Theory of Landau Orbital Ferromagnetic States for Electrons in a Metal*

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A theory of the Landau orbital ferromagnetic states for the electrons in metals is presented. The conditions under which such states are realizable in a metal and are observable in a laboratory are presented and analyzed.

I. INTRODUCTION

Recently, in order to explore the origin of an extremely strong magnetic field present in dense astrophysical matter (such as white dwarfs and neutron stars or pulsars), Canuto, Chiu, Chiuderi, and the author¹ introduced a new mechanism that ascribes a ferromagnetic state for a system consisting of high-density electrons. Such a magnetized state is named¹ the LOFER state (Landau orbital ferromagnetic state). It was shown² that the LOFER state alone can furnish the extremely intense magnetic field believed to be present in such compact stars.³ It is then intriguing to find out whether or not one can realize the LOFER state in a metal and thus observe it in a laboratory.

In this article, we present the theory of LOFER states for electrons in a metal and discuss the conditions under which a LOFER state can be observable in a laboratory. We do not indicate any specific metal which might be favorable for the laboratory observations of the LOFER state. We merely present the conditions or the guiding rules which enable one to look for a specific metal suitable for this observation. In view of the low temperatures that can be attained in a laboratory at present and the purity of metallic samples one can obtain these days, the LOFER state is at present unlikely to be observed in a metal in a laboratory. Much of the possibility of observing such a state in a laboratory seems to depend on the future development of techniques for obtaining pure metals, low temperatures, and high pressures.

In Sec. II we discuss how the LOFER states arise in a metal and in Sec. III we discuss the stability of such states. We show that the LOFER state is quasistable in the sense that, once a system is in such a state, it will stay in the state for any forseeable future. In Sec. IV we present the conditions under which the LOFER states are realizable and observable in a metal. We discuss and analyze some methods to achieve the favorable conditions for the observations of such a state in a laboratory. Finally, two appendices are added to give definitions, to show relations among some relevant thermodynamic functions, and to give an estimation for the energy barrier that is needed to discuss the stability of the LOFER state.

II. LOFER STATES OF ELECTRONS IN A METAL

It is well known⁴ that the electrons in a metal are well described by a model in which they are treated as a

system of free, charged fermions of spin $\frac{1}{2}$, satisfying an empirically determined dispersion law of

$$\mathcal{E}(\mathbf{p}) = \mathcal{E}(p_x, p_y, p_z). \tag{1}$$

In the presence of a dc magnetic field H, the motion of these electrons is governed by the Hamiltonian obtained from Eq. (1) by adding the Zeeman energy term $\mu_B \mathbf{\sigma} \cdot \mathbf{H}$ (where $\mu_B = e\hbar/2mc = -0.578817 \times 10^{-8} \text{ eV G}^{-1}$ is the Bohr magneton and σ is the Pauli spin operator) and by replacing the momentum operator p with the operator $\hat{\boldsymbol{\pi}} = \mathbf{p} - (e/c)\mathbf{A}$ (where $\mathbf{H} = \text{curl}\mathbf{A}$) to satisfy the requirement of invariance under gauge transformations. The charge e appearing in the above is the same as that of a bare electron.5 (For convenience, the charge e is understood hereafter to denote its absolute value $e \mid .)$ When the z axis is chosen to coincide with the direction of the uniform magnetic field H, the vector potential **A** can be written as $\mathbf{A} = (-Hy, 0, 0)$. Then, the operators $(c/eH)^{1/2}\hat{\pi}_y$ and $(c/eH)^{1/2}\hat{\pi}_x$ are canonically conjugate:

$$\lceil (c/eH)^{1/2}\hat{\boldsymbol{\pi}}_x, (c/eH)^{1/2}\hat{\boldsymbol{\pi}}_y \rceil = i\hbar. \tag{2}$$

Hence, from the semiclassical quantization rule, one obtains 6

$$\mathcal{J}\pi_y d\pi_x = 2\pi (n+\gamma) \left(e\hbar H/c\right). \tag{3}$$

Here n is a non-negative integer, and γ is a non-negative constant less than 1. For instance, for a square dispersion law [i.e., $\mathcal{E}(\mathbf{p}) = (1/2m) \, p^2$], γ is $\frac{1}{2}$. [Equations (2) and (3) are valid even when the magnetic field H is nonuniform in the y direction.⁷] Consequently, the single-particle energies for the quasielectrons are classified by the quantum numbers (n, p_x, p_z, σ_z) , and are given by $\mathcal{E}_n(p_z) \pm \mu_B H$. Each level is degenerate owing to the quantum number p_x , and the energy spectrum consists of a set of spectral branches characterized by the quantum number $n=0, 1, \ldots$ For given p_z and spin polarization, the spacing between two neighboring branches is given by^{6,8}

$$\Delta \mathcal{E}_n(p_z) = \mathcal{E}_{n+1}(p_z) - \mathcal{E}_n(p_z) \cong 2\pi \left[e\hbar H / c \left(\frac{\partial S}{\partial \mathcal{E}} \right) \right]. \quad (4)$$

Here, the quantity $S(\mathcal{E}, p_z)$ denotes the cross-sectional area of p_z , bounded by the constant energy surface in the momentum space and is assumed to be a slowly 4618

varying function of &. It is given by the relation

$$S(\mathcal{E}, p_z) = \iint_{\mathcal{E}=\text{const}, p_z=\text{const}} \oint d\pi_x d\pi_y$$

$$= \mathcal{J} \pi_y(\mathcal{E}, p_z, \pi_x) d\pi_x$$

$$= 2\pi (n + \gamma) (e\hbar H/c). \tag{5}$$

To understand the effects of the presence of such discrete branches of the energy spectrum, we consider the case where the temperature T is zero and where there are many branches of the spectrum below the Fermi energy \mathcal{E}_F (that is, $\Delta \mathcal{E}_n \ll \mathcal{E}_F$ for some values of p_z). As shown in Eq. (4), the spacing $\Delta \mathcal{E}_n$ is proportional to the magnetic field H, while the Fermi energy \mathcal{E}_F can be shown to depend far less sensitively on H than does $\Delta \mathcal{E}_n$. Thus, as the strength of the applied magnetic field H is increased (or decreased), the branch of the energy spectrum initially right below (or right above) the Fermi energy level crosses over and becomes unoccupied (or occupied) by the quasielectrons. This effect causes a physical quantity to be periodic with respect to varying strengths of the magnetic field H. In fact, any macroscopic observable can be written as the sum of two distinct parts, namely, the nonoscillatory part (which is denoted by the superscript n.o.), which varies monotonically with the magnetic field, and the oscillatory part (which is classified by the superscript osc). The oscillatory part comes from the manifest discreteness in the branches of the quasielectron energy spectrum when there is an applied uniform magnetic field H. And thus such terms become evident only in the quantum domain provided that the thermal broadening as well as impurity broadening of the energy spectrum does not exceed the spacings of the branches of the spectrum.

According to Lifshitz and Kosevich,⁸ the thermodynamic potential Ω per unit volume and the magnetization M (= $-\partial\Omega/\partial H$) per unit volume are given as follows (we choose the unit $\hbar=1$):

$$\Omega(T, \mu, H) = \Omega^{\text{n.o.}} + \Omega^{\text{osc}}, \tag{6}$$

$$M = M^{\text{n.o.}} + M^{\text{osc}}, \tag{7}$$

where

$$\Omega^{\text{n.o.}} \cong \Omega_0 - \frac{1}{2} (\chi_P + \chi_L) H^2, \tag{8}$$

$$egin{align*} \Omega^{
m osc} &\cong igl[(2m^*\mu^*)^{3/2}/2\pi^2eta igr] (\omega_c/2\mu^*)^{3/2}a^{-1/2} \ & imes \sum_{l=1}^{\infty} rac{\cos(\pi l m^*/m) \; \exp(-2\pi l/\omega_c au)}{l^{3/2} \sinh(2\pi^2 l/eta \omega_c)} \ . \end{aligned}$$

$$\times \cos[2\pi l(\mu^*/\omega_c) \mp \frac{1}{4}\pi - 2\pi l\gamma], \quad (9)$$

$$M^{\text{n.o.}} \cong (\chi_P + \chi_L) H,$$
 (10)

$$M^{\text{osc}} = -[e(2m^*\mu^*)^{1/2}/2\pi\beta c](2\mu^*/\omega_c)^{1/2}a^{-1/2}$$

$$imes \sum_{l=1}^{\infty} rac{\cos(\pi l m^*/m) \; \exp(-2\pi l/\omega_c au)}{l^{1/2} \sinh(2\pi^2 l/eta\omega_c)}$$

$$\times \sin[2\pi l(\mu^*/\omega_c) \mp \frac{1}{4}\pi - 2\pi l\gamma], \quad (11)$$

$$a = (1/2\pi) \mid \partial^2 S(\mu, p_z) / \partial p_z^2 \mid_e. \tag{12}$$

Here, $\Omega_0 = \Omega \mid_{H=0}$ denotes the thermodynamic potential per unit volume in the absence of the magnetic field H, and μ is the chemical potential per electron ($\mu = \mathcal{E}_F$ when $T=0^{\circ}K$). The subindex e is used to denote a quantity evaluated at the extremal orbital,8 and the quantities m^* and μ^* are defined through the relations $2\pi m^* \mu^* = S_e(\mu)$ and $2\pi m^* = (d/d\mu) S_e(\mu)$. Here $S_e(\mu)$ denotes the value of $S(\mu, p_z)$ evaluated at the extremal orbital. [For a square dispersion law, $S(\xi, p_z) =$ $\pi(2m\mathcal{E}-\bar{p}_z^2)$, $S_e(\mu)=2\pi m\mu$, and $(d/d\mu)S_e(\mu)=2\pi m$, so that $m^* = m$, $\mu^* = \mu$, and a = 1. If there is more than one extremal orbital, one should add up the contribution like Eqs. (9) or (11) for each of the extremal orbitals to obtain the expressions for Ω^{osc} or M^{osc} . The symbol \(\simeg \) denotes restriction to the leading term in the expansion of (ω_c/μ^*) and $(\beta\mu^*)^{-1}$, where $\beta^{-1} = k_B T$ $(k_B$ is the Boltzmann constant) and $\omega_c = eH/m^*c$ is the cyclotron frequency. The quantity χ_P is the Pauli paramagnetic susceptibility9 and is given by8

$$\chi_P \cong \frac{1}{2} N(\mathcal{E}_F) (e/mc)^2. \tag{13}$$

Here $N(\mathcal{E}_F)$ is the number density of states at the Fermi surface per unit volume and per one of the spin polarizations. The quantity χ_L denotes the Landau diamagnetic susceptibility⁶ and is given by⁴

$$\chi_L \cong -\frac{1}{6}N(\mathcal{E}_F) \left(\frac{e}{c}\right)^2 \left\langle \frac{\partial^2 \mathcal{E}}{\partial p_x^2} \frac{\partial^2 \mathcal{E}}{\partial p_y^2} - \left(\frac{\partial^2 \mathcal{E}}{\partial p_x \partial p_y}\right)^2 \right\rangle_{av}, (14)$$

where $\langle \cdots \rangle_{av}$ denotes the average of the quantity taken over the Fermi surface. For the case of the square dispersion law, one obtains the well-known result $\chi_L \cong$ $-\frac{1}{3}\chi_P$. For a smooth Fermi surface, one expects $(-\chi_L)$ to be roughly $\frac{1}{3}(m/m^*)^2\chi_P$. The quantity τ is the relaxation time which reflects the scattering of a quasielectron by impurities, lattice imperfections, phonons, etc. This τ is different from the transport collision time $\tau_{\rm tr}$. As is well known, 10 $\tau_{\rm tr}$ is the collision time with the forward scattering being softened out. In the region of our interest, namely, where $\omega_c \ll \mu^*$ and $\beta^{-1} \ll \mu^*$, τ can be considered as a quantity very weakly dependent on the magnetic field H, and thus, for practical purposes, one can consider τ as independent of H. The factor $\exp(-2\pi l/\omega_c\tau)$ in Eqs. (9) and (11), which is due to Dingle, 11 represents the broadening of the branches of the energy spectrum due to the presence of impurities. Thus the finiteness of the relaxation time τ makes the oscillatory term exponentially small. Similarly, the presence of thermal broadenings of the branches of the energy spectrum is represented by the factor

$$\left[\sinh\left(2\pi^2l/\beta\omega_c\right)\right]^{-1}$$

in Eqs. (9) and (11).

It should be noted that the argument of the sinusoidal functions in the expressions for the oscillatory terms is $2\pi l\mu^*/\omega_c \mp \frac{1}{4}\pi - 2\pi l\gamma$. Since $(2\pi l\mu^*/\omega_c)$ is proportional to H^{-1} and $\mu^*\gg\omega_c$ in our region of interest, one has the following two properties for the oscillatory terms. First, they are rapidly oscillating functions of H and cannot

be expanded in powers of H. Second, a derivative with respect to H is μ^*/ω_c larger than the original quantity. Consequently, the nonoscillatory terms can be obtained by using perturbation theory with the terms containing H as perturbations, while the oscillatory terms can never be obtained by using any such perturbation method. And, since $M = -(\partial \Omega/\partial H)$, $M^{\rm osc}$ is of the order of μ^*/ω_c larger than $\Omega^{\rm osc}$, which allows the oscillatory part of the magnetization to be observable, and what one observes is, in fact, the de Haas-van Alphen effect.⁴ In deriving the oscillatory terms given by Eqs. (9) and (11), the method of stationary phase¹² is used to evaluate certain integrals asymptotically.⁸

We set $x = 2\pi^2/\beta\omega_c$ and consider the region where

$$x \gtrsim 1$$
 (15)

is satisfied. In such a region, the terms with l=1 in Eqs. (9) and (11) dominate over the other harmonics (namely, those terms with $l \ge 2$), and Eqs. (9) and (11) are simplified to

$$\Omega^{\text{osc}} \cong \left[(2\pi^{2}m^{*}\beta^{-1})^{3/2}/2\pi^{2}\beta \right] \cos(\pi m^{*}/m) a^{-1/2}x^{-3/2}
\times \exp(-x\beta/\pi\tau) (\sinh x)^{-1} \cos(\beta\mu^{*}x/\pi \mp \frac{1}{4}\pi - 2\pi\gamma),
(16)
M^{\text{osc}} \cong -\left[e(2m^{*}\beta^{-1})^{1/2}/2\pi^{2}c \right] \mu^{*} \cos(\pi m^{*}/m) a^{-1/2}
\times x^{1/2} \exp(-x\beta/\pi\tau) (\sinh x)^{-1} \sin(\beta\mu^{*}x/\pi \mp \frac{1}{4}\pi - 2\pi\gamma).
(17)$$

In measuring the de Haas-van Alphen effect in the noble metals in the region governed by Eq. (15), Shoenberg¹³ observed oscillatory terms much richer in harmonic content than was to be expected from Eq. (17). He resolved this discrepancy between observation and theory by pointing out that the electrons in the metal do not see the field \mathbf{H} alone but actually see the field $\mathbf{B} = \mathbf{H} + 4\pi \mathbf{M}$.¹³ That is, an electron in a metal does not discriminate the magnetic field due to the external source from that due to the motions of all other electrons in the system. Thus, the correct expressions for Eqs. (2)-(17) are obtained by substituting

$$B = H + 4\pi M \tag{18}$$

in place of H for the conventional expressions given by Eqs. (2)–(17). This conjecture by Shoenberg¹³ that H should be replaced by B in the conventional expressions was proven by Pippard¹⁴ based on the relations between thermodynamic functions. In fact, noting the principle of minimal electromagnetic interactions and that $\operatorname{curl} \mathbf{A}(\mathbf{x}) = \mathbf{B}(\mathbf{x})$, one obtains the rigorous justification of Shoenberg's conjecture as was done by the author. Definitions and relations among the thermodynamic functions are listed in Appendix A). Thus, hereafter Eqs. (2)–(17) are understood to be modified by replacing H with B everywhere, and the cyclotron frequency ω_c is $\omega_c = eB/m^*c = e(H + 4\pi M)/m^*c$.

Because of this substitution of H by B, the magnetization M governed by Eqs. (7), (10), and (11) satisfies

a nonlinear equation. In fact, this nonlinear equation for M, in the case of a strong external magnetic field H, has been exhaustively studied in the past. 13,14 However, an intriguing question is whether or not the nonlinear equation has nontrivial solutions for M even when the external magnetic field H is absent. Such a nontrivial solution represents the presence of a spontaneous magnetization that is consistent with the dynamics of the electrons and the Maxwell equations. The author and his co-workers¹ have shown that in fact such nontrivial solution exist for a system containing electrons at a high density. A stable state corresponding to a relative energy minimum exhibiting such a spontaneous magnetization is named¹ the Landau orbital ferromagnetic state (or, briefly, the LOFER state). In the remainder of this article, unless otherwise mentioned, we will consider the case where H=0 so that $B=4\pi M$ or $\omega_c = 4\pi eM/m^*c$.

To understand how a LOFER state arises, we first consider the case where $|M^{\text{n.o.}}| \gg |M^{\text{osc}}|$ so that $M \cong M^{n.o.}$. This is the case when the temperature is high enough or the metal is impure. For this case, as long as $4\pi(\chi_P + \chi_L) < 1$, there is no nontrivial solution for M (that is, there is no spontaneously magnetized state). As discussed earlier, $(-\chi_L)$ is roughly equal to $\frac{1}{3}(m/m^*)^2\chi_P$ and $4\pi\chi_P$ is equal to $(\alpha/\pi)(v_F/c)$, which is roughly 10^{-5} in order of magnitude for a metal. [Here $\alpha = e^2/\hbar c = (137.088 \cdots)^{-1}$ is the fine-structure constant and v_F is the Fermi velocity.] Thus, as long as $m^*/m \gtrsim$ 10^{-2} , $M^{\text{n.o.}}$ alone cannot give rise to a LOFER state. (From this, one can conclude that a theory based on treating the electromagnetic interaction as a perturbation, cannot give rise to a LOFER state.) It should be noted that for a square dispersion law, $4\pi(\chi_P + \chi_L) =$ $(8\pi/3)\chi_P = (2\alpha/3\pi)(v_F/c) \simeq O(10^{-5})$, and for metals, the condition $4\pi(\chi_P + \chi_L) \ll 1$ usually is satisfied.

Since, in general, the case where $M\cong M^{\text{n.o.}}$ (or, equivalently, $|M^{\text{n.o.}}|\gg |M^{\text{osc}}|$) does not exhibit a LOFER state, one has to recognize the vital importance of the presence of the oscillatory part of the magnetization M^{osc} in discussing how the LOFER states can arise. We thus consider the case where $|M^{\text{osc}}|\gtrsim |M^{\text{n.o.}}|$. For simplicity as well as for the sake of practical interest, we restrict ourselves to the region where temperature is high enough so that the condition (15) is valid but is not high enough so that $|M^{\text{n.o.}}|\gg |M^{\text{osc}}|$ is invalid. In such a region, from Eqs. (7), (10), and (17), one obtains the equation governing the (nonzero) spontaneous magnetization as

$$1 - 4\pi (\chi_P + \chi_L) \cong -\cos(\pi m^*/m) (\alpha \mu^*/\pi^3)$$

$$\times (2\beta/m^*c^2a)^{1/2} x^{3/2} e^{-x\beta/\pi\tau} (\sinh x)^{-1}$$

$$\times \sin[(\beta \mu^*/\pi) x \mp \frac{1}{4}\pi - 2\pi\gamma]. \quad (19)$$

We note that because of $\beta\mu^*\gg 1$, the sinusoidal function $\sin[(\beta\mu^*/\pi)x\mp\frac{1}{4}\pi-2\pi\gamma]$ is an extremely rapidly oscillating function of x. We also note that the quantity $x^{3/2}/\sinh x$ attains its maximum values of $(1.1457\cdots)^{-1}$

at $x = 1.288 \cdots$. Thus, Eq. (19) has a solution if $\cos^2(\pi m^*/m) (\alpha/\pi^3)^2(\beta\mu^*) (2\mu^*/m^*c^2a)$

$$\times \exp(-2.576\cdots\beta/\pi\tau)$$

> $(1.3128\cdots)\lceil 1-4\pi(\chi_P+\chi_L)\rceil^2$,

or it has a solution if $T \leq T_c$ (or $\beta \geq \beta_c$), where T_c (or $\beta_c^{-1} = k_B T_c$) is given by

$$(\beta_{c}\mu^{*})^{-1} = (0.7617\cdots) \cos^{2}(\pi m^{*}/m) (\alpha/\pi^{3})^{2}$$

$$\times (2\mu^{*}/m^{*}c^{2}a) [1 - 4\pi(\chi_{P} + \chi_{L})]^{-2}$$

$$\times \exp[-(2.576\cdots)\beta_{c}/\pi\tau]. \quad (20)$$

Here, the numerical factor $1.3128\cdots = (0.76173\cdots)^{-1}$ comes from the factor $(x^{3/2}/\sinh x)^{-2}$ evaluated at $x=1.288\cdots$. [Equation (6) of Ref. 1 is in error. There, the factor $(x^*)^{3/2}/\sinh x^*$ should be replaced by its square and then evaluated at $x^*=1.288\cdots$, which makes Eq. (6) of Ref. 1 equal to Eq. (20) given above, provided that the sample is pure (i.e., $\tau \to \infty$) and $4\pi(\chi_P + \chi_L) \ll 1$. The author is most grateful to Professor Shoenberg for pointing out this error to us.¹⁶

The strength of the spontaneous magnetization M can be estimated by using the relation $2\pi^2/\beta\omega_c=1.288\cdots$. The resulting magnetization is given by

$$4\pi M(T_c) \cong 1.139 \times 10^5 (m^*/m) k_B T_c (^{\circ}K)^{-1} G.$$
 (21)

[Relations similar to Eqs. (20) and (21) are also obtained by Shoenberg. [As mentioned earlier, because of the presence of the rapidly oscillating function $\sin[(\beta\mu^*/\pi)x \mp \frac{1}{4}\pi - 2\pi\gamma]$, below T_c there are many other nontrivial solutions for M in the neighborhood of a given solution. The difference ΔM between two neighboring solutions is roughly $(\omega_c/\mu^*)M$ and thus such solutions are densely populated.

For a pure sample (i.e., $\tau \rightarrow \infty$) and for the square dispersion law, Eq. (20) reduces to

$$k_B T_c / \mathcal{E}_F \cong 0.76 (\alpha v_F / \pi^3 c)^2 \simeq 10^{-7} (v_F / c)^2.$$
 (22)

Equation (22) is the same as Eq. (5) of Ref. 1. For the electrons in a metal governed by the square dispersion law, T_c and $4\pi M(T_c)$ are of the order of 10^{-7} oK and 10⁻² G, respectively. These values of temperature and magnetic field are much too low and too weak to be managed by present-day laboratory facilities. However, as was pointed out in Ref. 1, for the electrons in compact stars [such as white dwarfs and neutron stars (or pulsars), the values of spontaneous magnetizations resulting from the LOFER states can account for the strength of magnetic fields expected to be present in such astrophysical objects. ¹⁻³ For example, for $\varepsilon_F \sim 1$ MeV (which corresponds to a white dwarf), $4\pi M \sim$ 10^9 G and $T_c \sim 10^4$ °K, while for $\mathcal{E}_F \sim 100$ MeV (corresponding to a neutron star), $4\pi M \sim 10^{13}$ G and $T_c \sim$ 108 °K. (Of course, the relativistic effects modify these values, but not much.2) Thus, the LOFER states have significant astrophysical applications.

III. STABILITY OF LOFER STATE

To discuss the stability of a LOFER state with respect to variations in the magnetization M, we consider the Gibbs free-energy density $G(T, \mu, \mathbf{B})$. Since, in this section, we are interested mainly in the variable **B** (or **M**), the thermodynamic variables T and μ are not written down explicitly so that $G(\mathbf{B})$ denotes $G(T, \mu, \mathbf{B})$, etc. From Eqs. (A8), (A11), and (A15), one obtains a theorem stating that a LOFER state corresponds to an extremum point of the free energy $G(\mathbf{B})$ [i.e., $(\partial G/\partial \mathbf{B})_{T,\mu}=0$]. Of the extrema of $G(\mathbf{B})$, the LOFER states are defined to correspond to the minima of $G(\mathbf{B})$. There exists a one-to-one correspondence between the LOFER states and the minima of $G(\mathbf{B})$ [i.e., a set of **B** such that $(\partial G/\partial \mathbf{B})_{T,\mu} = 0$ and $(\partial^2 G/\partial \mathbf{B}^2)_{T,\mu} > 0$. The reason why the maxima of $G(\mathbf{B})$ are excluded from the consideration of the LOFER states is that such maxima correspond to magnetically unstable states. In fact, there is a well-known stability theorem for magnetic materials. This theorem¹⁷ states that, for an isotropic magnetic material, magnetic stability requires (a) $\mathbf{B} \cdot \mathbf{H} > 0$, and (b) $[\partial (\mathbf{H} \cdot \mathbf{B} B^{-1})/\partial B]_{T,\mu} <$ 0. When the condition (a) is violated (i.e., $\mathbf{B} \cdot \mathbf{H} < 0$), such a system is absolutely unstable against having a component of magnetization perpendicular to the direction of H; while, when the condition (b) is violated {i.e., $\lceil \partial (\mathbf{H} \cdot \mathbf{B} B^{-1}) / \partial B \rceil_{T,\mu} < 0$ } such a magnetic system is absolutely unstable against change in the component of magnetization parallel to **H**. Since in our case $\mathbf{H} = 0$, the stability condition (a) states that a system in a LOFER state is in a critical state.¹⁷ However, this possible instability does not interest us because, even if such an instability could occur, it does not change the component of the magnetization that is associated with the LOFER state. Thus, we consider the case when B and H are parallel and let the field H vanish. In such a case, the instability condition (b) becomes $(\partial H/\partial B)_{T,\mu} > 0$, which by virtue of Eq. (A8) is equivalent to the condition $(\partial^2 G/\partial B^2)_{T,\mu} > 0$. Then the states corresponding to the maxima of G(B) violate the condition (b) and thus such states are absolutely unstable against fluctuations in M. Such an instability is well known. For example, in the presence of a uniform magnetic field, it has been shown (based on microscopic theory) that a uniformly magnetized state for the electrons in a metal is unstable with respect to a spatially nonuniformly magnetized state when the condition $(\partial H/\partial B)_{T,\mu} < 0$ 15,18 [or, $(\partial M/\partial B)_{T,\mu} > (1/4\pi)$ since $B = H + 4\pi M$ is fulfilled.

As one can see from Eqs. (A11) and (6), the Gibbs free-energy density G(B) can be written as $G(B) = G^{\text{n.o.}}(B) + G^{\text{osc}}(B)$. Here $G^{\text{n.o.}}(B) = (1/8\pi)B^2 + \Omega^{\text{n.o.}}(B)$ and $G^{\text{osc}}(B) = \Omega^{\text{osc}}(B)$. Below T_c , because of the oscillatory behavior of $G^{\text{osc}}(B)$, G(B) has successive maxima and minima, and these minima correspond to the LOFER states. To resolve the question which of two given LOFER states is more stable, one has to consider the Gibbs free-energy density $\widetilde{G}(H)$. For a given configura-

tion of the true current (or, external magnetic field), the value of the corresponding $\tilde{G}(H)$ determines which of the states is more stable. If the value of $\bar{G}(H)$ is lower, the corresponding state is more stable. However, because of Eq. (A10), $\widetilde{G}(H) = G(B)$ when H = 0. Thus, a LOFER state corresponding to a lower value of G(B)is more stable than the one corresponding to a larger value of G(B). Consequently, a LOFER state makes a macroscopic transition to another LOFER state corresponding to a lower value of G(B). During this transition, the field B changes from one value to another in a continuous manner. And it is easy to conceive that the most probable path the system would follow during the transition is the one given by the free-energy density G(B). Needless to say, a field B appearing in G(B)which is not an extremum of G(B) corresponds to a nonequilibrium configuration. Only at an extremum of G(B) does the field B correspond to an equilibrium configuration. Since a LOFER state corresponds to a minimum of G(B), there is a maximum of G(B) in between two given LOFER states and this maximum of G(B)gives rise to an energy barrier. That is, in order to be able to make a macroscopic transition from one LOFER state to another LOFER state, this barrier must be overcome by the system. It is not necessary that the whole system makes the transition. Rather, such a transition occurs through a local fluctuation in M (or B) which involves a subsystem in a small volume u. But, in order that the transition be macroscopic, this volume u cannot be smaller than that of a cylinder with a cross-sectional area equal to $\pi \times$ (the cyclotron radius R_c)² and a height equal to $\hbar \times (\rho_z)$ corresponding to the extremal orbital)⁻¹. Recalling that for the square dispersion law, the extremal orbital occurs at $p_z=0$, we restrict ourselves to the case where the magnitude of p_z corresponding to the extremal orbit does not exceed \hbar/R_c . Then, we have

$$u > \pi R_c^3 \cong (v_F/m^*\omega_c^2) (2\pi\mu^*/\omega_c).$$
 (23)

If one makes a reasonable assumption that the *a priori* probability of the system in the volume u to be found in the state corresponding to B is proportional to the Boltzmann factor $\exp[-u\beta G(B)]$, the probability that this subsystem u crosses over the energy barrier $u\Delta G_b$ is

$$P \propto \exp(-u\beta\Delta G_b)$$
. (24)

Here ΔG_b is the energy barrier per unit volume that the subsystem must overcome in order to make the macroscopic transition.

To give an estimate for the barrier height ΔG_b , we restrict ourselves to the case where the temperature is infinitesimally lowered from the critical temperature T_c . As the temperature T is lowered from T_c , the barrier height ΔG_b increases, resulting in more stable LOFER states. Thus, the consideration restricted near T_c would give an upper bound for the transition probability P for the given LOFER state. Furthermore, because of

mathematical simplicity, we consider the spontaneous magnetization M that satisfies the condition given by Eq. (15).

The G(B)-versus-B curve can be visualized as follows. The nonoscillatory part $G^{n,o}(B)$ forms a monotonic background on top of which the oscillatory part $G^{\text{osc}}(B)$ is superimposed and gives many extrema of G(B). Very close to, but below, T_c there are two extremum points of G(B); one is the maximum and the other is the minimum. At T_c these two extrema coincide at B_0 . That is, as the temperature is lowered from T_c the maximum point B_{max} of G(B) and the minimum at B_{min} move away from each other starting at B_0 . Obviously the point B_0 corresponds to an inflection point of G(B)when the temperature is T_c . Because of the monotonic background due to $G^{n.o.}(B)$, in order that the LOFER state [which corresponds to the minimum of G(B)] make a transition to another equilibrium state (such as the state with M=0), the subsystem has to overcome the barrier due to the presence of the maximum of G(B). This barrier height ΔG_b is given by Eq. (B11). From Eqs. (23) and (B11), we therefore have

$$u\beta\Delta G_b > (2.5\alpha)^{-1} (2\pi\mu^*/\omega_c^{0}) (c/v_F) (\Delta\beta/\beta_c)^{3/2}.$$
 (25)

Here $\omega_c^0 = eB_0/m^*c$. Thus, taking $c/v_F \gtrsim 10^2$, one has

$$u\beta\Delta G_b > 10^3, \tag{26}$$

provided that

$$1\gg (T_c-T)/T_c \geq (\omega_c^{0}/2\pi\mu^*)^{3/2}$$
. (27)

Equation (27) is consistent with Eq. (B8). The characteristic time of the system is of the order of $(\omega_c^0)^{-1}$ which is far less than 10^{-20} sec. Noting that one million years is equal to 3.1×10^{14} sec, we have the decay time τ_D of the LOFER state when the temperature T is below T_c given by Eq. (27) as

$$\tau_D \cong (\omega_c^{\ 0})^{-1} \exp(\beta \mu \Delta G_b)$$

 $\gg 10^{-34} e^{10^3}$ million years. (28)

Note that $e=2.718\cdots$, which gives the lower bound of τ_D as an astronomical time. Therefore, such a LOFER state is quasistable.

So far we have restricted ourselves to consider the stability of a LOFER state under thermal fluctuations. This consideration is valid as long as the system together with the thermal bath are well isolated from other energy sources. In practice, the presence of the background perturbations such as cosmic rays should make the transition time τ_D considerably less than that given by Eq. (28).

Finally, there can be nonuniform states such as magnetic domain structures which are more stable than a uniformly magnetized LOFER state. However, once a system is in a LOFER state, it will take the time given by Eq. (28) to go into a more stable domain structure or a nonuniformly magnetized state, provided that the system is well isolated from other energy sources.

A rigorous and complete analysis of the stability of

a LOFER state does not seem to be feasible. To obtain a plausible result, we made the following working hypotheses:

- (a) The system is spatially extended, well isolated, and initially in a uniformly magnetized (LOFER) state
- (b) In order to disturb a given LOFER state, the majority of the electronic states that are responsible for the LOFER state to exist should be disturbed. Such states are those corresponding to energy eigenvalues located on the extremal orbitals. Thus, for the cases when the domain wall energy is positive, there is a minimum size u for the subsystem [which is given by Eq. (23) that can make the macroscopic transition as a whole. (For the cases when the domain wall energy is negative, a nonuniform state, rather than a domain structure, is obtained. However, the presence or the absence of such a nonuniform state involving the characteristic distance much smaller than the cyclotron radius cannot be easily settled by theoretical analysis. Based on the results of Ref. 15, we presume that such a nonuniform state is unlikely to occur.)
- (c) The *a priori* probability of the system to be found in the state corresponding to B is proportional to the Boltzmann factor $\exp[-u\beta G(B)]$, where G(B) is the Gibbs free energy.

All these plausible assumptions could be violated when applied to real metals. Especially, in view of the fact that practically speaking a sample cannot be well isolated from other energy sources and that the associated barrier energy is so small for a metal that the background energy fluctuations could easily make a uniformly magnetized LOFER state unstable against the formation of domain structure. Thus, the contents of Sec. III should be considered as a plausible argument.

IV. DISCUSSION

As was discussed in Sec. III, for the square dispersion law, the critical temperature T_c for electrons in a metal is too low to be managed by present-day laboratory facilities. Thus, in order to observe the LOFER state in a metal, one has to enhance the value of T_c given by Eq. (20). This enhancement of T_c can be achieved as follows. First, in order to minimize the disadvantage due to the presence of the factor $\exp[-(2.576\cdots)\beta_c/\pi\tau]$ in Eq. (20), the sample should be as pure as possible. Since τ is different from the transport relaxation time $\tau_{\rm tr}$, the sample should be free of not only impurities but also lattice point defects, grain boundaries, etc. Noting that the exponential factor actually comes from $\exp(-2\pi/\omega_c\tau)$ and that $\exp(-1.0) = 0.367\cdots$, one requires $\omega_c \tau \gtrsim 2\pi$ in order to minimize the disadvantage due to the presence of the finite τ . This condition $\omega_c \tau \gtrsim 2\pi$ is equivalent to

$$l \gtrsim 2\pi R_c,$$
 (29)

where l (= $v_F \tau$) is the mean free path of the electrons and R_c (= v_F/ω_c) is the cyclotron radius. These require-

ments of purity are peripheral technological problems which may be overcome in the future. Equation (29) states that it is advantageous (with regards to the problems of the purity of the sample) to have a large value for the spontaneous magnetization M associated with the LOFER states.

But there are also intrinsic requirements which are related to the geometry of the Fermi surface for the metal. Thus, to enhance T_c , one should choose a metal that possesses (a) m^*/m very close to an integer [so that $\cos^2(\pi m^*/m) = 1$, (b) $a = (2\pi)^{-1} |\partial^2 S(\mu, p_z)/\partial p_z^2|_e$ as small as possible, and (c) $\mu^* = S_e(\mu) [(d/d\mu) S_e(\mu)]^{-1}$ as large as possible. One cannot take much advantage of the requirement (c) through a choice of a sample with $\lceil (d/d\mu) S_e(\mu) \rceil = 2\pi m^*$ being smaller than $2\pi m$. This is because it contradicts the requirement (a). The optimum geometry of the Fermi surface is, therefore, the one having the extremal orbital with the following three properties: (a) The cyclotron mass m^* is close to the bare mass m of an electron, (b) the curvature of $S(\mu, p_z)$ for variable p_z at the extremal orbital is as small as possible, and (c) the cross-sectional area $S_e(\mu)$ is as large as possible. Considering the usual metallic density, it is unlikely that one can take much advantage of using property (c) given above. However, property (b) can be used to enhance T_c appreciably. To form a rough idea of the magnitudes of the quantities involved, we consider for example a metal with $m^*=m$ and $\mu^*=5$ eV. In order to obtain $T_c=0.1$ m °K, the quantity a for the metal should be 10^{-3} (whereas a=1 for the square dispersion law), and the sample should be pure enough so that there are no impurities or lattice imperfections within the distance of the order of a centimeter. These values of T_c and τ suggest that the possibility of observing the LOFER states in metals belongs to the future and depends on the development of techniques to obtain extremely low temperatures as well as extremely pure metals.

In view of the fact that techniques for obtaining extremely high pressures develop so rapidly, we note that one can increase the magnitude of μ^* [or $S_e(\mu)$] through applying an extremely high pressure to the metal resulting in an enhancement of T_c . This method of enhancing T_c is in accordance with the line suggested by the high values of T_c and M for the compact stars. However, we made no detailed studies yet on enhancing T_c through applying high pressure to a metal.

In the case when there is more than one extremal orbital, the contributions from each of these extremal orbitals should be added up to give Eqs. (9) and (11). However, it is not always advantageous to have many extremal orbitals in order to obtain a high value of T_c . This is because the contributions from these extremal orbitals easily add up destructively leaving a low value of T_c . An analysis of the LOFER states for a metal having the Fermi surface with more than one extremal orbital is quite complicated and will not be discussed here

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APPENDIX A: SOME THERMODYNAMIC FUNCTIONS AND RELATIONS AMONG THESE FUNCTIONS

In this this appendix, we introduce some relevant thermodynamic functions and give the relations among these functions. As is well known,¹⁷ the work δW done on the field by the external emf (which maintains the external currents) is

$$\delta W = (1/4\pi) \int_{V} d^3x \, \mathbf{H} \cdot \delta \mathbf{B}. \tag{A1}$$

Here $\mathbf{B} = \mathbf{H} + 4\pi \mathbf{M}$ and V is the volume of the magnetic material which is assumed to be fixed. The Helmholtz free-energy density $F(T, n, \mathbf{B})$ is introduced as

$$dF(T, n, \mathbf{B}) = -SdT + \mu dn + (1/4\pi)\mathbf{H} \cdot d\mathbf{B}. \quad (A2)$$

Here n denotes the number density of the particles and μ is the chemical potential per particle. S is the entropy per unit volume. Since the source of the field \mathbf{H} is the true current while that of the field \mathbf{B} (= $\mathbf{H}+4\pi\mathbf{M}$) is the true current as well as the magnetization current, the free energy $F(T, n, \mathbf{B})$ does not represent that corresponding to the system controlling the external magnetic field. The Helmholtz free-energy density $\widetilde{F}(T, n, \mathbf{H})$ is defined by

$$\tilde{F}(T, n, \mathbf{H}) = F(T, n, \mathbf{B}) - (1/4\pi)\mathbf{H} \cdot \mathbf{B}, \quad (A3)$$

and it is the one that should be considered when the system having a definite number of particles is in contact with a thermal bath and is in the presence of an external magnetic field. Corresponding Gibbs free-energy densities $G(T, \mu, \mathbf{B})$ and $\widetilde{G}(T, \mu, \mathbf{H})$ are given by

$$G(T, \mu, \mathbf{B}) = F(T, n, \mathbf{B}) - \mu n, \tag{A4}$$

$$\widetilde{G}(T, \mu, \mathbf{H}) = \widetilde{F}(T, n, \mathbf{H}) - \mu n.$$
 (A5)

These thermodynamic functions have the following properties:

(a)
$$dG = -SdT - nd\mu + (1/4\pi)\mathbf{H} \cdot d\mathbf{B}$$
, (A6)

$$d\tilde{G} = -SdT - nd\mu - (1/4\pi)\mathbf{B} \cdot d\mathbf{H}, \tag{A7}$$

(b)
$$(\partial G/\partial \mathbf{B})_{T,\mu} = (1/4\pi)\mathbf{H},$$
 (A8)

$$(\partial \tilde{G}/\partial \mathbf{H})_{T,\mu} = -(1/4\pi)\mathbf{B},\tag{A9}$$

(c)
$$\widetilde{G}(T, \mu, \mathbf{H}) = G(T, \mu, \mathbf{B}) - (1/4\pi)\mathbf{H} \cdot \mathbf{B}$$
. (A10)

Thermodynamic potentials per unit volume of the magnetic material $\Omega(T, \mu, \mathbf{B})$ and $\widetilde{\Omega}(T, \mu, \mathbf{H})$ are given by

$$\Omega(T, \mu, \mathbf{B}) = G(T, \mu, \mathbf{B}) - (1/8\pi)B^2,$$
 (A11)

$$\Omega(T, \mu, \mathbf{H}) = \widetilde{G}(T, \mu, \mathbf{H}) + (1/8\pi)H^2. \quad (A12)$$

These functions have the following properties:

(a)
$$d\Omega = -SdT - nd\mu - \mathbf{M} \cdot d\mathbf{B}$$
, (A13)

$$d\widetilde{\Omega} = -SdT - nd\mu - \mathbf{M} \cdot d\mathbf{H}, \tag{A14}$$

(b)
$$(\partial \Omega/\partial \mathbf{B})_{T,\mu} = -\mathbf{M},$$
 (A15)

$$(\partial \widetilde{\Omega}/\partial \mathbf{H})_{T,\mu} = -\mathbf{M},\tag{A16}$$

(c)
$$\widetilde{\Omega}(T, \mu, \mathbf{H}) = \Omega(T, \mu, \mathbf{B}) + 2\pi M^2$$
. (A17)

The thermodynamic potential $\Omega(T, \mu, \mathbf{B})$ can be obtained from the microscopic theory by using the following relation:

$$\Omega(T, \mu, \mathbf{B}) = -(\beta V)^{-1} \ln\{\mathrm{Tr}[\exp(-\beta(\hat{H} - \mu \hat{N}))]\}. \tag{A18}$$

Here \hat{N} is the number operator and \hat{H} is the Hamiltonian governing the motions of the particles constituting the magnetic material. For example, in our case,

$$\widehat{H} = \sum_{\sigma} \int d^3x \, \psi_{\sigma}^{\dagger}(\mathbf{x}) [\mathcal{E}(\boldsymbol{\pi}) + \mu_B \boldsymbol{\sigma} \cdot \mathbf{B}] \psi_{\sigma}(\mathbf{x}),$$

where $\psi_{\sigma}^{\dagger}(\mathbf{x})$ and $\psi_{\sigma}(\mathbf{x})$ are the field operators¹⁰ for electrons. Equation (A18) can be easily proven as was done in Ref. 19.

APPENDIX B: HEIGHTS OF ENERGY BARRIER ΔG_0 NEAR CRITICAL POINT (T_c, B_0)

We recall that the region of our interests is $\beta\mu^*\gg 1$, $(\omega_c/\mu^*)\ll 1$, and $x\gtrsim 1$. In such a region, the chemical potential μ can be shown²⁰ to depend very weakly on T and B. And thus, this dependence of μ on T and B will be ignored (since they contribute only high-order corrections) and we omit to write down μ dependence of the free-energy density G(T, B) explicitly.

The critical temperature T_c (or $k_BT_c = \beta_c^{-1}$) is given by the highest value of temperature T that enables the Gibbs free-energy density G(T,B) to possess at least one extremum point with respect to variable B [that is, there exists $B\neq 0$ such that $(\partial G/\partial B)_T = 0$]. We introduce the field B_0 (or $\omega_c^0 = eB_0/m^*c$) such that it satisfies (a) $\sin(2\pi\mu^*/\omega_c^0 \mp \frac{1}{4}\pi - 2\pi\gamma) = 1$, and (b) the closest to $(2\pi^2/\beta_c\omega_c) = 1.288\cdots$. From Eq. (20), one can easily see that these values of T_c and B_0 satisfy Eq. (19). And (T_c, B_0) is defined as the critical point of G(T, B) in the TB plane. As is noted in Sec. III, at T_c , the field B_0 is an inflection point of the free-energy density $G(T_c, B)$:

$$(\partial G/\partial B)_0 = (\partial^2 G/\partial B^2)_0 = 0.$$
 (B1)

Here the subindex zero denotes the evaluation of the derivatives at the critical point (T_c, B_0) . Fur-

thermore, because of $\sin(2\pi\mu^*/\omega_c^0 \mp \frac{1}{4}\pi - 2\pi\gamma) = 1$ [or $\cos(2\pi\mu^*/\omega_c^0 \mp \frac{1}{4}\pi - 2\pi\gamma) = 0$ and since $G^{\text{osc}}(T_c, B_0)$, $(\partial G^{\text{osc}}/\partial \beta)_0$, $(\partial^2 G^{\text{osc}}/\partial \beta^2)_0$, etc. are proportional to $\cos(2\pi\mu^*/\omega_c^0 \mp \frac{1}{4}\pi - 2\pi\gamma)$, one has

$$G^{\text{osc}}(T_c, B_0) = (\partial G^{\text{osc}}/\partial \beta)_0$$
$$= (\partial^2 G^{\text{osc}}/\partial \beta^2)_0 = \dots = 0.$$
 (B2)

To obtain an expression for the Gibbs free-energy density G(T, B) near the critical point (T_c, B_0) , we note that the power expansion of G(T, B) with respect to $\Delta B = B - B_0$ is possible if and only if $|\Delta B/B_0| <$ $(\omega_c^0/2\pi\mu^*)$ because of the presence of the factor $\cos(2\pi\mu^*/\omega_c \mp \frac{1}{4}\pi - 2\pi\gamma)$ in the expression for $G^{\text{osc}}(T, B)$. Thus, in order to be able to expand in powers of ΔB and $\Delta\beta$ (= β - β_c), we consider the neighborhood of the critical point given by

$$\Delta \beta / \beta_c \ll 1$$
, (B3)

$$|\Delta B/B_0| \ll (\omega_c^0/2\pi\mu^*). \tag{B4}$$

Then, in such a region, the Gibbs free-energy density G(T,B) is given by

$$G(T, B) - G(T_c, B_0) = [(\partial G^{\text{n.o.}}/\partial \beta)_0 + (\partial^2 G/\partial B \partial \beta)_0 \Delta B]$$

$$\times \Delta\beta \{1 + O(\Delta\beta/\beta_c) + O[(\mu^*/\omega_c^0)(\Delta\omega_c/\omega_c^0)]\}.$$
 (B5)

In deriving Eq. (B5), we made use of Eqs. (B1) and (B2).

As was mentioned in Sec. III, as T is lowered from T_c , the maximum point B_{max} and the minimum point B_{min} of G(T, B) (which coincided at B_0 when $T = T_c$) begin to move away from B_0 . These movements are governed, of course, by Eq. (19). Taking the variations of Eq. (19) with respect to β and B, and noting that $(1-x_c \coth x_c) = 0.496 \cdots$ (where $x_c = 1.288 \cdots$), one

$$\Delta \beta / \beta_c \cong [2\pi (\mu^*/\omega_c^0) (\Delta \omega_c / \omega_c^0)]^2$$

$$+\lceil 0.004 - (2\pi/\omega_c^0 \tau) \rceil (\Delta\omega_c/\omega_c^0)$$
.

Here $\Delta\omega_c$ corresponds to $B_{\text{max}}-B_0$ or $B_{\text{min}}-B_0$. Recalling that $(2\pi/\omega_c\tau) \lesssim 1$ and restricting oneself to the region given by

$$(\omega_c^0/2\pi\mu^*)^2 \ll |\Delta\omega_c/\omega_c^0| \ll \omega_c^0/2\pi\mu^*,$$
 (B6)

one has

$$\Delta\omega_c/\omega_c^0 \cong \pm (\omega_c^0/2\pi\mu^*) (\Delta\beta/\beta_c)^{1/2}$$
. (B7)

Equation (B7) gives the shifts of B_{max} and B_{min} from B_0 due to changes in T. Also, Eq. (B6) is equivalent to

$$(\omega_c^0/2\pi\mu^*)^2 \ll \Delta\beta/\beta_c \ll 1.$$
 (B8)

Equation (B8) is consistent with Eqs. (B3) and (B4). From Eqs. (B5) and (B7), we obtain the energy barrier per unit volume $\Delta G_b \left[= G(T, B_{\text{max}}) - G(T, B_{\text{min}}) \right]$ as

$$\Delta G_b \cong 2B_0 \mid (\partial^2 G/\partial B\partial \beta)_0 \mid (\omega_c^0/2\pi\mu^*)\beta_c(\Delta\beta/\beta_c)^{3/2}$$
. (B9)

Noting Eq. (A8) and $H = B - 4\pi M$, we have

$$(\partial^2 G/\partial B\partial \beta)_0 \cong -(\partial M^{\text{osc}}/\partial \beta)_0 \cong -(B_0/8\pi\beta_c)$$
. (B10)

We therefore have

$$\Delta G_b \cong (\omega_c^0/\pi\mu^*) (B_0^2/8\pi) (\Delta\beta/\beta_c)^{3/2}.$$
 (B11)

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