MAGNETIC SUSCEPTIBILITY IN STRONG MAGNETIC FIELDS

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Submitted to JETP editor May 26, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 47, 1958-1965 (November, 1964)

The magnetic moment of conduction electrons in strong magnetic fields is determined. It is found that the magnetic field strength dependence of the ground state energy and level density can be deduced from an experimental investigation of the magnetic susceptibility for an arbitrary dispersion law. It is shown that for a quadratic dispersion law in strong magnetic fields the total magnetic moment (diamagnetic or paramagnetic) approaches saturation.

1. INTRODUCTION

At the present time, the oscillating part of the magnetic susceptibility (the de Haas-van Alphen effect) has been well studied theoretically and experimentally (see, e.g., [1]). The oscillations, as is well known, occur at low temperatures T ($T < \mu H$) in weak magnetic fields ($\mu H < \zeta_0$, where μ is the Bohr magneton for the conduction electron, and ζ_0 is the limiting Fermi energy; the temperature will always be measured in energy units). The situation is much worse with respect to the investigation of the monotonic part χ of the magnetic susceptibility (which is all that remains in strong magnetic fields and high temperatures). Experimentally, this is because it is difficult to separate the monotonic susceptibility of the conduction electrons from the susceptibility of the lattice. The theoretical determination of the susceptibility for a non-quadratic dispersion law for the electrons cannot be made because, as it turns out, knowledge of the exact rules of quantization is required even in weak magnetic fields (see [2]). In this connection Rumer's calculation [3] for a free electron gas[1] is essential for an understanding of the dependence. Of particular interest, naturally, is the region of strong magnetic fields

$$\mu H \gg T, \zeta_0, \tag{1}$$

where the magnetic susceptibility of the electron gas depends on magnetic field (in zeroth approximation χ does not depend on H for $\mu H \ll \zeta_0$ and $\mu H \ll T$).

However, Rumer's treatment fails to give an understanding of the situation for the general case in precisely the region of Eq. (1). To show this, let us turn to the simplest case of a dispersion law $\epsilon = p^2/2m^*$ (ϵ is energy, p momentum), where the effective mass m^* is not the same as the mass m_0 of a free electron. Then the energy levels have the form (z is the magnetic field direction)

$$\varepsilon_n^{\pm} = p_z^2/2m^* + (n + 1/2) \times \mu H \pm \mu_0 H/2 = \varepsilon \pm \mu_0 H/2 \tag{2}$$

(the last term is associated with the spin paramagnetism), so that the energy of the ground state equals

$$\varepsilon_{min} = \frac{eH\hbar}{2c}\left(\frac{1}{m^*} - \frac{1}{m_0}\right) = \frac{1}{2}(\mu - \mu_0)H.$$

In magnetic fields of Eq. (1), when the separation between the levels is the greatest in energy, all the electrons accumulate near ϵ_{min}. From the expression for ϵ_{min} it is clear that the case of free electrons, when $m^* = m_0$, is a special one, for only in this case does the energy ϵ_{min} equal zero at any field H, whereas when $m^* \neq m_0$, the magnitude $|\epsilon_{min}| \to \infty$ as $H \to \infty$. As a result, it is natural to expect in the general case a substantially different dependence $\chi(T, H)$ than the one offered in [3].

The purpose of this paper is to elucidate the form of this dependence. First, in Sec. 2, we shall carry out the calculation of $\chi(H, T)$ for the case of Eq. (2) both with $\mu \sim \mu_0$ and with $\mu \gg \mu_0$ (the case $\mu \ll \mu_0$, i.e., $m^* \gg m_0$, is not met in real conductors) and estimate the magnetic fields that satisfy the condition (1). The basic problem of Sec. 2 is to obtain $\chi(H, T)$ in explicit form and to demonstrate the idea of the calculation. In Sec. 3 we discuss the case when the μH that satisfies the inequality (1) is significantly less than all

[1] Also of great interest is the calculation of magnetic susceptibility for semiconductors with a loop of extrema in a perpendicular loop of magnetic field, [4] which has been carried out on the basis of exact quantization rules.

characteristic energies associated with the form of the dispersion law, so that the dispersion law, although anisotropic, is quadratic. Finally, Sec. 4 is devoted to an arbitrary dispersion law, when the quantization rule is in general unknown. In this case it is natural to change the statement of the problem and consider as given not the dispersion law in the absence of a magnetic field, but a ground state in a given magnetic field, and to study the dispersion law on the basis of experimental data. Equation (20), which allows this to be done, has an extremely simple form.

In this work we shall consider only the most interesting case of one zone with a constant number of electrons. As can readily be appreciated, when there are several zones the susceptibility in strong magnetic fields will still be determined only by the group with the greatest effective mass; all the other groups "freeze out." Also "frozen out" are the anomalously small sections of the large group which were considered by one of the authors.[5] In those cases where the number of conduction electrons is itself not a constant and tends to zero at T = 0, as happens in semiconductors, the basic dependence of the susceptibility on temperature and magnetic field is caused by the "freezing out" of electrons with increasing magnetic field and is obviously an exponential one.

2. MAGNETIC SUSCEPTIBILITY FOR THE CASE OF AN ISOTROPIC QUADRATIC DISPERSION LAW

To find the total magnetic moment

$$M = -(\partial F / \partial H)_{TVN}, \tag{3}$$

as is known, it is sufficient to know the free energy of the system (see [6])

$$F = N\zeta + \Omega, \qquad \Omega = -\frac{2eHT}{ch^2}\sum_{n=0}^{\infty}\int_0^{\infty} dp_z \times\left\{\ln\left[1+\exp\left(\frac{\zeta-\varepsilon_n^+}{T}\right)\right] + \ln\left[1+\exp\left(\frac{\zeta-\varepsilon_n^-}{T}\right)\right]\right\} \tag{4}$$

($\epsilon_n^{\pm}$ is given by Eq. (2), and the quantities F, N, and Ω entering in Eq. (4) are defined per unit volume). The chemical potential ζ is found from the requirement that the number of particles be constant:

$$N = -\frac{\partial\Omega}{\partial\zeta} = \frac{2eH}{ch^2}\sum_n\int_0^{\infty} dp_z\left\{\left[\exp\left(\frac{\varepsilon_n^+-\zeta}{T}\right)+1\right]^{-1} + \left[\exp\left(\frac{\varepsilon_n^--\zeta}{T}\right)+1\right]^{-1}\right\}. \tag{5}$$

Since $\mu H \gg \zeta_0$, we shall make an assumption that is fundamental for the sequel, namely, that (accurate to an exponential in $\mu H/T$) all the electrons are concentrated near the level n = 0—this is physically obvious and is easily verified (see also [7]). With this the problem becomes unidimensional ($\epsilon_0 = p_z^2/2m^* + \frac{1}{2}(\mu - \mu_0)H$), and by setting

$$p_z = \sqrt{2m^*Tx}, \quad \alpha_{\pm} = [\zeta - \tfrac{1}{2}(\mu \mp \mu_0)H]/T, \quad \gamma = 4\zeta_0^{3/2}/3\mu HT^{1/2}, \quad A = NT/\gamma H, \tag{6}$$

we obtain equations for the determination of $\alpha_{\pm}$ and Ω:

$$\int_0^{\infty}\frac{dx}{\sqrt{x}}\left(\frac{1}{\exp(x-\alpha_+)+1}+\frac{1}{\exp(x-\alpha_-)+1}\right)=\gamma, \tag{7}$$

$$\Omega = -AH\int_0^{\infty}\frac{dx}{\sqrt{x}}\{\ln[1+\exp(\alpha_+-x)] + \ln[1+\exp(\alpha_--x)]\}. \tag{8}$$

We shall find first of all the susceptibility of a system of particles with masses m* that are a very great deal smaller than the mass of a free electron m_0, i.e., we shall consider the case $\mu \gg \mu_0$. Equation (7) can be solved in two limiting cases:

1. Case $\gamma \gg 1$ (i.e., $\zeta_0 \ll \mu H \ll \zeta_0(\zeta_0/T)^{1/2}$, so that in every case $\zeta_0 \gg T$). Here $\alpha_+ \gg 1$, $\alpha_+/(\mu H/T) \ll 1$ (this can be verified after determination of ζ and α_+), so that $\mu H/T - \alpha_+ \sim \mu H/T$ and the terms in Eq. (5) of higher order of smallness in $\mu H/T$ are omitted (terms with $n \neq 0$ are exponentially small). Thus, only those electrons in the ground energy level n = 0 are actually important.

In principle, the magnitude of α_- for $\alpha_+ \gg 1$ can be arbitrary. If $\alpha_- \gg 1$, then

$$\gamma \sim \int_0^{\alpha_+}\frac{dx}{\sqrt{x}}+\int_0^{\alpha_-}\frac{dx}{\sqrt{x}} = 2(\sqrt{\alpha_+}+\sqrt{\alpha_-}), \quad \sqrt{\alpha_{\pm}} = \frac{\gamma}{4}+\frac{\mu_0 H}{\gamma T};$$

$$\zeta = \tfrac{1}{2}\mu H + T(\gamma/4)^2 + (\mu_0 H/\gamma T)^2. \tag{9}$$

For this, in order to satisfy the inequality $\alpha_- \gg 1$, it is necessary to fulfill the condition $\gamma/4 \pm \mu_0 H/\gamma T \gg 1$, or

$$(\mu H/\zeta_0)^3 \ll \tfrac{4}{9}\mu/\mu_0 \tag{10}$$

(which is possible only for $\mu \gg \mu_0$). Using (8) and (9), we find the thermodynamic potential

$$\Omega \sim -\tfrac{4}{3}AH(\alpha_+^{3/2}+\alpha_-^{3/2}) = NT\{\gamma^2/24 + 8(\mu_0 H/\gamma T)^2\}$$

and then the magnetic moment

$$M = -\tfrac{1}{2}N\mu\left[1-\frac{4}{27}\left(\frac{\zeta_0}{\mu H}\right)^3-\frac{9}{2}\frac{\mu H}{\zeta_0}\left(\frac{\mu_0 H}{\zeta_0}\right)^2\right]. \tag{11}$$

On the other hand, if $\alpha_- \lesssim 1$, the second term in Eq. (7) can be eliminated. Then $\gamma = 2(\alpha_+)^{1/2}$. The requirement $\alpha_- < 1$ is fulfilled under condition that

$$(\mu H/\zeta_0)^3 > 4\mu/9\mu_0. \tag{12}$$

In this case the chemical and thermodynamic potentials are determined as

$$\zeta = {}^1/_2\mu H + T\gamma^2/4 - \mu_0 H/2,$$
$$\Omega = -{}^4/_3 AH\alpha_+^{3/2} = -{}^1/_6 NT\gamma^2, \tag{13}$$

hence we have for the magnetic moment

$$M = -{}^1/_2 N\mu\{1 - \mu_0/\mu - {}^{16}/_{27}(\zeta_0/\mu H)^3\}. \tag{14}$$

2. In the other limiting case $\gamma \ll 1$ (i.e., in the energy scale $\mu H \gg \zeta_0(\zeta_0/T)^{1/2}$) it is necessary, as can be seen from Eq. (7), that $-\alpha_\pm \gg 1$, and near the level n = 0 the electron gas obeys Boltzmann statistics (i.e., $(e^x + 1)^{-1} \sim e^{-x}$). Then

$$\gamma \sim \int_0^\infty \frac{dx}{\sqrt{x}}(e^{-x+\alpha_+} + e^{-x+\alpha_-}) = \sqrt{\pi}(e^{\alpha_+} + e^{\alpha_-}),$$
$$\zeta = \frac{\mu H}{2} - T\ln\frac{2\sqrt{\pi}}{\gamma}\cosh\frac{\mu_0 H}{2T}. \tag{15}$$

From this

$$\Omega = -AH\gamma = -NT, \quad F = N\zeta - NT,$$
$$M = -\frac{1}{2}N\mu\left\{1 - \frac{2T}{\mu H} - \frac{\mu_0}{\mu}\tanh\frac{\mu_0 H}{2T}\right\}. \tag{16}$$

In case $\mu \sim \mu_0$ the calculation is completely analogous, except that the inequality (10) and, consequently, the dependence (11) for the magnetic moment cease to be fulfilled.

Since $\mu H \gg \zeta_0$, T and $\mu \gtrsim \mu_0$, then in virtually all investigated field regions the same dependence M(H) is observed (below is written also the well known expression for M in weak field):

$$M = -\frac{N}{2}(\mu - \mu_0), \quad \mu H \gg \max(\zeta_0, T), \tag{17a}$$

$$M = -\frac{N\mu^2}{3(\zeta_0 + T)}\left(1 - 3\frac{\mu_0^2}{\mu^2}\right)H,$$
$$\mu H \ll \max(\zeta_0, T). \tag{17b}$$

Thus the magnetic moment depends on magnetic field in the following way. If in the absence of a magnetic field the electron gas is not degenerate, $\zeta_0 < T$, then the absolute magnitude of the magnetic moment initially, for $\mu H \ll T$, grows linearly with field, according to (17b), being diamagnetic if $m^* < m_0/(e)^{1/2}$ and paramagnetic if $m^* > m_0/(3)^{1/2}$; then, when $\mu H \gg T$, it approaches, in accordance with (17a), saturation, diamagnetic for $m^* < m_0$ and paramagnetic for $m^* > m_0$. In the intermediate field region $\mu H \sim T$, there can be extrema; in particular if $m_0 > m^* > m_0(3)^{-1/2}$, then surely there will be at least one maximum [since there must be a transition from a growing paramagnetic moment (17b) to a diamagnetic moment that is growing in modulus (16)]. There is no temperature dependence of the moment in zeroth approximation, but in subsequent approximations in limiting strong fields $\mu_0 H \gg T$, $\mu H \gg \zeta_0$, $\zeta_0(\zeta_0/T)^{1/2}$, T, the moment has, according to (16), a universal character and is isotropic.

If in the absence of a magnetic field the electron gas is degenerate, $\zeta_0 > T$, then in the field regions $\mu H \gg \zeta_0$ and $\mu H \gg 2\pi^2 T$, everything goes as described above, and in the field region $2\pi^2 T < \mu H \lesssim \zeta_0$ de Haas-van Alphen oscillations of the magnetic susceptibility occur (see [2]).

Curiously, the saturation of the magnetic moment in strong fields occurs as if each electron had an intrinsic moment $(\mu_0 - \mu)/2$.

From Eq. (17a) it is clear that Rumer's case $\mu = \mu_0$ is a special one, when (17a) gives M = 0; then we have to use the complete formulas (14) and (15), according to which

$$M = 8N\zeta_0^3/27\mu^2 H^3, \quad \zeta_0 \ll \mu H \ll \zeta_0\sqrt{\zeta_0/T}, \quad \zeta_0 > T; \tag{18a}$$
$$M = NT/H, \quad \mu H \gg \zeta_0, \ \zeta_0\sqrt{\zeta_0/T}, \ T. \tag{18b}$$

The essential difference between the dependences M(T, H) in the cases $m^* = m_0$ [Eqs. (18a) and (18b)] and $m^* \neq m_0$ [Eq. (17a)] is obvious.

We shall now ascertain how well the condition $\mu H \gg \zeta_0$ is fulfilled, and thereby find the limits of applicability of the equations obtained. In good metals $\zeta_0 \sim 10^{4\circ}$K, for the ground electronic groups $m \sim m_0 \sim 10^{-27}$ g, and the required inequality corresponds to $H \gtrsim 10^9$ Oe. To decrease H small ζ_0 are desirable (i.e., low densities of conduction electrons), as well as small electron masses m. Consequently, it is necessary to turn to semimetals, where $\mu H \gg \zeta_0$ does not by any means correspond to presently unattainable fields. Thus, for Bi the parameter $\zeta_0 \sim 100^\circ$K, i.e., $H \sim 10^4$ Oe. (However, this calculation is not directly applicable to metals like Bi with a substantially nonquadratic dispersion law [8]; it is necessary to use the results of Sec. 4.)

The cases of semiconductors and of anomalously small zones in good metals, when (1) is fulfilled, will appear as the subject of a separate communication (see also Sec. 1).

3. ANISOTROPIC QUADRATIC DISPERSION LAW

We turn now to the case of an arbitrary dispersion law. In strong magnetic fields $\mu H \gg \zeta_0$, as has already been mentioned, one needs to find the quantization rules for the first Landau levels before he can determine the magnetic susceptibility. In all real magnetic fields the separation between the levels is small compared to the width of the conduction band $\Delta\epsilon$, so that even $\mu H \gg \zeta_0$ corresponds to an almost empty or almost completely filled band and the dispersion law can be considered to be quadratic. Then the quantization has the form

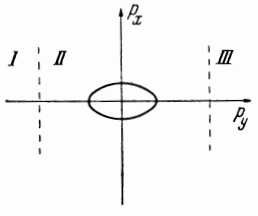
$$S(\varepsilon, p_z) = \left(n + \frac{1}{2}\right) ehH/c, \quad n = 0, 1, 2\ldots \qquad (19)$$

(S is the area of the section of the equal-energy surface by the plane p_z = const).

The quantization rule (19) can be derived rigorously, starting from the Hamiltonian in magnetic field (with no account taken of interzone transitions) given in [9] [Eq. (6.20)]. We remark in passing that this equation is approximately valid for arbitrary p, but is by no means exact; it is impossible to obtain an exact Schroedinger quantum operator from a classical dispersion law. Thus, for example, the two distinct operators

$$\cos \hat{p}_x a_x \cos \hat{p}_y a_y + \cos \hat{p}_y a_y \cos \hat{p}_x a_x,$$
$$\cos (\hat{p}_x a_x + \hat{p}_y a_y) + \cos (\hat{p}_x a_x - \hat{p}_y a_y)$$

both satisfy all the general requirements considered in [9]. In regions far from the classical orbit (I and III in the figure), the usual quasiclassical solution can be found. In the narrow region II ($\mu H \ll \Delta\epsilon$), considering the smallness of p_y, we expand ϵ in p_y, transform to the Fourier representation, and, considering the smallness of p_x (p_x and p_y are canonical conjugate operators to within a constant multiplier), we expand in p_x. In this we arrive at the same formula that would be obtained by the replacement of p_x and p_y by the corresponding operators in the expansion $\epsilon(p_x, p_y)$ for small p_x, p_y, i.e., in quadratic form. Solving the resulting harmonic oscillator equation and joining it to the solutions in the regions I and III of the figure, we obtain Eq. (19).



We shall not dwell to any extent on the case of arbitrary quadratic dispersion. The form of the dependence $\chi(H)$ in this case is determined by the general formula (20) of Sec. 4 with account taken of the quantization rules (19). We mention only that since the value of the effective mass changes with a change in field direction, then the signs of the inequalities used can change, and thereby the character of the dependence $\chi(H)$.

Of course, the expansion in quadratic form and Eq. (19) are valid only in the absence of degeneracy (associated with high symmetry) or the presence of any kind of additional parameter of the dimensions of energy comparable to or less than ζ_0. Thus, for example, for Bi, although $\zeta_0 \sim 100°K$, its Fermi surface is not ellipsoidal, and the susceptibility has a more complicated dependence on magnetic field (see [8]).

4. GENERAL CASE OF ARBITRARY DISPERSION

We now obtain the equation for the magnetic moment in case the dispersion law at H = 0 for small p does not reduce to a quadratic law (see, for example, [4,8]). We are interested in fields so strong that the separation between the levels and the energy of the ground state (due both to the diamagnetism and the paramagnetism of the electrons) are large in comparison with T and ζ_0 (but, of course, $H \ll c\hbar/ea^2$, where a is the lattice period). Such fields are attainable at present for semiconductors and metals like Bi, As, and Sb. Quantization in such fields is naturally not possible in the general case.

However, considerations similar to those employed in Sec. 2 (for $\mu H \gg \zeta_0$, ζ_0 ($\zeta_0/T)^{1/2}$, T) show that the susceptibility is determined only by the ground state and lead to the following result:

$$M = -N\frac{\partial \varepsilon_0}{\partial H} + NT\frac{\partial}{\partial H}\ln \nu_0, \qquad (20)$$

where N is the density of electrons, $\epsilon_0(H)$ and $\nu_0(H)$ are the energy and density of states for the ground state, i.e., absolutely the lowest of all the quantum numbers of the level. It can be shown that, as in Sec. 2, the lowest approximation $M = -N\partial\epsilon_0/\partial H$ is valid already when $\mu H \gg \zeta_0$, T.

For a known quantization rule obtained, for example, in particular cases of metals of the Bi type by Abrikosov and Fal'kovskiĭ [8] and of semiconductors of the Te type by Firsov [10] and with a loop of extrema by Rashba, [4] the magnetic susceptibility can be determined from Eq. (20). Thus, for quadratic dispersion, we have from (19) and (20)

$$M = -\tfrac{1}{2}N(\mu - \mu_0) + NT/H. \qquad (21)$$

For an unknown quantization law, Eq. (20) gives the possibility of finding from experimental data the dependence on the magnetic field for $\epsilon_0(H)$ in strong fields from the principal term of the susceptibility, and $\nu_0(H)$ in limiting strong magnetic fields from the weak temperature dependence.

CONCLUSIONS

A. In strong magnetic fields ($\mu H \gg \zeta_0$, T) the magnetic moment in lowest approximation is independent of temperature and is determined only by the dependence of the ground state energy on magnetic field:

$$M = -N\partial\varepsilon_0 / \partial H. \qquad (22)$$

Fields like this can be attained, evidently, for poor metals (Bi type).

B. In limiting strong magnetic fields (the form of the estimate depends on the character of the quantization for a given dispersion law) the magnetic moment has a small addition, linearly dependent on temperature; the coefficient of proportionality is determined by the density of states in the ground state $N\partial \ln \nu_0/\partial H$.

C. For a quadratic dispersion law

$$\partial\varepsilon_0 / \partial H = {}^1/_2(\mu - \mu_0), \quad \partial \ln \nu_0 / \partial H = 1/H$$

and the magnetic moment approaches saturation, corresponding to diamagnetism for $m^* < m_0$ and to paramagnetism for $m^* > m_0$.

D. For an isotropic quadratic dispersion law there can be found small additions (Eqs. (11), (14), (16)) to the basic formula, Eq. (17a), that have a different form in the different regions into which the region of fields $\mu H \gg \zeta_0$, T is divided (in weak fields $\mu H \ll \max(\zeta_0, T)$ the well known formula, Eq. (17b) is valid).

E. The case of a free electron gas studied by Rumer,[3] for which $\mu = \mu_0$, is a special one. Here the magnetic moment approaches zero in accordance with Rumer's Eqs. (18a)—(18c).

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Translated by L. M. Matarrese
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