Nonlinear de Haas-van Alphen Effect and Magnetic Domains in Beryllium

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The results of experiments that show that the wave shape of the magnetization oscillations (de Haas-van Alphen effect) depends on sample shape, and some results of magnetothermal oscillation experiments are presented. A theory that is an extension of Shoenberg's idea that the effective field on the conduction electrons is the induction B is developed. It is concluded that magnetic domains, similar in several respects to ferromagnetic domains, occur when the oscillating differential magnetic susceptibility $\partial 4\pi I/\partial B$ is greater than unity.

I. INTRODUCTION

DURING recent experiments[1,2] on the de Haas-van Alphen effect of beryllium, it was found that some of the results did not agree with the current theoretical interpretation[3–5] and so the theory was reinspected.

It was found that magnetic domains are expected to exist whenever the differential magnetic susceptibility $\partial 4\pi I/\partial B$ exceeds unity. The argument is presented in the next section, and it will be seen that it is based on classical magnetics and thermodynamics.

In Sec. III experiments will be described and it is shown that their results confirm the theory.

In Sec. IV several other possible theories are considered; however, they do not explain the experimental results.

The results of a general treatment of the B-H effect are described in Appendix I and another derivation of the idea that $M=M(B)$ is presented in Appendix II.

II. THEORY OF THE NONLINEAR EFFECT IN BERYLLIUM

In this section it is shown that a simple extension of Pippard's[4] work leads to theoretical contradictions. Also, it is shown that a magnetic domain structure exists because it has the lowest free energy. The expected results of magnetization and magnetothermal oscillation experiments are calculated. (Comparison to experiment will be made in Sec. III).

The large amplitude of the magnetization oscillations and the sample-shape-dependence of the experimental results (Sec. III) indicate that the predominate effect is Shoenberg's B-H effect,[3] i.e., the magnetization is a function of the magnetic induction B inside the sample, and B is a function of the applied magnetic field, the magnetization intensity, and the sample-shape-dependent demagnetization factor.

This paper is primarily concerned with explaining H-A effect experimental results of beryllium, and there are certain features of it that simplify the theoretical problem. With the applied field direction near the hexagonal axis, there are two predominate H-A frequencies ($F\simeq 1.0\times 10^7$ G) that differ by about 3% so that there is a beating pattern in the data that has ~ 34 oscillations per beat. Watts[6] has identified the pieces of the Fermi surface that cause these frequencies and has shown them to be nearly cylindrical in shape and aligned with the hexagonal axis. Because of this, it follows that the direction of magnetization is nearly along the axis and that the phase of the oscillations depends only on the component of the field along the axis. (See Appendix I.) In this section the symbols H and B will mean the axial components of the magnetic field intensity and the magnetic induction field, respectively.

Pippard[4] has considered the problem of magnetic interaction and has shown by an argument involving the entropy density that Shoenberg's conjecture is true, i.e., that the H-A oscillations are a simple oscillating function of the magnetic induction B. (See Appendix II.) Over limited ranges of field the magnetization can be closed approximated by

$$4\pi I = 4\pi I_0 \sin kB. \tag{1}$$

He considered the case of a thin rod oriented along the direction of magnetization and showed that the magnetization would be as shown in Figs. 1(a) and 1(b). The multiple valued function in Fig. 1(b) is not physical, and he resolved the problem by considering the free energy ($G=-\int I dH_0$) which is shown in Fig. 1(c). The magnetization will follow the dashed vertical lines shown in Fig. 1(b) because that corresponds to the lowest free energy. There might be supercooling to the metastable states, but the region of the solid curve between the open circles in Fig. 1(b) is unstable and such states can never really exist.

The case of a thin disk perpendicular to the magnetization is quite different and Pippard did not present it. A straightforward extension of the theory leads to contradiction with the behavior of a rod. If one assumes that the magnetization of the disk is uniform, then the induction inside the sample B is equal to the applied field H_0 and so Eq. 1 becomes

$$4\pi I = 4\pi I_0 \sin kH_0 \quad \text{or} \quad y = a \sin x_0, \tag{2}$$

[1] M. H. Halloran and F. S. L. Hsu, private communications and Bull. Am. Phys. Soc. **10**, 350 (1965).
[2] J. H. Condon, Bull. Am. Phys. Soc. **10**, 350 (1965).
[3] D. Shoenberg, Phil. Trans. Roy. Soc. (London) **A255**, 85 (1962).
[4] A. B. Pippard, Proc. Roy Soc. (London) **A272**, 192 (1963).
[5] R. D. Plummer and W. L. Gordon, Phys. Rev. Letters **13**, 432 (1964).
[6] B. R. Watts, Phys. Letters **3**, 284 (1963) and Proc. Roy. Soc. (London) **A282**, 521 (1964).

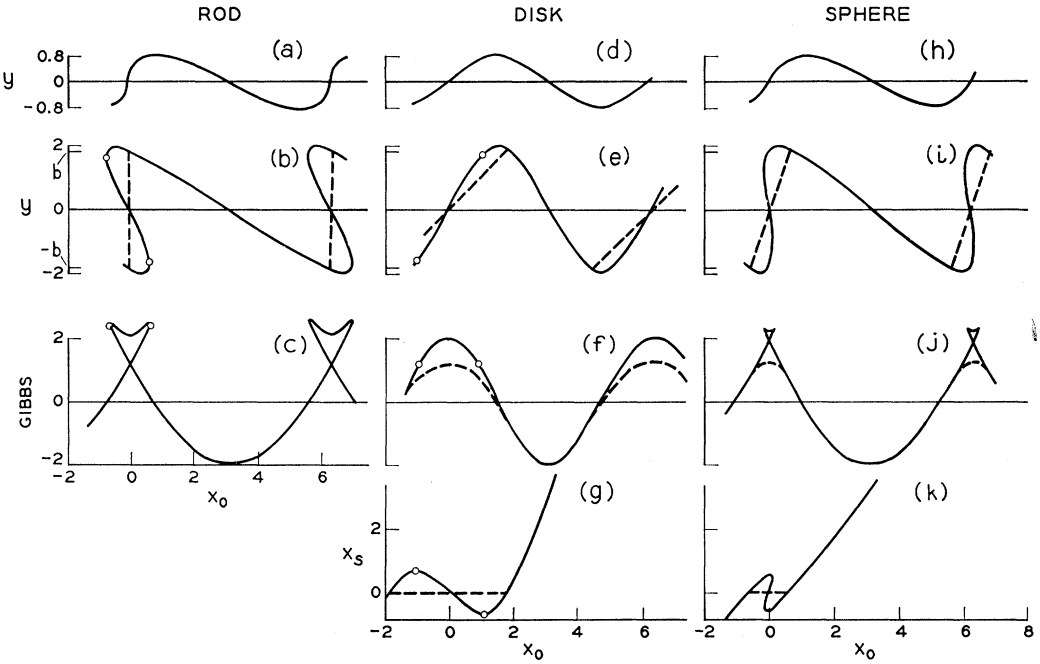

FIG. 1. (a) Pippard's solutions of the magnetization of a rod with his a factor equal to 0.8; (b) his solution with $a=2.0$, the dashed line shows the behavior required by the thermodynamic argument of Pippard; (c) the normalized Gibbs function that corresponds to (b) above; (d) the solution of Eq. 2 with $a=0.8$, the magnetization of a disk-shaped sample; (e) similar to (d) but with $a=2.0$, the dashed line shows the behavior during domain formation; (f) the normalized Gibbs function of a disk shaped sample, the dashed line is the lower free energy of the domain structure; (g) the magnetic field intensity inside a disk-shaped sample as a function of the applied magnetic field; (h)–(k) Are similar to (d)–(g) except they are for a spherical sample. The regions between the open circles are thermodynamically unstable. The quantities y and x are defined in Eq. 2 of text, and Gibbs is the same as g of Eq. 4.

where $y=4\pi kI$, $x_0=kH_0$ and $a=4\pi kI_0$. This is shown by the solid curves in Figs. 1(d) and 1(e). However, this is inconsistent with the behavior of the rod. Consider the disk as composed of a large number of parallel thin rods whose length is the thickness of the disk and require each to have the same magnetization (i.e., require uniform magnetization of the disk). Each rod must behave as the rod previously considered, but now the applied H for each rod H_r is the magnetic field inside the disk, $B-4\pi I$. But since $B=H_0$ for a disk,

$$H_r=H_0-4\pi I \quad \text{or} \quad x_r=x_0-y. \tag{3}$$

The function x_r is shown in Fig. 1(g); the y of Fig. 1(e) was used to obtain it. The three solutions of $x_r=0$ correspond exactly to the three solutions of $x_0=0$ in Fig. 1(b). For the magnetization of the disk to be sinusoidal [solid curve Fig. 1(e)] the magnetization of the rods must follow the solid curve of Fig. 1(b), but Pippard showed that rods cannot behave that way. In fact, because the part of the curves between the 0's in Fig. 1(b) is unstable, the corresponding part in Fig. 1(e) must also be unstable. Since that part of the curve cannot exist, there must be another state that has lower free energy.

A possible explanation of the instability is shown in Fig. 2. Suppose the sample has the uniform zero magnetization described by the solid curve at $x_0=0$ in Fig. 1(e). Now suppose there were some slight perturbation (a thermal fluctuation perhaps) that caused the region in the vicinity of A to have a slight positive magnetization [Fig. 2(a)]. The induction in the regions near B will be decreased by a small amount and those regions will become negatively magnetized. [Fig. 2(b)] These magnetizations will cause the induction in the regions A and C to be increased and will cause (further) inincrease of the magnetizations there [Fig. 2(c)]. This instability will grow and propagate if the feedback factor is greater than unity. The feedback factor is the

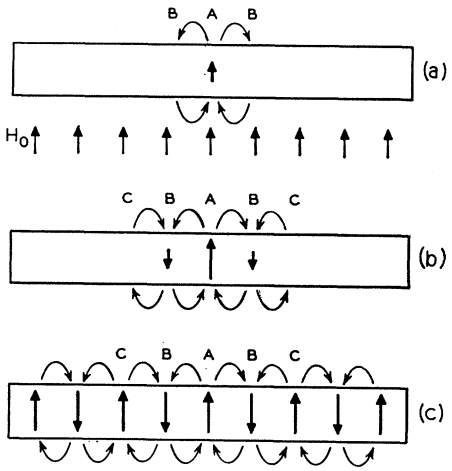

FIG. 2. A description of the growth and propagation of the domain structure that can occur when the differential susceptibility $d4\pi I/dB$ is greater than unity.

differential susceptibility, $d4\pi I/dB$. (The maximum value of $d4\pi I/dB$ is equal to the a factor.) This simple model suggests that there is a stable magnetic domain structure in the sample.

A magnetic domain structure is consistent with the behavior of the rod. Consider the disk to be made of rods as before but do not require their magnetization to be equal. Whenever x_r is an integral multiple of 2π [Fig. 1(g)], the rods may have either of two magnetizations, $\pm b$ [Fig. 1(b)], and so the average magnetization can be anywhere between $-b$ and $+b$ depending on the relative populations of the different magnetizations. The paths shown by the dashed lines in Figs. 1(e) and 1(g) are compatible with the behavior of the rod. The sample follows the usual curve being uniformly magnetized during part of the cycle (solid curve) and breaks up into narrow domains during the rest of the cycle (dashed line). The magnetic field intensity inside the sample is independent of the applied field when domains are present.

The domain structure has a lower free energy than the uniform magnetization has. The free energy is

$$G=-\int IdH_0 \quad \text{or} \quad g=-\int ydx_0=4\pi k^2 G. \qquad (4)$$

Figure 1(f) shows the free energies corresponding to the magnetizations shown in Fig. 1(e). The domain structure has a lower free energy and must be considered the correct description.

In fact, the domain structure has the lowest free energy of all descriptions. If the disk had a state with lower free energy, then the free energy of the rod at the crossing at $x_0=0$ [Fig. 1(c)] would have to be lower than shown, and the behavior of the rod would be different. If Pippard's solution for the rod is correct then the free energy of the lowest thermodynamic state of a disk is the free energy of the domain structure.

It has been argued by some that a large wall-energy term would prevent domain formation, but clearly a finite wall-energy term cannot prevent domain formation in arbitrarily large samples.

Sample shapes are rarely very rod like or disk like in the experiments, and it is important to consider the case of an ellipsoid with demagnetization factor L, $(0<L<4\pi)$. The solution is straight-forward following closely the treatment of a disk. The state without domains [solid line Fig. 1(i)] has distortion intermediate to the rod and the disk and the state with domains (dashed line) has slope $4\pi/L$. Even if the ellipsoid is long and thin, but finite, it is preferential for it to break up into domains.

This fact implies that there is a driving force to make the domains very narrow, but there is also a counteracting force. As soon as the size of the domains becomes comparable to the size of the real-space cyclotron orbit an appreciable number of electrons will average the magnetic induction by crossing domain walls, thereby raising the free energy. This increased free energy can be thought of as a domain wall energy.

There are several striking analogies with ferromagnetism: 1. The occurrence of domains when the differential susceptibility, $d(4\pi I)/dB$, exceeds unity; 2. the domain wall energy just explained; and 3. the fact that the relative populations of the different magnetizations may be changed by changing the applied field. However beryllium is not like iron; the source of the magnetization is the H-A effect, not spin alignment.

Although the domains have been described as rod like for the purposes of exposition, a structure of narrow plate shaped domains that contain the direction of magnetization has a lower wall energy.[7]

A. The Magnetization Oscillations

In this section and the next, the results of the experiments will be derived from the theory assumming that there are no supercooling effects and that the domain wall energy term is small. (To assume otherwise leads to disagreement with the experiment. See Sec. IV.)

The peak amplitude of the magnetization oscillations is independent of the sample shape as can be seen by comparing Figs. 1(b), 1(e), and 1(c). The amplitude b is a function of a and is shown in Fig. 3(a). Since there are two beating periods in beryllium, the a factor is a function of field like that shown in Fig. 3(b). The resultant envelope shape [Fig. 3(c)] is what is expected for any ellipsoidal sample shape. If the sample is rod shaped, then $\sim$34 saw-tooth-shaped oscillations [as in Fig. 1(b)] are expected between the nodes of the envelopes; if the sample is a disk, then $\sim$34 triangular-shaped oscillations [Fig. 1(e)] are expected.

B. The Magnetothermal Oscillations

The entropy density of an isothermal sample is $S=-\partial G/\partial T$. If the condition is adiabatic, the oscillating temperature is

$$\Delta T_{\rm osc}=\frac{T}{C}(\partial G/\partial T)_{\rm osc}, \quad (\Delta T_{\rm osc}\ll T), \qquad (3)$$

where C is the specific heat. G depends on the temperature only through the amplitude of the H-A oscillations,

$$\frac{\partial G}{\partial T}=\frac{1}{4\pi k^2}\frac{dg}{da}\frac{da}{dT}, \qquad (6)$$

where

$$a\propto Z/\sinh(Z),$$

and

$$Z\equiv\frac{2\pi^2 c m_0 k_B}{eh}\frac{m_c}{m_0}\frac{T}{B},$$

so that

$$\Delta T_{\rm osc}=-\frac{\pi c m_0 k_B}{2ehB}\frac{1}{\gamma k^2}\frac{m_c}{m_0}L(Z)a\frac{dg}{da}, \qquad (7)$$

[7] L. Landau and E. Lifshitz, Physik. Z. Sowjetunion 8, 153 (1935).

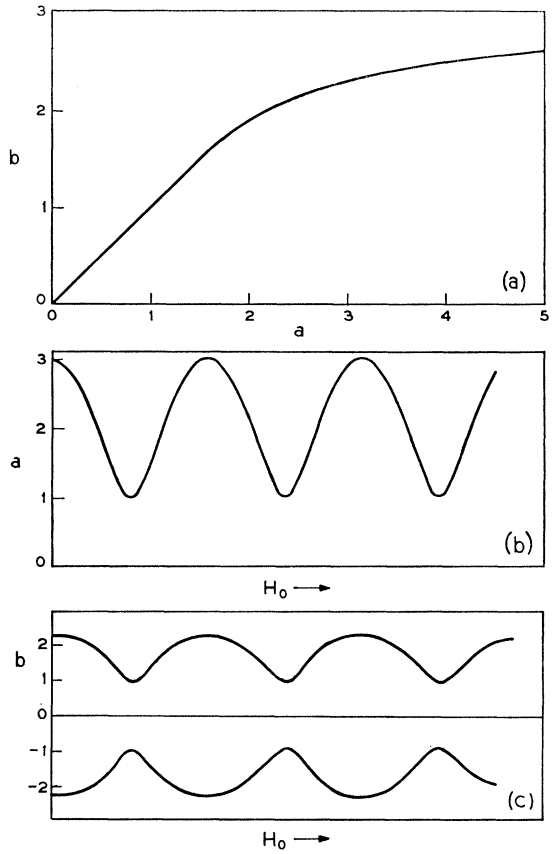

FIG. 3. (a) The peak amplitude of the magnetization oscillations b as a function of Pippard's a factor; (b) behavior of Pippard's a factor when there are two beating H-A periods; (c) the envelope shape of the magnetization oscillations according to the theory. Compare with Figs. 6(a), 6(c), and 11(a).

where m_c is the cyclotron mass of the H-A period that has local angular frequency k, γT is the electronic heat capacity per unit volume, and $L(Z)$ is the function coth $(Z)-Z^{-1}$. The maxima and minima of $g(a)$ are not sample-shape-dependent as can be seen by comparing Figs. 1(c), 1(f), and 1(j). The maxima and minima of $g(a)$ are shown in Fig. 4(a) and the maxima and minima of a dg/da are shown in Fig. 4(b). An example of an expected magnetothermal oscillation envelope shape is shown in Fig. 4(d).

The wave shapes of the magnetothermal oscillations are also given by Eq. 7, but their derivation is complicated and only the results are presented, Fig. 5.

Finally, the "nodes" of magnetothermal oscillations occur at the same field as the nodes of the magnetization oscillations.

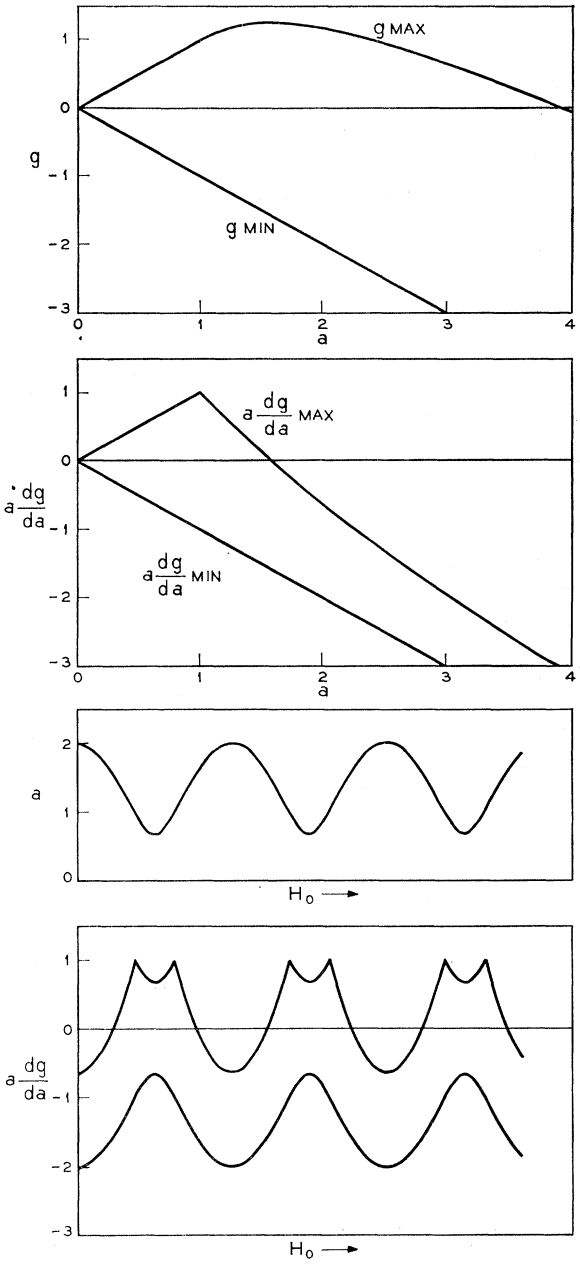

FIG. 4. (a) The maximum and minimum values of the Gibbs function g as a function of Pippard's a factor. For $a>1$ the maximum occurs at the crossing in Fig. 1(c); (b) the maximum and minimum function $a\ dg/da$ that is required in Eq. 7 of the text; (c) a schematic representation of the behavior of Pippard's a factor when there are two beating H-A oscillations; (d) the envelope shape of the magnetothermal oscillations according to the theory in the text. Compare with Figs. 10(c) and 11.

III. THE EXPERIMENTS

In this section the experiments carried out by the author and by others are described and the experimental results are compared to the theory presented in the previous section. The agreement is very good: (1) All the experiments showed reversibility, which is in agreement with the theory. (2) The envelope shapes of both the magnetization and the magnetothermal oscillations agree with those predicted by the theory. (3) The wave shapes of the magnetization oscillations are also in good agreement with prediction (no comparison could be made for magnetothermal oscillations). (4) The measured absolute amplitudes are in fair agreement with calculation. (5) The theory does not predict the difference frequency that Plummer and Gordon[5]

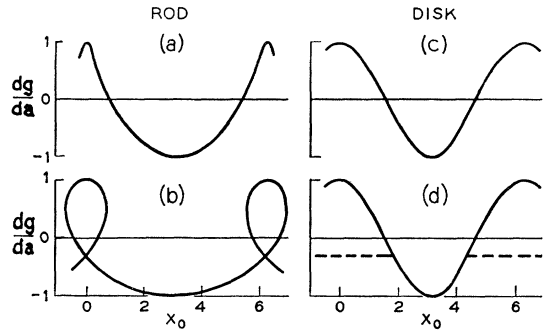

FIG. 5. The waveshape of the magnetothermal oscillations predicted by Eqs. 7(a) and (b) are for a rod-shaped sample, (c) and (d) are for a disk-shaped sample. The curves are drawn for Pippard's a factor $=0.8$ (a) and (c) and 2.0 (b) and (d). Only the lower branches of (b) and (d) are expected to occur. The upper branches correspond to the nonequilibrium thermodynamic solutions.

report. But since the author could not confirm that result, he believes that their result must be due to their experimental method 6. An experimental search for the domains was made and had negative results. Because of the technique, a negative result would be expected if the domains are smaller than ~ 0.5 mm, and that is the conclusion made from this experiment.

In Sec. IV other theories will be discussed and compared to these experimental results. It will be concluded that, even though the domains have not been observed by any direct method, the circumstantial evidence for their existence is very strong.

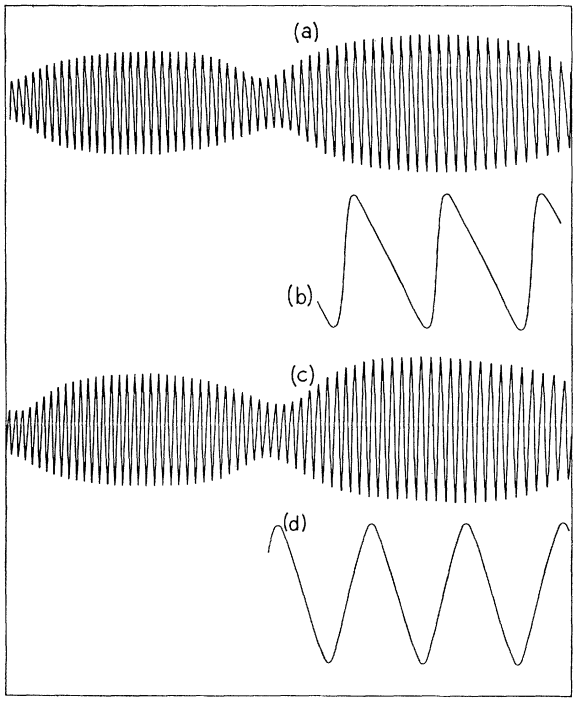

FIG. 6. (a) The H-A oscillations of the torque of the rod shaped sample at 1.4°K and $24.9 \leq H \leq 30.0$ K G when H is 5° away from the hexagonal axis; (b) an expanded plot showing the wave shape of the three oscillations at the second antinode of (a); (c) the H-A oscillations of the disk shaped sample with experimental conditions identical with (a) above; (d) expanded plot of the torque of the disk shaped sample similar to (c).

A. Torsion Balance Experiments

A torsion balance similar to one previously described[8] was calibrated and used to study the H-A effect of both a rod-shaped ($0.7 \times 0.7 \times 2.0$ mm along c, mass $=1.05$ mg) and a disk-shaped ($2.0 \times 2.0 \times 0.17$ mm, mass $=0.80$ mg) sample of beryllium. The samples were shaped by spark cutting and chemical etching and showed low ($\sim 0.1°$) crystalline microstructure in the back reflection Laue pictures used to orient the samples. The torque (Fig. 6) showed the reversibility, wave shape, and envelope shape that the theory gives. [Compare the envelope of Fig. 6(a) and (c) with Fig. 3(c), compare Fig. 6(b) with Fig. 1(b) and Fig. 6(d) with Fig. 1(e).] The calibration of the balance and knowledge of direction of magnetization (See Appendix I) were used to determine the average magnetization intensity of the samples. The results for the rod are shown in Fig. 7 (the results for the disk are nearly identical and are not presented). Also shown in Fig. 7 is the amplitude for which the a factor is unity and it can be seen that the a factor of the results is smaller than the wave-shape distortion and envelope shape would indicate. There are two probable explanations; the harmonic content of the H-A effect could be such that the sinusoidal wave shape of Eq. 1 is not adequate (such a situation is discussed in Appendix I) or the crystalline microstructure could be so large that the contributions from different parts of the sample partially cancel. This latter effect was shown in a larger sample of irregular shape and mass $=39.6$ mg. The wave shape of the torque oscillations were very complicated but were completely reversible. The envelope shape corresponds to the theory, but the amplitude (Fig. 8) was

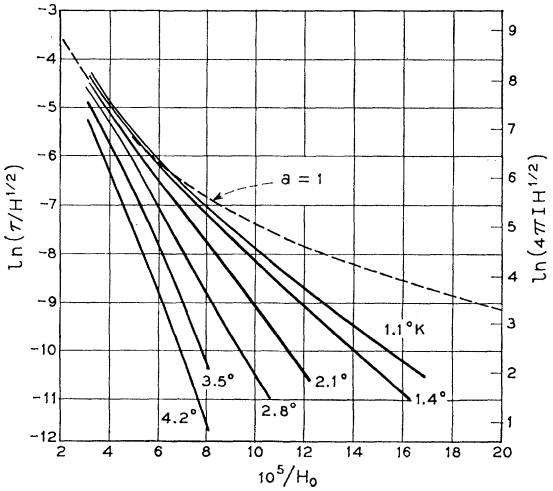

FIG. 7. The amplitude of the torque with the applied field 5° from the $\hat{c}$ axis and the average magnetization of the rod shaped sample. The curve marked $a=1$ represents the amplitude for which Pippard's a factor is unity. (All units are in the cgs-emu system.)

[8] J. H. Condon and J. A. Marcus, Phys. Rev. A134, 446 (1964).

always smaller than $a=1$. Wave-shape distortion (presumably due to the B-H effect) was present when the experimental a factor was ~ 0.1. The x-ray diffraction pictures of this sample showed a microstructure of $\sim 0.5°$ and this microstructure explains the smallness and very long-period oscillations of the amplitude.

B. The Induction Experiments

Since the previous experiments did not confirm one of Plummer and Gordon's results[5] (See D below) and the absolute measurements presented above depended on the knowledge of the direction of the magnetization, a separate set of experiments was carried out using a quite different method. The results agreed with the previous results and with the theory of Sec. II.

A rod ($1.0\times 1.5\times 9.5$ mm along $\hat{c}$) was prepared by spark cutting and chemical etching and ~ 200 turns of fine (AWG49) insulated copper wire were closely wound on to the central section of it. This coil and a larger coil which sensed the applied field were connected to two electronic integrators, the outputs of which were displayed on the y and x axes of an oscilloscope as B in the sample and H applied, respectively. The results (Fig. 9) showed reversibility and showed that the magnetization was saw-tooth shaped [as in Fig. 1(b)]. This method provides the sensitivity calibration simultaneously with the data; the peak to peak to peak amplitude of the oscillations of $4\pi I$ is 27 G $\pm 10\%$ (compared to 30 G from the torsion balance experiments on a different sample at the same temperature and field) and the sharp step is shown to be paramagnetic as Pippard's treatment predicts, [Fig. 1(b)]. It was difficult to determine the envelope shape because of the large base-line shift inherent in the method.

In order to double check the envelope shape, a coil

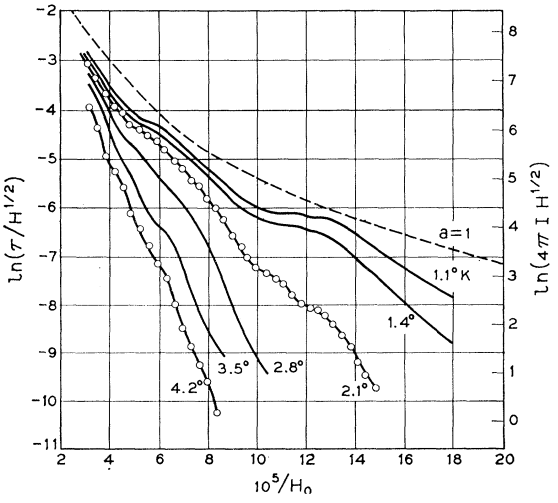

FIG. 8. This plot is similar to Fig. 7 except that it is of the results from a large irregularly shaped sample that showed 0.5° crystalline microstructure in the x-ray studies. The open circles are representative data points; the other data points of this plot and those of the plot in Fig. 7 have been omitted for clarity.

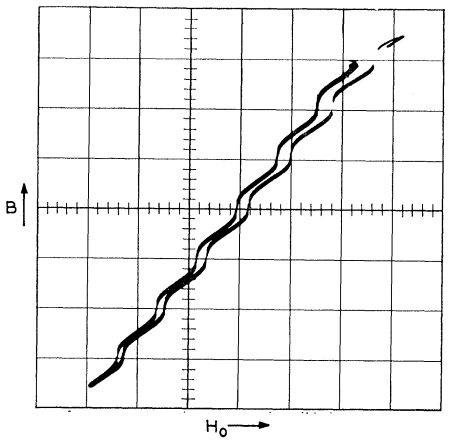

FIG. 9. B versus H in beryllium at 1.3° and a mean field of 28.25 k G. The applied field was first swept downward and then upward. The lack of retrace is due entirely to the drift in the poor quality integrator for H_0. The sweep range is ~ 0.5 K G.

was wound on to a much larger sample[9] (~ 6.0 mm cube) and the output of it was combined with the output of the applied-field sensing coil in such a way that only the single dM/dt was integrated and displayed on the oscilloscope. The envelope shape did correspond to theory. Experimental difficulties preclude any statement about wave shape, reversibility, or absolute magnitude.

C. The Magnetoresistive Probe Experiments

A commercial magnetoresistance probe[10] was used to map the field distribution in the vicinity of a large flat plate ($10.\times 15.\times 0.5$ mm) of beryllium in order to search for the domains predicted by the theory. The results were quite noisy (~ 5 G p-p) but were completely consistent with a uniform magnetization of the triangular wave shape predicted by the theory. Since the sensitive area of the probe was 0.4×1.0 mm and spaced away from the sample by 0.5 mm, the result can be interpreted as setting an upper limit of ~ 0.5 mm on the size of the domains. This experiment showed the reversibility, absolute amplitude, and envelope shape that the previous experiments showed but with less accuracy.

The same magnetoresistance probe was mounted on the end of a rod-shaped sample of beryllium and the results triple checked the previous results, reversibility, envelope shape, wave shape, and absolute amplitude.

D. Plummer and Gordon's Experiments

Plummer and Gordon[5] have studied the magnetization of rod- and disk-shaped samples by a modified mutual-inductance technique. Their results show the saw-tooth and triangular wave shape predicted by the theory. They do not report absolute amplitudes. Their

[9] This sample was one of those used in the magnetothermal oscillation experiments.

[10] Purchased from American Aerospace Corporation, Farmingdale, New York.

results are reversible, in agreement with the theory of Sec. II. However, they report the observation of a "difference frequency."

This difference frequency may be interpreted as a distortion of the envelope shape. Since no such effect was seen in the author's experiments or is predicted by the theory in its present form, it is presumed that their result is dependent on their technique, i.e., a moderately rapidly alternating magnetic field. The author believes that their result is due to details of the dynamics of domain-wall motion, but since very little is known about the statics of domains at present, the problem of dynamics is unsolvable.

E. The Magnetothermal Oscillation Experiments

The magnetothermal oscillations are the temperature oscillations of an adiabatically isolated sample as the magnetic field strength or direction is changed.[11] Studies in Be have been carried out by Le Page, Garber, and Blatt[12] and by Halloran and Hsu.[1] Discussion will be limited to the latter's results because they were more readily available to the author. Only those results that are pertinent to the point of this paper will be discussed.

The results of the magnetothermal experiments on a $\sim$6 mm cube of Be at four different average temperatures are shown in Fig. 10. The envelope shape of the data at 2.1°K agrees very well with Fig. 4(d), (the other envelope shapes in Fig. 10 are explainable by chosing different ranges of a factors). This sample was known to have $\sim$0.5° crystalline microstructure and so it is understandable that the absolute peak to peak temperature oscillations observed experimentally were smaller than those calculated from Eq. 7 using $m_c=0.17$ m_0[13] $\gamma=0.17$ mJ/deg^2—mole[14] and the H-A period, $P=1.02\times10^{-7}G^{-1}$, (see Table I).

The wave shape of magnetothermal effect was studied in rod- and disk-shaped samples, but because the results were not reproducible for similarly shaped samples (presumably because of microstructure) no comparison can be made to the theory. In all the magnetothermal experiments, the results were reversible in agreement with the theory.

Finally, the magnetothermal oscillations and magnetization oscillations have been studied over the same field range (Fig. 11), and although the experimental accuracy is not good enough to confirm the phase relationship of the fast oscillations, it is good enough to show that the "nodes" are in phase as predicted.

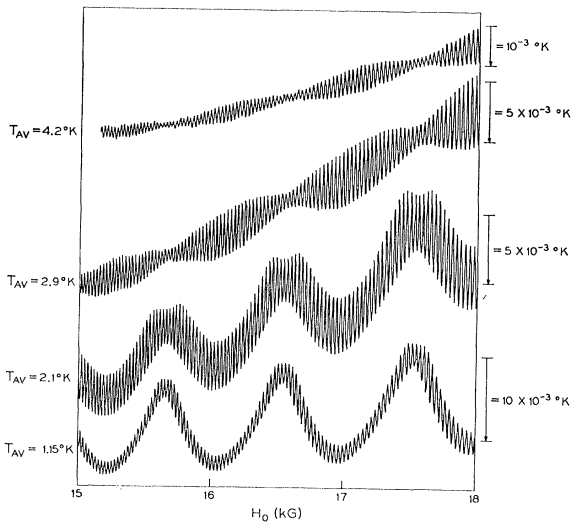

FIG. 10. The magnetothermal oscillations of beryllium at four different average temperatures. The upward slope of the curves is due to the magnetoresistance of the carbon thermometer and slow temperature drift of the liquid-He bath during the experiment.

IV. OTHER POSSIBLE CAUSES

In this section other theories will be discussed and compared with the experimental results. No other theory fits nearly as well as that of domains.

A. Supercooling

Pippard[4] proposed that supercooling could occur in Shoenberg's[3] impulsive-field-technique experiments. That is quite likely to be the case where the time of supercooling need be only $\sim10^{-6}$—10^{-7} sec, but the author's experiments were carried out $\sim10^5$ to 10^8 times more slowly and it seems quite unlikely that Pippard's eddy-current stabilization could be effective over such long times. If there were supercooling: 1. Hysteresis would be predicted and no such hysteresis is observed. 2. The magnetization of a disk would take a rapid irreversible jump from the open circles [of Fig. 1(e)] to the dashed line of the domain structure; the experimental results definitely rule this out. 3. It would not be possible for Plummer and Gordon to see the paramagnetic spikes that they do by the method they use. 4. There would be a gradual irreversible "loss" of free energy of the system, which, if even only 5% of that energy went into heating the sample, leads to disagreement with the experimental results.

B. No Domains

If the arguments of Sec. II were ignored or it was assumed that the domain-wall-energy term was large enough to prevent domain formation in the sample size studied then there would be other difficulties. 1. The envelope shapes of the magnetization and magnetothermal oscillations of disk-shaped samples would be those shown by the dashed lines in Fig. 11. 2. The wave shape of the magnetization oscillations of a disk would

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[11] J. E. Kunzler, F. S. L. Hsu and W. S. Boyle, Phys. Rev. 128, 1084 (1962).

[12] J. LePage, M. Garber and J. F. Blatt, *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt, D. V. Edwards, F. N. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965).

[13] $m_c=0.17$ m_0 was obtained from the temperature dependence of the H-A effect and also by Azbel-Kaner cyclotron resonance, W. M. Walsh (private communication).

[14] G. Ahlers, Phys. Rev. (to be published).

be sinusoidal not triangular as the experiments showed. 3. Finally, this theory is self-contradictory as was pointed out in Sec. II.

C. Oscillating Magnetostriction

It is possible that there is some "feed back" mechanism other than the B-H effect that could cause similar behavior. This subsection and the next two discuss such cases. Even though it is clear at the outset that the sample shape dependence of the results cannot be explained unless there is a shape-dependent parameter in the theory, and there is none, it will be shown that other feedback mechanisms have little importance.

The magneto-strictive strain ϵ of sample is $\epsilon \backsimeq -S(\partial\Omega/\partial\epsilon)$, where Ω is the free energy per unit volume and S is the compliance of the sample. The major contribution of the oscillating part is due primarily to the change of extremal cross sections of the Fermi surface with change of strain, i.e., $\partial\Omega/\partial\epsilon=\Omega'(cS_m/e\hbar B)(\partial \ln S_m/\partial\epsilon)$, where Ω' is the derivative $\partial\Omega/\partial(cS_m/e\hbar B)$ and S_m is the extremal area. But the magnetization intensity is given by $I=\Omega'(cS_m/e\hbar B)1/B$ and so

$$\epsilon=-SB(\partial \ln S_m/\partial\epsilon)I.$$

This result is quite general. The symbols ϵ and $\partial/\partial\epsilon$ are in reality six-component vectors and S is a 6×6 matrix.

It is possible that there is magnetostrictive feedback similar to the magnetic feedback, i.e.,

$$\epsilon=\epsilon_0 S_m\left(\frac{cS_m}{e\hbar B}\left(1+\frac{\partial \ln S_m}{\partial\epsilon}\epsilon\right)\right)$$

or

$$y=a_{MS}\sin(x+y),$$

where

$$a_{MS}=\frac{cS_m}{e\hbar B}\frac{d\ln S_m}{d\epsilon}.$$

The ratio of relative importance of a_{MS} to the magnetic a factor can be reduced to $a_{MS}/a=S^{-1}\epsilon_0^2/4\pi I_0^2$. Chandrasekhar, Fawcett, and White[15] have measured ϵ_0 to be about 10^{-7} and so a_{MS}/a is found to be $\sim1/1000$ and a_{MS} cannot be directly responsible for the effects seen.

The internal stresses in a sample with magnetic domain structure will probably make a significant contribution to the domain-wall energy.

TABLE I. Sample comparisons of calculated and observed amplitudes of the magnetothermal oscillations.

T_{av}(°K)	$H_0(kG)$	ΔT_{calc}(°K)[a]	ΔT_{obs}(°K)
2.1	17.6	10.5×10^{-3}	5.8×10^{-3}[b]
1.15	17.6	7.3×10^{-3}	5.0×10^{-3}[b]
2.1	29.7	37×10^{-3}	18×10^{-3}[c]

[a] Calculated from Eq. 7 using $(a\,dg/da)_{p-p}=2$.
[b] From Fig. 10.
[c] From Fig. 11.

[15] B. S. Chandrasekhar, E. Fawcett and G. White (private communication).

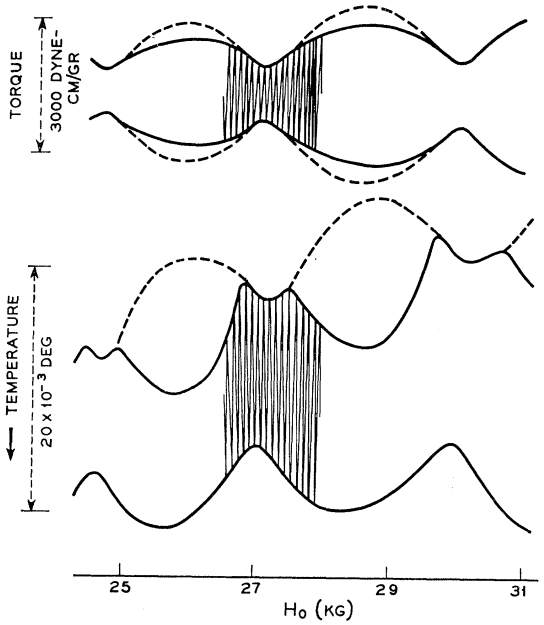

FIG. 11. The torque and magnetothermal oscillations of two different samples at different temperatures but at the same orientation of magnetic field relative to the crystal and over the same field range. The dashed lines show the behavior expected from two beating periods. This figure shows the phase relationship between the envelopes of the magnetization oscillations and the temperature oscillations. There is an error due to the photographic service: the plot for the torque should be expanded slightly in the horizontal direction. The dashed lines show the behavior expected if there were no magnetic feedback effect.

D. Oscillating Fermi Energy

It is possible for the Fermi energy to oscillate causing periodic changes in the extremal areas and thereby causing effects similar to the B-H effect. However, a simple calculation of the ratio of strength of this effect to that of the B-H effect yields $H^2(dS_m/dE)^2/4\pi\rho(E_F)S_m^2$. Using values for Be this ratio is about 1/300.

E. Oscillating Band Structure

Kittel[16] has shown that the electronic band structure may be changed by an inhomogeneous magnetic field within the real-space cell. This magnetic-field inhomogeneity has the same periodicity as the real lattice. It might be possible, therefore, that the band structure oscillates with the H-A period and for oscillating extremal areas to be produced in this way. The upper limit of the importance of this effect relative to the B-H effect has been estimated to be the inverse of the Landau level number, or $\sim1/300$.

V. CONCLUSIONS

From theoretical considerations that involve only magnetism and thermodynamics it has been shown that magnetic domains will occur whenever $d4\pi I/dB$ exceeds unity. The proof does not depend at all on the origin of the magnetization but only on the fact that

[16] C. Kittel, Phys. Rev. Letters **10**, 339 (1963).

equation of state has a region where $d4\pi I/dB>1$. The situation is very similar to ferromagnetic domains.

The theory of magnetothermal oscillations and the effect of Shoenberg's B-H effect on them has been presented. A theory of the oscillatory magnetostrictive effect has also been given.

The experimental work has shown that conditions are correct for domain formation in beryllium. The experiments that can discriminate between various plausible theories always have results that agree with the domain theory. Even though there is at present no direct evidence for domains, the circumstantial evidence is very strong.

ACKNOWLEDGMENTS

I thank several people: M. H. Halloren W. A. Reed, L. F. Mattheiss, and P. Soven for many helpful discussions. In particular, I thank E. I. Blount for correcing a serious error in an earlier argument, M. H. Halloren and F. S. L. Hsu for making available their experimental results, and A. Greenfield for making recommendations in the presentation of the material.

APPENDIX I

In the main text it was stated that the direction of magnetization was along the hexagonal axis.

The same 6 mm cube sample used in the experiment of Sec. IIIB also had coils wound on to it in the $(10\bar{1}0)$ $(1\bar{2}10)$ planes. The outputs of these coils and that of the (0001) coil were measured and it was concluded that whenever the applied field was within 10° of being parallel to the hexagonal axis, the direction of magnetization was directly along that axis within experimental error ($\lesssim 1°$). The same result is expected on theoretical grounds. The free energy is an oscillating function such as $F=A\cos(2\pi/PB)$, where P is the period of the oscillations. The period of H-A oscillations due to a cylindrical segment of the Fermi surface vary as $P=P_0\cos\theta$, where θ is the angle between field direction and the cylinder axis. Now,

$$F=A\cos(2\pi/P_0B\cos\theta)=A\cos(2\pi/P_0B_c),$$

where B_c is the component of field along the cylinder axis. The magnetization is given by the derivatives of the free energy $M_i=-\partial F/\partial B_i$, and since the free energy depends only on the component of field along the cylinder axis, the magnetization must be along that axis.

The same method can be generalized. The thermodynamic potential per unit volume can be approximated by

$$\frac{1}{V}\Omega_{L+K}(B)=\frac{1}{V}\sum_{l=1}A_l(B,\theta)\cos\left(l\frac{C(\theta)}{B}+\psi_l(\theta)\right). \quad \text{(AI.1)}$$

The function $C(\theta)$ describes the dependence of extremal cross-sectional area upon magnetic field direction, and $A_l(B,\theta)$ and $\psi_l(\theta)$ describe the very slow variation of wave shape and amplitude with magnetic field. Expansion of Ω about some field $\mathbf{B}_0$ yields

$$(1/V)\Omega_{L+K}(B)\simeq(1/V)\Omega_{L+K}(\mathbf{B}_0)+\omega(\mathbf{k}\cdot\nabla\mathbf{B}), \quad \text{(AI.2)}$$

where the function ω is periodic in 2π. It will be shown in Appendix II that,

$$4\pi\mathbf{I}=(4\pi/V)\nabla_{\mathbf{B}}\Omega_{L+K}(B) \quad \text{(AI.3)}$$
$$=4\pi\mathbf{k}\omega'(\mathbf{k}\cdot\mathbf{B})$$

(the Δ before B has been dropped for convenience). Equation AI.3 shows that the oscillating magnetization from one H-A period is a fixed direction times an oscillating scalar function.

For an arbitrarily shaped ellipsoidal sample the magnetic induction inside the sample is

$$\mathbf{B}=\mathbf{H}_0+4\pi(\mathbf{I}-\mathbf{L})\cdot\mathbf{I}, \quad \text{(AI.4)}$$

where H_0 is the applied field, so that

$$4\pi kI=4\pi k^2\omega'\{\mathbf{k}\cdot\mathbf{H}_0+[\hat{\mathbf{k}}\cdot(\mathbf{I}-\mathbf{L})\cdot\hat{\mathbf{k}}]4\pi kI\}. \quad \text{(AI.5)}$$

This equation is very similar to Pippard's and may also be multiple valued at certain values of H_0. The multiple valued solutions occur if the tensor $\nabla_B 4\pi\mathbf{I}$ has an eigenvalue larger than unity and the sample is thin in a direction perpendicular to the corresponding eigenvector. If the sample is a different shape, domains that are thin perpendicular to that eigenvector will form causing a lowering of the free energy.

If the wave shape of the oscillations given by Eq. AI.3 was already sawtooth shaped because of the contributions of higher harmonics before consideration of the B-H effect, then the amplitude limiting and wave-shape distortion will be greater than predicted by Pippard's treatment which assumes sinusoidal wave-shape. The experiments show this greater amplitude limiting and wave-shape distortion.

If there are two H-A periods whose magnetizations are not parallel to each other or to the applied magnetic field, the situation is complicated. Multiple valued solutions and domain formation will occur if $\nabla_B 4\pi\mathbf{I}$ has an eigenvalue greater than unity. The domains will be plate-like in shape and will contain the eigenvector of $\nabla_B 4\pi\mathbf{I}$ that has the largest eigenvalue.

The component of the monotonic magnetization parallel to the applied field will not be changed by the domain formation; however, other components can be changed. Likewise, long-period Fourier components of the component of magnetization parallel to the field will not be changed by the domain formation, but it is possible that difference frequency oscillations of the magnetization will occur perpendicular to the applied field direction.

APPENDIX II

Presented here is another proof of the statement $M=M(B)$. It is included because it takes a different form than Pippard's argument[4] although it yields the same conclusion, and because it points out more clearly one possible difficulty in that treatment, i.e., the argument through the entropy density may have the diffi-

culty that the distribution of energy levels in the dilute and dense systems can be different.

Kittel[16] considered the problem of magnetic interaction in terms of Lorentz' single magnetic field $\mathbf{h}$. He argued that $\mathbf{h}$ could be expressed as the sum of two terms: $\mathbf{B}$, the uniform magnetic induction in the sample, and $\mathbf{h}_{osc}$, a field that is periodic with the lattice and that averages to zero in one real-space cell. Since $\mathbf{h}_{osc}$ is periodic, its effect may be included in the original Bloch Hamiltonian; and Kittel shows its effect to be negligible.

The effect of B is known; the momenta P_i of the Hamiltonian must be replaced by P_i defined such that $[P_i,P_j]=B_k$. Everything in Lifshitz and Kosevich's work[17] goes back to their Eq. 1.2, i.e., $[P_i,P_j]=H_k$, so in all the rest of their paper B should appear rather than H.

The thermodynamic potential, Ω_{L+K}, that Lifshitz and Kosevich[17] calculate is nearly the Helmholtz function of the "system."[18] (The "system" is magnet, sample and interaction between them, minus the magnet alone.) However, it is clear from considerations of the internal energy that the interaction energy $\mathbf{M}\cdot\mathbf{H}_0$ must be added to their result, i.e., $A=\Omega_{L+K}+\mathbf{M}\cdot\mathbf{H}_0$. The Gibbs function of the system differs from the Helmholtz function by $\mathbf{M}\cdot\mathbf{H}_0$ and is then $G=\Omega_{L+K}(B)$. But one other effect has been neglected, that of the interactions of the individual magnetic moments. An additional internal energy (and therefore Gibbs function) term ΔG due to individual interactions between states must be subtracted, since it has been doubly added already,

$$\Delta G=\sum_{i<j}\epsilon_{ij}f(\epsilon_i)f(\epsilon_j), \qquad \text{(AII.1)}$$

where $f(\epsilon_i)$ is the Fermi function of the energy of the ith state. If the interaction is taken to be magnetic dipole in nature, then ΔG may be expressed as

$$\Delta G=-\tfrac{1}{2}\sum\mu_i\mu_j g_{ij}f(\epsilon_i)f(\epsilon_j), \qquad \text{(AII.2)}$$

where g_{ij} is the geometrical factor of the coupling of the dipoles μ_i and μ_j. The sum $\sum\mu_i g_{ij}f(\epsilon_i)$ is the induction in the sample due to the moments μ_i and so the double sum becomes

$$\Delta G=-2\pi\mathbf{I}\cdot(\mathbf{I}-\mathbf{L})\cdot\mathbf{I}V, \qquad \text{(AII.3)}$$

where $\mathbf{L}$ is the demagnetization factor and V is the volume of the sample. Now subtracting this from the previous result, the Gibbs function of the system is given by,

$$G=\Omega_{L+K}(B)+2\pi\mathbf{I}\cdot(\mathbf{I}-\mathbf{L})\cdot\mathbf{I}V. \qquad \text{(AII.4)}$$

Using $\mathbf{M}=-\nabla_{H0}G$, the result $\mathbf{M}=-\nabla_B\Omega_{L+K}(B)=\mathbf{M}_{L+K}(B)$ is obtained. Now it is apparent that this argument for which I am indebted to P. Soven gives the same result as Pippard's. However, if the energies [as in Eq. (AII.2)] coupling individual states must be considered, as it is apparent they must be, it then follows that the "states" are not really the eigen-solutions to the complete Hamiltonian of the problem. The calculation of the thermodynamic potential as done by Lifshitz and Kosevich is appropriate only when the states are eigen-states. The simple subtracting of ΔG is therefore in doubt. The coupling suggests that a renormalization might be necessary in the exact solution.

Note added in proof. Since this paper was submitted, Plummer and Gordon[19] have shown that the modified mutual inductance method can produce difference frequency oscillations, and so there is no longer a discrepancy between their experimental results and those of Sec. III A-C.

Broshar, McCombe, and Siedel[20] have published experimental results similar to those presented here and also presented earlier.[1,21] However, because they ignore the possibility of magnetic domain formation, their conclusion that there is an electron-electron interaction is in error.

Their conclusion appears to be based on three points: (1) There was no observation of the infinite differential magnetic susceptibility that they expected; (2) The temperature and field dependence of the amplitude was not explainable; and (3) The envelope shape of the magnetothermal oscillations of a disk-shaped sample was not what they expected.

Infinite differential susceptibility is not expected for the sample shape they use and furthermore there is reason to believe that their sample has $\sim 0.5°$ crystalline substructure.[22]

The unusual temperature and field dependence of the amplitude is explainable in terms of the harmonic content and magnetic domains.

Their confirmation of the earlier [1] determination of the magnetothermal oscillations of disk-shaped samples is pleasing. It was this earlier result that convinced the author of the validity of the idea of magnetic domains,[2] i.e., the comparison of Figs. 4(d) and 10(d) and 11(b).

If Broshar, McCombe, and Siedel's hypothesis of "broadened" Landau levels was correct, then the sharp steps in Fig. 9 could not occur.

M. H. Halloran[23] has found very large amplitude difference frequency magnetothermal oscillations in silver. This and the data of Shoenberg[3] and Joseph and Thorsen[24] indicate that magnetic domains must exist in silver and gold also.

[17] I. M. Lifshitz and A. M. Kosevich, Zh. Eksperim. i Teor. Phys. **29**, 730 (1955) [English transl.: Soviet Phys.—JETP **2**, 636 (1956)].

[18] Cf., D. Ter Haar, *Elements of Statistical Mechanics* (Holt, Rinehart, and Winston, Inc., New York, 1954).

[19] R. D. Plummer and W. L. Gordon, Phys. Letters, April 1966.

[20] W. Broshar, B. McCombe, and G. Siedel, Phys. Rev. Letters **16**, 235 (1966).

[21] J. LePage, M. Garber and F. J. Blatt, *Low Temperature Physics LT9* (Plenum Press, New York, 1965), p. 799.

[22] X-ray diffraction studies (L. D. Fullerton, Bell Telephone Laboratories) show that a sample cut from the same boule as the one used by Broshar *et al.* (Dr. D. Kaufman, Nuclear Metals Division, Concord, Massachusetts) has from 0.5 to 1.0° microstructure throughout.

[23] M. H. Halloran, private communication.

[24] A. S. Joseph and A. C. Thorsen, Phys. Rev. **138**, A1159 (1965); Phys. Rev. **140**, A2046 (1965).