Cd2 at 4.2°K, but were observed at 1.4°K. The period in Cd1 does not change with temperature; but the amplitude of the effect increases essentially linearly as the temperature is decreased (Table II). The temperature dependence of the resistivity of each sample and the corresponding resistance ratios are shown in Table II and Fig. 3(c).

Discussion. —At all three temperatures investigated, the resistivity of Cd2 (thick sample) is larger than that of Cd1, although the effect of surface scattering would be expected to cause the contrary. This result illustrates a feature of Sondheimer's[2] theory: In zero magnetic field for completely diffuse boundary scattering and for the case $a/\lambda_B < 1$, the ratio of the resistivity of a thin plate to that of the bulk material is given

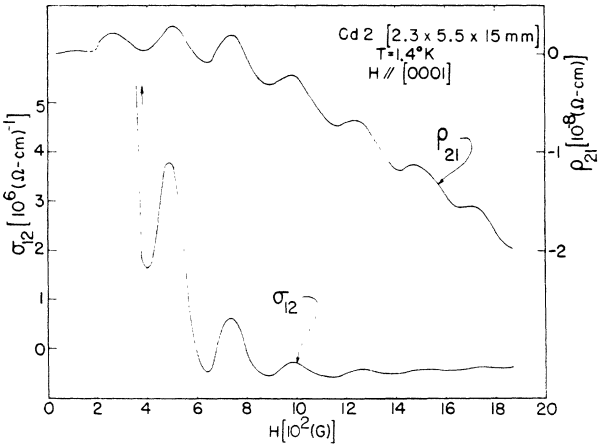

FIG. 2. Magnetomorphic oscillations in the Hall resistivity ρ_{21} and the Hall conductivity σ_{12} for sample Cd2.

by

$$\frac{\rho}{\rho_B} = \frac{4}{3}\frac{\lambda_B}{a}\frac{1}{\ln(\lambda_B/a)}, \tag{1}$$

where a is the sample thickness, λ_B is the bulk mean free path, and ρ_B is the bulk resistivity. For two samples of thicknesses a_1 and a_2, this gives

$$\frac{\rho_1}{\rho_2} = \frac{a_2}{a_1}\frac{\ln(\lambda_B/a_2)}{\ln(\lambda_B/a_1)}. \tag{2}$$

For $a_2/a_1 > 1$, the situation $\rho_1/\rho_2 < 1$ occurs for certain values of λ_B. For $a_2/a_1 = 3$ (roughly the present case), the condition for $\rho_1/\rho_2 < 1$ is simply

$$\lambda_B < \sqrt{3}\, a_2; \tag{3}$$

Table I. Size dependence of magnetomorphic oscillations at 1.4°K.

	Cd1	Cd2
Period (G)	840	250
$\lvert\tilde{\rho}_{21}\rvert$[a] (Ω-cm)	24×10^{-9}	0.4×10^{-9}
First peak ρ_{21} (G)	600	185
$R_{300°K}/R_{4.2°K}$	12 700	3 860

[a] $\lvert\tilde{\rho}_{21}\rvert$ measured at $H \simeq 2$ kG.

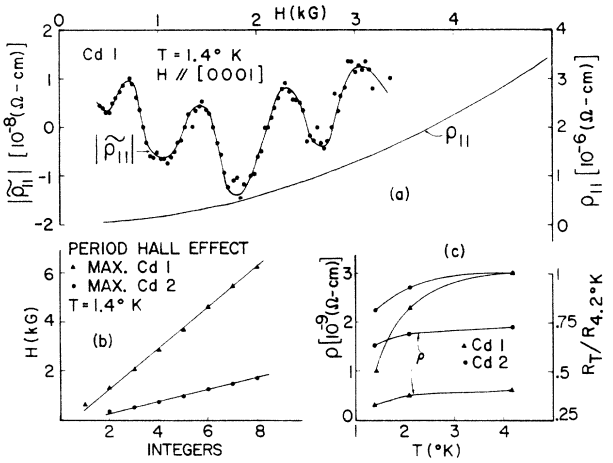

FIG. 3. (a) Magnetomorphic oscillations in the transverse magnetoresistivity ρ_{11}. The oscillatory term $|\tilde{\rho}_{11}|$ was isolated by subtraction of a quadratic term determined from a least-squares fit of the experimental points. (b) Plot of the field values corresponding to the maxima of the ρ_{21} oscillations vs integers for each sample. (c) Temperature dependence of the resistance ratio $R_T/R_{4.2°K}$ and the zero-field resistivity for each sample vs temperature.

this limit is about 4 mm for $a_2 = 2.3$ mm.

The value of λ_B is calculated from Eq. (2) from the measured ρ_1 and ρ_2 values. The result is $\lambda_B \simeq 2.6$ mm, essentially independent of temperature.

The influence of size on the relative change in resistance, Fig. 3(c), is analogous to Olsen's observations[5] on thin indium wires and corroborates the mechanism of enhanced effectiveness of small-angle phonon scattering in thin samples.[5,6]

The only available theories[4,7-9] of magnetomorphic effects are based on a free-electron model. Sondheimer's theory[2] for thin infinite sheets predicts an oscillatory behavior of the resistivity and the Hall effect[10] in a transverse magnetic field. Maxima of ρ/ρ_B occur periodically in $\beta = a/r = aeH/pc$ with a period of $\Delta\beta \simeq 6$, where ρ_B is the resistivity of the bulk metal, r the radius of the free-electron cyclotron orbit, and $p = m^*\bar{v}$ the momentum of the electrons at the Fermi surface. The first peak is a maximum corresponding to $a = r$, and

$$H_0 = pc/ae. \quad (4)$$

The oscillations described here agree with Sondheimer's theory in the following respects (although the one-electron theory is hardly adequate for divalent metals such as cadmium): the periodicity in H, the displacement of the first peak toward lower fields for increased sample thickness, the increase in amplitude with decrease of temperature, the dependence of the period on sample thickness, and the $|\tilde{\rho}_{21}|$ lag in phase. There is disagreement on two important points: The first peak occurs in all instances at a higher field than would be expected from the period [Fig. 3(b)]; and the first peak in the magnetoresistivity is a minimum [Fig. 3(a)].[3]

To test the Sondheimer theory further, we take the value $H_0 = (1/6)P$ as corresponding to $r = a$.

Table II. Temperature dependence of galvanomagnetic effects.

T (°K)	4.2	3.0	2.1	1.4
$\lvert\tilde{\rho}_{21}\rvert$ (Cd1)[a] (Ω-cm)	1.2×10^{-9}	9×10^{-9}	15×10^{-9}	24×10^{-9}
ρ_{21} (Cd1)[b] (Ω-cm)	-5.7×10^{-8}	-5.4×10^{-8}	-14×10^{-8}	-14×10^{-8}
ρ_{11} (Cd1)[b] (Ω-cm)	1.5×10^{-6}	3.7×10^{-6}	4.6×10^{-6}	4.7×10^{-6}
R (Cd1)[c] (Ω)	1.53×10^{-8}		1.26×10^{-8}	0.77×10^{-8}
R (Cd2)[c] (Ω)	1.03×10^{-8}		0.95×10^{-8}	0.83×10^{-8}

[a] $|\tilde{\rho}_{21}|$ measured at $H \simeq 2$ kG.

[b] ρ_{21} and ρ_{11} measured at $H = 5836$ G; distance between magnetoresistance probes 10 mm for Cd1, 7 mm for Cd2.

[c] R measured in zero field.

The first experimental peak occurs at $\sim 3.5H_0$ rather than at H_0. Equation (4) then gives $p = 1.79\times10^{-19}$ g-cm/sec, which, for the free-electron value $\bar{v} = 1.62\times10^{8}$ cm sec^{-1}, gives the effective mass $m^* = 1.22m_0$. This result is identical to that of Galt, Merritt, and Schmidt[11] obtained from cyclotron resonance in the [0001] direction.

We attempt to correlate the present results with those obtained from a two-band fit[12] of the magnetoconductivity, using[13]

$$\sigma_{11} = \frac{\rho_{11}}{\rho_{11}^2+\rho_{21}^2} = ec\sum n_i\left(\frac{H_i}{H^2+H_i^2}\right),$$

$$\sigma_{12} = \frac{\rho_{21}}{\rho_{11}^2+\rho_{21}^2} = c\sum(\pm e)n_i\left(\frac{H}{H^2+H_i^2}\right),$$

where n_i is the number of carriers in the ith band,

$$H_i = m_i^*c/e\tau = p_ic/e\lambda, \qquad (5)$$

and $\tau = \lambda/\bar{v}$ is the relaxation time (assumed isotropic and equal for both bands). In the present case, the mean free path λ is taken to be the effective value of the mean free path when boundary scattering is important. The band fit of the magnetoconductivity for Cd1 gives $H_1 = 18$ G, $n_1 = 4.2\times10^{21}$ cm^{-3}, $H_2 = 78$ G, and $n_2 = 4.8\times10^{21}$ cm^{-3}. Here n_1 and n_2 are interpreted, respectively, as the number of holes and electrons, in agreement with the change of sign of the Hall effect in cadmium (see Figs. 1 and 2). The band fit for Cd2 was inconclusive.

A comparison of Eqs. (4) and (5) reveals that the two independently determined quantities H_0 and H_i are related as

$$H_0/H_i = \lambda_{\mathrm{eff}}/a. \qquad (6)$$

We assume that the oscillations are due to the electron band since this band is the dominant contributor to the conductivities in the range of field where the oscillations occur. An estimate of λ_{eff} is obtained from Eq. (6) for sample Cd1 as

$$\lambda_{\mathrm{eff}} = 1.4 \text{ mm} \simeq 1.75a_1.$$

A second estimate is obtained from Eq. (1) as

$$\lambda_{\mathrm{eff}} \simeq 1.6a_1,$$

where ρ and ρ_B are assumed to differ only in mean free paths λ_{eff} and λ_B, respectively. Yet a third estimate follows from a simple consideration of the thin sheet geometry, that is,

$$\lambda_{\mathrm{eff}} \lesssim 1.85a_1.$$

Thus, the effective mean free path for conduction in the direction of the sample length seems to be a valid concept and to be somewhat greater than the smallest dimension of the sample.[14]

Assume an oscillation in the mobility (or of the effective mean free path) associated with the electron band and introduce such an oscillation into the Sondheimer-Wilson theory through

$$H_2 \to (H_2)_0[1 + B\sin(2\pi H/840\text{ G})],$$

with B a parameter, $0 < B < 1$. This procedure results in oscillations in ρ_{11} and ρ_{21} of the proper period and, for $B \simeq 0.1$, the proper magnitude. Further, the peak which would be identified experimentally as the first peak occurs at the observed field value. For the first two peaks, $|\tilde{\rho}_{21}|$ lags $|\tilde{\rho}_{11}|$ by about 300 G; for succeeding peaks, the phase difference goes to zero. Finally, the first peak of ρ_{11} is a minimum. We note that one difficulty of this conjecture is the problem of the relative phase of the ρ_{21} and ρ_{11} oscillations.

From the foregoing and other recent work,[1] we see that size effects are important in multivalent metal samples of smallest dimension ~1-10 mm. This suggests caution in the measurement of transport properties of these metals in small samples.

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[1]There has been considerable recent interest in size effects in multivalent metals; e.g., B. N. Aleksandrov and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. 41, 1333 (1961) [translation: Soviet Phys.-JETP 14, 948 (1962)]; V. F. Gantmakher, Zh. Eksperim. i Teor. Fiz. 43, 345 (1962) [translation: Soviet Phys.-JETP 16, 247 (1963)] M. Ya. Azbel' and R. N. Gurzhi, Zh. Eksperim. i Teor. Fiz. 42, 1632 (1962) [translation: Soviet Phys.-JETP 15, 1133 (1962)]; B. N. Aleksandrov, Zh. Experim. i Teor. Fiz. 43, 399 (1962) [translation: Soviet Phys.-JETP 16, 286, 871 (1963)]. These studies relate to effects in zero magnetic field and in longitudinal magnetic fields.

[2]E. H. Sondheimer, Phys. Rev. 80, 401 (1950); Advan. Phys. 1, 1 (1952).

[3]Compare with the oscillations found in the magnetoresistance of Na by J. Babiskin and P. G. Siebenmann, Phys. Rev. 107, 1249 (1957).
[4]The cadmium bar was obtained from Cominco Products, Inc., and had a stated purity of 99.9999%.
[5]J. L. Olsen, Physica 31, 713 (1958)
[6]F. J. Blatt and H. G. Satz, Physica 33, 1007 (1960).
[7]G. Chambers, Proc. Roy. Soc. (London) A202, 378 (1950).
[8]R. B. Dingle, Proc. Roy. Soc. (London) A201, 545 (1950).
[9]D. K. C. MacDonald and K. Sarginson, Proc. Roy. Soc. (London) A203, 223 (1950).
[10]Apparently a truncation in the numerical calculations resulted in the nonoscillatory curves in Sondheimer's Figs. 4 and 5, and led to the stated conclusion that ρ_{21} should show no oscillations. The oscillatory behavior of the Hall effect from Sondheimer's theory is shown in the insert in Fig. 1. The period of the oscillation in ρ_{21} is the same as that in ρ_{11} but ρ_{21} lags in phase by 90°.
[11]J. K. Galt, F. R. Merritt, and P. H. Schmidt, Phys. Rev. Letters 6, 458 (1961).
[12]A. H. Wilson, The Theory of Metals (Cambridge University Press, 1953), 3rd ed.
[13]C. G. Grenier, J. M. Reynolds, and N. H. Zebouni, Phys. Rev. 129, 1088 (1963).
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SPIN DENSITY IN COBALT

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In a recent note Menzinger and Paoletti[1] (MP) reported measurements of the magnetic form factor of fcc cobalt determined from polarized neutron measurements. They find that the shape of the form-factor curve is temperature dependent and they deduce from their data that either the mean-square amplitude due to thermal vibrations of the spin density is only ~0.37 that of the nuclear vibrations or that the spin density is temperature dependent.

It is the purpose of this note (1) to show that the temperature dependence of the spin density is a considerably more likely explanation, (2) to show the variation of the difference in spin-up and spin-down charge density with temperature, and (3) to calculate the occupation of the e_g (doubly degenerate) and t_{2g} (triply degenerate) orbitals of fcc cobalt from the data of MP.

In order to make an estimate of the effect suggested by MP we assume that the spin density close to the nucleus (where the potential is large) follows the nucleus in its thermal displacement from its lattice site, but we permit the outer part of the spin density near the atomic radius to remain relatively motionless. There is a smooth radial variation of displacements from the inner part of the spin density to the outer part approximated by a Debye-Waller factor for each volume of spin density given by

$$\exp[-\tfrac{1}{2}k^2\langle u^2\rangle(1-r^2/a^2)], \qquad (1)$$

where $\langle u^2\rangle$ is the component of the expectation value of the mean-square displacement of the nuclei along the scattering vector and a the atomic radius. Since each volume element of spin density has its own mean-square displacement, we must evaluate both the form factor and Debye-Waller factor simultaneously. We thus find for the structure factor F in fcc cobalt

$$F \cong 4\{b\exp(-M) \pm (P/1-3y)\exp[-(M+2k^2y\alpha)]\},$$
$$\alpha \cong 0.04\ \text{Å}^2,$$
$$y \cong k^2\langle u^2\rangle/40, \qquad (2)$$

where $\exp(-M)$ is the Debye-Waller factor for the nuclei, b the nuclear scattering amplitude, P the measured magnetic scattering amplitude[2] of MP at room temperature, and the term $\exp[-(M+2k^2y\alpha)]/(1-3y)$ the Debye-Waller factor for the spin density. In the region of the MP measurements (T from 300° to 900°K, k from 5 to 9/Å), we find the Debye-Waller factor for the spin density to vary from $\sim 0.91\exp(-M)$ to $\sim 1.0\exp(-M)$, i.e., a generally greater reduction in intensity than for the nuclei. At first sight this seems contrary to our expectations, but in the regions of k from 5 to 9/Å the outer part of the spin density is out of phase with the inner part and its smaller mean-square displacement leads to a larger negative contribution to the form factor. According to MP their observations suggest a Debye-Waller factor for the spin density of $\exp(-0.37M)$ which is approximately $1.04\exp(-M)$ to $1.42\exp(-M)$ over this region of k values.

As an alternative explanation we have assumed