Semiclassical Theory of Magnetic Energy Levels and Magnetic Susceptibility of Bloch Electrons*

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In this paper the semiclassical condition for the quantization of magnetic energy levels of Bloch electrons is extended to one order higher in the magnetic field. The result enables us to make a calculation of the magnetic susceptibility of Bloch electrons which includes both the de Haas–van Alphen effect and the steady susceptibility. While the basic calculation is made for fairly simple closed orbits, the results have been extended to include spin splitting, self-intersecting orbits, and open orbits, but omitting consideration of banding of magnetic levels and magnetic breakdown. Possible new oscillations due to self-intersecting orbits are discussed.

DERIVATIONS of the magnetic susceptibility of Bloch electrons have either concentrated on the de Haas–van Alphen effect,[1] or the steady susceptibility.[2-4] In this paper we give a unified derivation of both parts of the susceptibility based on a semiclassical treatment of the levels which is valid when banding of Landau levels[5] and magnetic breakdown[6] are not important.

In Part I we discuss the magnetic levels. The primary result (Sec. 1) is a generalization of Onsager's[7] quantization condition for closed magnetic orbits. This generalization amounts to replacing the constant of integration γ [see Eq. (1)] by a function of energy, and evaluating it to first order in the magnetic field. An equation-of-motion approach is used to obtain the result. In the remainder of part I the result is extended in several ways. In Secs. 2 and 3 we include higher order terms in the effective Hamiltonian obtained in R, and in particular extend the method to cover spin splitting. In Sec. 4 the problem of self-intersecting orbits is discussed, and in Sec. 5 the results are extended to the case of open orbits.

Part II is concerned with the magnetic susceptibility. The main result (Sec. 6) is a calculation of the total magnetic susceptibility for closed electron orbits by a method quite similar to that of Lifshitz and Kosevich[1] but which, because of the additional terms in γ, gives the correct steady susceptibility. The problem of the variation of the period with the magnetic field is considered in Sec. 7. The extension of the results to more complicated orbits is made in Sec. 8, and the possibility of new oscillations due to self-intersecting orbits is discussed.

Part I: MAGNETIC ENERGY LEVELS OF BLOCH ELECTRONS

1. Basic Derivation

In describing the behavior of Bloch electrons in electric and magnetic fields, semiclassical methods have been most successful. The theory has been reviewed extensively by Lifshitz and Kaganov.[8] The semiclassical quantization condition for magnetic levels is the well-known Onsager equation,

$$A(\mathscr{E}_n) = (n+\gamma)(2\pi eB/c), \tag{1}$$

where B is the magnetic field, and A is the cross-sectional area of the orbit in $\mathbf{k}$ space, which is the intersection of a constant-energy surface with the surface $k_z = \text{constant}$, where z is the direction of the magnetic field $\mathbf{B}$. This equation applies to orbits which are closed and nonintersecting, and we shall make these assumptions in the present section.

Equation (1) has been derived by applying the Bohr-Sommerfeld quantization condition to the problem,[8] or by using the WKB method.[9] In this section we shall derive Eq. (1) by an equation-of-motion method, which is capable of generalization to higher order in the magnetic field.

We begin with an effective one-band Hamiltonian which appears in the momentum-space Schrödinger

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[1] I. M. Lifshitz and A. M. Kosevich, Zh. Eksperim. i Teor. Fiz. **29**, 730 (1955) [English transl.: [Soviet Phys.—JETP **2**, 636 (1956)]. For the free-electron case see R. E. Peierls, *Quantum Theory of Solids* (Clarendon Press, Oxford, England, 1956), Chap. 7.

[2] J. E. Hebborn and E. H. Sondheimer, J. Phys. Chem. Solids **13**, 105 (1960); C. P. Enz, Helv. Phys. Acta **33**, 89 (1960); G. H. Wannier and U. N. Upadyaya, Phys. Rev. **136**, A803 (1964); J. E. Hebborn, J. M. Luttinger, E. H. Sondheimer, and P. J. Stiles, J. Phys. Chem. Solids **25**, 741 (1964).

[3] E. I. Blount, Phys. Rev. **126**, 1636 (1962).

[4] L. M. Roth, J. Phys. Chem. Solids **23**, 433 (1962), hereafter referred to as R. We shall follow most of the notation of this reference, including setting $\hbar = 1$.

[5] G. E. Zilberman, Zh. Eksperim. i Teor. Fiz. **33**, 387 (1957); **34**, 515 (1958) [English transl.: Soviet Phys.—JETP **6**, 299 (1958); **7**, 355 (1958)]; P. G. Harper, Proc. Phys. Soc. (London) **A68**, 874, 879 (1955); A. D. Brailsford, *ibid.* **A70**, 275 (1957).

[6] M. Cohen and L. Falikov, Phys. Rev. Letters **5**, 544 (1960); A. B. Pippard, Proc. Roy. Soc. (London) **A270**, 1 (1962); U. N. Upadyaya (to be published).

[7] L. Onsager, Phil. Mag. **43**, 1006 (1952).

[8] I. M. Lifshitz and M. I. Kaganov, Usp. Fiz. Nauk **69**, 419 (1959); **78**, 411 (1962) [English transl.: Soviet Phys.—Uspekhi **2**, 831 (1959); **5**, 878 (1962)].

[9] G. E. Zilberman, Zh. Eksperim. i Teor. Fiz. **32**, 296 (1957) [English transl.: Soviet Phys.—JETP **5**, 208 (1957)].

equation,

$$\mathcal{3C}(\kappa)\psi(\mathbf{k}) = \mathcal{E}\psi(\mathbf{k}). \tag{2}$$

This equation has been obtained by the author in R and by others.[3,10,11] Here κ is the kinetic-momentum operator, given in $\mathbf{k}$ space by

$$\kappa = \mathbf{k} + e/c\mathbf{A}(i\nabla_k), \tag{3}$$

where $\mathbf{A}$ is the vector potential for the magnetic field. This is essentially equivalent to the real-space operator

$$\mathbf{P} = \frac{1}{i}\nabla + \frac{e}{c}\mathbf{A}(\mathbf{r}). \tag{4}$$

The components of κ have the commutation relations

$$[\kappa_x, \kappa_y] = ieB/c, \tag{5}$$

assuming the magnetic field to be in the z direction. Thus, except for a constant, the two components act like a canonical coordinate and momentum.

The effective Hamiltonian $\mathcal{3C}(\kappa)$ is defined to be the symmetrized operator, that is, the components of κ always appear symmetrically. Thus, if we know $\mathcal{3C}(\mathbf{k})$, then the operator $\mathcal{3C}(\kappa)$ is defined. In R, an expression of $\mathcal{3C}(\mathbf{k})$ is given in powers of the magnetic field, the first term of which is the band energy $\mathcal{E}(\mathbf{k})$. In this section we shall consider only this lowest order term, extending the result to second order in the field in the next section. Also, we assume nondegenerate bands, neglecting spin effects which will also be discussed in Secs. 2 and 3.

We shall be dealing with symmetrized operators and will need a multiplication theorem which is proved in R. If $A(\kappa)$ and $B(\kappa)$ are symmetrized operators, then

$$A(\kappa)B(\kappa) = C(\kappa), \tag{6}$$

where $C(\kappa)$ is the symmetrized operator obtained from

$$C(\mathbf{k}) = [\exp(-i\mathbf{h} \cdot \nabla_k \times \nabla_{k'})]A(\mathbf{k})B(\mathbf{k}')|_{k'=k}. \tag{7}$$

Here, $\mathbf{h} = eB/2c$. This theorem enables us to evaluate products as expansions in powers of h of symmetrized functions. The type of expansion involved is referred to by Kohn[11] as a commutator expansion.

Consider now Eq. (2). Suppose we have a raising operator, C^+, such that $C^+\psi$ is a solution of the Schrödinger equation

$$\mathcal{3C}C^+\psi = [\mathcal{E} + \omega(\mathcal{E})]C^+\psi. \tag{8}$$

If we can find C^+ and $\omega(\mathcal{E})$, we can construct the spectrum. Equation (7) holds if

$$[\mathcal{3C}, C^+] = C^+\omega(\mathcal{3C}). \tag{9}$$

We now make the assumption that $C^+ = C^+(\kappa)$ is a symmetrized function of κ. It is understood, however, that this applies only to κ_x and κ_y; $\kappa_z = k_z$ is treated as

[10] G. H. Wannier and D. R. Fredkin, Phys. Rev. **125**, 1910 (1962).
[11] W. Kohn, Phys. Rev. **115**, 1460 (1959).

a constant. Now $\omega(\mathcal{3C})$, it should be pointed out, is not a symmetrized function of κ, since it is explicitly a function of $\mathcal{3C}$. This distinction will become clearer below. For now, let us try to find C^+ and ω to lowest order in the magnetic field. Since we expect ω to be proportional to B, we need not consider further commutation properties. The left side can be evaluated to first order in B, and we obtain

$$-2i\mathbf{h} \cdot (\nabla_k \mathcal{E}(\mathbf{k})) \times \nabla_k C_0^+ = C_0^+\omega_0(\mathcal{E}, k_z). \tag{10}$$

Now, letting $C_0^+ = e^{i\phi(\mathbf{k})}$, and $\nabla_k\mathcal{E} = \mathbf{v}$, we have

$$2\mathbf{h} \times \mathbf{v} \cdot \nabla_k\phi = \omega_0. \tag{11}$$

To solve this equation, we divide both sides by $v_\perp = (v_x^2 + v_y^2)^{1/2}$ and integrate around the classical cyclotron path:

$$\omega_0 \int^k \frac{d\lambda}{v_\perp} = \int^k d\lambda \frac{2\mathbf{h} \times \mathbf{v}}{v_\perp} \cdot \nabla_k\phi = \frac{eB}{c} \int^k d\lambda \cdot \nabla_k\phi. \tag{12}$$

The last integral is just ϕ, except for an arbitrary function of energy and k_z. Now, in order for C^+ to be single-valued, ϕ must change by integral multiples of 2π on traversing the orbit. Therefore

$$2\pi s = (c\omega_0/eB) \oint d\lambda/v_\perp. \tag{13}$$

For $s = 1$, we obtain the level spacing or cyclotron frequency for ω_0, and the result is equivalent to the Shockley integral[12]:

$$m^* \equiv eB/\omega_0 c = \oint d\lambda/2\pi v_\perp. \tag{14}$$

It is interesting to relate ϕ to the time spent by a classical electron traversing the orbit[8]

$$\phi = \omega_0 t(\mathbf{k}),$$

$$t(\mathbf{k}) = (c/eB) \int^k d\lambda/v_\perp. \tag{15}$$

We shall return to this notation in considering the electron spin.

Equation (14) can be related to the Onsager condition, Eq. (1), by writing the area in the form

$$A \equiv \int dk_x dk_y = \int d\mathcal{E} \oint d\lambda/v_\perp. \tag{16}$$

To calculate the level spacing from Eq. (1), we differentiate:

$$\frac{\partial A}{\partial \mathcal{E}}\omega_0 = 2\pi eB/c. \tag{17}$$

But the derivative is just $\oint d\lambda/v_\perp$, according to Eq. (16), which makes Eq. (17) equivalent to Eq. (14). To this lowest order, γ is then a constant of integration.

[12] W. Shockley, Phys. Rev. **79**, 191 (1950).

We could now attempt to obtain higher order corrections to C^+ and ω from Eq. (9), but it turns out to be simpler to use a slightly different approach. Suppose we assume that Onsager's relation holds with γ a function of energy. This is equivalent to assuming that for the nth eigenfunction

$$A(\mathfrak{IC})\psi_n = (n+\Gamma(\mathfrak{IC}))\frac{2\pi eB}{c}\psi_n, \tag{18}$$

where we have replaced γ by Γ to distinguish it from the final result of the next section, which includes the field dependence of $\mathfrak{IC}$. Then, by the same argument we used before, and using $h=eB/2c$ we have for C^+,

$$[A(\mathfrak{IC})/4\pi - h\Gamma(\mathfrak{IC}), C^+] = hC^+. \tag{19}$$

We now have, however, the same situation with A and Γ as we previously had with $\omega(\mathfrak{IC})$, namely that instead of a symmetrized function of κ, we have a function of $\mathfrak{IC}$. We can, however, find a function $A_S(\kappa)$ which is an expansion of A in even powers of h in terms of symmetrized functions of κ. The corresponding function $A_S(\mathbf{k})$ is given by

$$A_S(\mathbf{k}) = A(\mathcal{E}) + h^2 A_2(\mathbf{k}) + h^4 A_4 + \cdots. \tag{20}$$

We shall need only A_2 here, and it is given by

$$A_2(\mathbf{k}) = -(1/12)\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}\{3A''\mathcal{E}_{\alpha\gamma}\mathcal{E}_{\beta\delta} + 2A'''v_\alpha v_\gamma\mathcal{E}_{\beta\delta}\}, \tag{21}$$

as is shown in Appendix A. We have here introduced a notation for the numerous cross-products which occur. The Greek indices represent two-dimensional vector or gradient components (i.e., $\mathcal{E}_{\alpha\beta} \equiv \nabla_\alpha\nabla_\beta\mathcal{E} = \partial^2\mathcal{E}/\partial k_\alpha\partial k_\beta$) and ϵ is a 2×2 antisymmetric tensor:

$$\epsilon_{12} = -\epsilon_{21} = 1, \quad \epsilon_{11} = \epsilon_{22} = 0. \tag{22}$$

We use summation convention, and the prime represents differentiation with respect to energy.

For Γ, we shall assume an expansion in h of $\Gamma(\mathfrak{IC})$

$$\Gamma = \Gamma_0(\mathfrak{IC}) + 2h\Gamma_1(\mathfrak{IC}) + (2h)^2\Gamma_2(H)\cdots, \tag{23}$$

and now symmetrizing, we obtain

$$\Gamma_S(\mathbf{k}) = \Gamma_0(\mathcal{E}) + 2h\Gamma_1(\mathcal{E}) + h^2(\Gamma_{02}(\mathcal{E}) + 4\Gamma_2(\mathcal{E}))$$
$$+ h^3(2\Gamma_{12}(\mathcal{E}) + 8\Gamma_3(\mathcal{E})) + \cdots, \tag{24}$$

where the second subscript, if any, corresponds to the symmetrization. We shall actually calculate only the first two terms of the expansion. It is possible in principle to go to higher order, in which case we see from Eq. (24) that in any order, the symmetrization terms depend only on lower order terms, so that the procedure is consistent.

We now apply our multiplication theorem to Eq. (19), giving[13]

$$-2i\sin(h\epsilon_{\alpha\beta}\nabla_\alpha\nabla_\beta')$$
$$\times(a_S(\mathbf{k}) - h\Gamma_S(\mathbf{k}))C^+(\mathbf{k}')|_{\mathbf{k}=\mathbf{k}'} = hC^+(\mathbf{k}), \tag{25}$$

[13] This form of the commutator is essentially that found in J. E. Moyal, Proc. Cambridge Phil. Soc. **45**, 99 (1947).

where we have written $a = A/4\pi$. We now substitute the expansions in Eqs. (20) and (24) into this equation, and also the following expansion of C^+

$$C^+ = C_0^+ + hC_1^+ + h^2 C_2^+. \tag{26}$$

We now equate like powers of h, but first we can greatly simplify the expression by noticing that for any function $F(\mathbf{k})$

$$2\epsilon_{\alpha\beta}\nabla_\alpha a\nabla_\beta F = \partial F/\partial\phi, \tag{27}$$

where ϕ is the function of Eq. (15). The resulting equations for C^+ are

$$i\frac{\partial C_0^+}{\partial\phi} + C_0^+ = 0, \tag{28}$$

$$i\frac{\partial C_1^+}{\partial\phi} + C_1^+ = i\frac{d\Gamma_0}{da}\frac{\partial C_0^+}{\partial\phi}, \tag{29}$$

$$i\frac{\partial C_2^+}{\partial\phi} + C_2^+ = i\frac{\partial\Gamma_0}{\partial a}\frac{\partial C_1^+}{\partial\phi}$$
$$+ 2i\frac{\partial\Gamma_1}{\partial a}\frac{\partial C_0^+}{\partial\phi} - 2i\epsilon_{\alpha\beta}\nabla_\alpha a_2\nabla_\beta C_0^+$$
$$+ (i/3)(\epsilon_{\alpha\beta}\nabla_\alpha\nabla'_\beta)^3 a(\mathbf{k})C_0^+(\mathbf{k}')|_{\mathbf{k}=\mathbf{k}'}. \tag{30}$$

Looking first at Eq. (28), we see immediately that the solution $C_0^+ = e^{i\phi}$ is the same function we met before. Equation (29) is an inhomogeneous equation, and in order for it to have a solution, the right-hand side must be orthogonal to the solution of the homogeneous equation, i.e., C_0^+. But since the right-hand side is proportional to $\partial C_0^+/\partial\phi = iC_0^+$, it can only be orthogonal if $d\Gamma_0/da = 0$. Thus we must have Γ_0 a constant, which is the Onsager result. In Eq. (30) we apply the same argument, i.e., requiring the right-hand side to be orthogonal to C_0^+ to evaluate $d\Gamma_1/da$. Thus, multiplying by $e^{-i\phi}$ and integrating around the cyclotron orbit we have

$$2(\partial\Gamma_1/\partial a) = 2\oint\frac{d\phi}{2\pi}\epsilon_{\alpha\beta}\nabla_\alpha a_2\nabla_\beta\phi$$
$$+ \frac{i}{3}\oint\frac{d\phi}{2\pi}e^{-i\phi}(\epsilon_{\alpha\beta}\nabla_\alpha\nabla'_\beta)^3 a(\mathbf{k})e^{+i\phi(\mathbf{k}')}|_{\mathbf{k}=\mathbf{k}'}, \tag{31}$$

where we have used $d\phi = 2\pi d\lambda/a'v_\perp$ as the integration variable.

The first term of Eq. (31) can be simplified by the following equation for a function $F(\mathbf{k})$

$$\oint d\phi\nabla_\perp F(\kappa) = \frac{\partial}{\partial A}\oint d\phi(\nabla_\perp A)F(\kappa), \tag{32}$$

which can be derived from the two-dimensional Green's

theorem

$$\int d^2\kappa \nabla_\perp F(\mathbf{k}) = \oint d\lambda \mathbf{n} F(\mathbf{k}), \qquad (33)$$

by expressing d^2k as $dAd\phi/2\pi$, and by differentiating with respect to A. Thus the first term of Eq. (31) becomes

$$-2\frac{\partial}{\partial a}\oint \frac{d\phi}{2\pi}\epsilon_{\alpha\beta}(\nabla_\alpha a)a_2\nabla_\beta\varphi = \frac{\partial}{\partial a}\oint\frac{d\phi}{2\pi}a_2. \qquad (34)$$

The *second* term of Eq. (31) is much more difficult to reduce, and we defer the calculation to Appendix B, stating only the result which, combined with Eq. (34) gives

$$\frac{\partial\Gamma_1}{\partial a} = \frac{1}{2}\frac{\partial}{\partial a}\oint\frac{d\phi}{2\pi}a_2 + \frac{1}{6}\frac{\partial^2}{\partial a^2}\oint\frac{d\phi}{2\pi}\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}a_{\alpha\gamma}a_{\beta\delta}, \qquad (35)$$

with a_2 obtained from Eq. (21). Equation (35) now contains a combination of energy and area functions, and it is straightforward to put all derivatives in terms of energy. The result is

$$\frac{\partial\Gamma_1}{\partial\mathscr{E}} = \frac{1}{96\pi}\frac{\partial^2}{\partial\mathscr{E}^2}\oint\frac{d\lambda}{v_\perp}\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}\mathscr{E}_{\alpha\gamma}\mathscr{E}_{\beta\delta}, \qquad (36)$$

so that integrating and carrying out the sums

$$\Gamma_1 = \bar{\Gamma}_1 + \frac{1}{48\pi}\frac{\partial}{\partial\mathscr{E}}\oint\frac{d\lambda}{v_\perp}(\mathscr{E}_{xx}\mathscr{E}_{yy} - \mathscr{E}_{xy}^2), \qquad (37)$$

where Γ_1 is a constant independent of energy.

We now wish to evaluate the constants $\bar{\Gamma}^i$ and to discuss the range of validity of the solutions. Let us consider the cyclotron orbits on a given $\mathbf{k}_z$ plane for various values of the energy. (For more detail see Sec. 5.) The topology of the orbit changes at maxima, minima, and saddle points. In the vicinity of a maximum or minimum, the orbits are simple and closed. However, at a saddle point, the orbits may change from closed to open, or two orbits may merge into one. Examples appear in Figs. 2 and 3; we shall concentrate on Fig. 2 for the present, as it involves only closed orbits. Since our whole scheme of approximation depends on the character of the orbit it breaks down at saddle points where, for example, Γ_1 is singular, as we shall see in Sec. 4. The solutions C^+ are completely separate for regions of $\mathbf{k}$ space separated by orbits through saddle points, or self-intersecting orbits. In particular we should evaluate $\bar{\Gamma}_0$ and $\bar{\Gamma}_1$ for each such region separately.

The simplest case is that of an orbit close enough to a maximum or minimum so that the extremum can be reached from the given orbit by changing the energy without passing through a saddle point. Then $\bar{\Gamma}_0$ and $\bar{\Gamma}_1$ can be calculated in the vicinity of the maximum or minimum. The result, which is obtained in Appendix C, is $\bar{\Gamma}_0 = \frac{1}{2}$ and $\bar{\Gamma}_1 = 0$. In Sec. 4 we shall show how to construct a bridge between two regions separated by a self-intersecting orbit. The result indicates that the choice $\bar{\Gamma}_0 = \frac{1}{2}$ and $\bar{\Gamma}_1 = 0$ applies for all closed orbits, except in the immediate vicinity of a self-intersecting orbit. The case of open orbits is considered in Sec. 5.

We have thus achieved our goal of extending Onsager's equation one further order in B. The particular form of Γ_1, Eq. (37), leads directly to the Landau-Peierls[14] term in the susceptibility, as we shall show in Sec. 6.

We should note in concluding this section that the present method is directly applicable to the more usual WKB situation and extends the result one higher order in Planck's constant. The expansion is related to Wigner's classical expansion of the partition function.[15]

2. Inclusion of Interband Terms

In this section we generalize the above result to include spin and interband effects. These two can be included by considering the one band effective Hamiltonian of R and others[3,10] to second order in h. If we assume nondegenerate bands we can write

$$\mathcal{3C}(\kappa) = \mathcal{3C}_0(\kappa) + h\mathcal{3C}_1(\kappa) + h^2\mathcal{3C}_2(\kappa), \qquad (38)$$

with $\mathcal{3C}_0(\mathbf{k}) = \mathscr{E}(\mathbf{k})$. The interesting case for particles with spin is, however, doubly degenerate in the absence of the field for a crystal with inversion symmetry. This case is discussed in some detail in the next section, and it is shown how to obtain the spin splitting. We shall assume here that we have a representation in which the Zeeman interaction is diagonal (such a representation is derived in fact in Sec. 3) in which case we can replace $\mathcal{3C}_1$ by $\pm\mathcal{3C}_1$, the sign depending on the spin state.

We now need an expression for the area function for the modified Hamiltonian, expanded in powers of h. We can use the following expansion for a volume integral:

$$I = \int_{\mathscr{E}_0(\kappa)+\epsilon_1(\kappa)<\mathscr{E}} dkg(k)$$

$$= \sum_n \frac{(-1)^n}{n!}\frac{\partial^n}{\partial\mathscr{E}^n}\int_{\mathscr{E}_0(\mathbf{k})<\mathscr{E}} dkg(k)[\mathscr{E}_1(k)]^n, \qquad (39)$$

where $\mathscr{E}_1$ comprises here the second two terms of Eq. (38). Eq. (39) is proved by writing the integral I as

$$I = \int d\mathbf{k}\, f(\mathscr{E}_0(\mathbf{k})+\mathscr{E}_1(\mathbf{k})-\mathscr{E})g(\mathbf{k}), \qquad (40)$$

where f is a step function (or Fermi function for zero temperature) and expanding formally in powers of $\mathscr{E}_1$.

[14] L. D. Landau, Z. Physik **64**, 629 (1930); R. Peierls, *ibid*. **80**, 763 (1933).

[15] E. P. Wigner, Phys. Rev. **40**, 749 (1932).

Using this result we can write for the area function

$$A = A(\mathcal{K}_0) - \oint (d\lambda/v_\perp)(\pm h\mathcal{K}_1 + h^2\mathcal{K}_2)$$

$$+ \frac{1}{2} \oint \frac{d\lambda}{v_\perp} h^2 \mathcal{K}_1^2, \quad (41)$$

so that with Eqs. (18) and (37) and using $\bar{\Gamma}_0 = \frac{1}{2}$ and $\bar{\Gamma}_1 = 0$, we finally obtain the generalized Onsager quantization condition as Eq. (1) with γ now given as

$$\gamma = \frac{1}{2} \pm \oint \frac{d\lambda}{4\pi v_\perp}\mathcal{K}_1 + \frac{eB}{c}\oint \frac{d\lambda}{4\pi v_\perp}\mathcal{K}_2$$

$$+ \frac{eB}{c}\frac{\partial}{\partial\mathcal{E}}\oint \frac{d\lambda}{4\pi v_\perp}\{\frac{1}{6}(\mathcal{E}_{xx}\mathcal{E}_{yy} - \mathcal{E}_{xy}^2) - \frac{1}{2}\mathcal{K}_1^2\}. \quad (42)$$

3. The Zeeman Interaction

In this section we shall consider in more detail the Zeeman interaction or spin splitting for Bloch electrons. In R and other work[3,10] it is shown that this is given by

$$\mathcal{K}_1(\kappa) = g_0\mu_B s_0 \cdot \mathbf{B} + \mu_B \mathbf{B} \cdot \mathbf{x} \times (\mathbf{p} + m\mathbf{v}), \quad (43)$$

where g_0 and s_0 are the free-electron g factor and spin, and μ_B is the Bohr magneton. $\mathbf{x}$ is the periodic part of the coordinate operator with matrix elements

$$\mathbf{x}_{nn'}(\mathbf{k}) = i\int d\mathbf{r} u_{n\mathbf{k}}^*(\mathbf{r})\nabla_\mathbf{k} u_{n'\mathbf{k}}(\mathbf{r}), \quad (44)$$

with $u_{n\mathbf{k}}$ the periodic part of the Bloch function, and $\mathbf{p}$ is the momentum operator whose diagonal part is $m\mathbf{v}$.

For zero magnetic field the Hamiltonian commutes with the time-inversion operator[16]

$$\mathcal{K} = -i\sigma_y\mathcal{C}, \quad (45)$$

where σ_y is the y component of the Pauli spin vector which results in a spin reversal, and $\mathcal{C}$ is the complex-conjugation operator. The invariance of $\mathcal{K}$ under time inversion implies a double degeneracy, the well-known Kramers degeneracy.[16] For crystals without spin this implies that the states $n\mathbf{k}$ and $n-\mathbf{k}$, for nondegenerate bands, are degenerate, since $\mathcal{C}$ takes $e^{i\mathbf{k}\cdot\mathbf{r}}$ into $e^{-i\mathbf{k}\cdot\mathbf{r}}$. The same is true with spin except that the degeneracy is between $n\mathbf{k}\uparrow$ and $n-\mathbf{k}\downarrow$. However, if the crystal also has inversion symmetry, the operator $\mathcal{g}\mathcal{K}$, with $\mathcal{g}$ the inversion operator, also commutes with $\mathcal{K}$, giving degenerate states $n\mathbf{k}\uparrow$ and $n\mathbf{k}\downarrow$, since $\mathcal{g}$ undoes the reversal by $\mathcal{K}$ of $\mathbf{k}$. We should note that in the presence of spin-orbit interaction the states $\uparrow$ and $\downarrow$ are not pure spin states but mixtures, a fact which does not change our argument. For crystals without inversion symmetry the $n\mathbf{k}\uparrow$-$n\mathbf{k}\downarrow$ degeneracy is removed by spin-orbit inter-

action, so that in principle at least the bands are non-degenerate. We shall assume that inversion symmetry is present, and note that the results would apply to a crystal lacking inversion symmetry if the $n\mathbf{k}\uparrow$-$n\mathbf{k}\downarrow$ splitting is very small.

When a magnetic field is applied the Kramers degeneracy is split. For our crystal with inversion symmetry it has been shown by Blount and Cohen[17] that it is possible to deduce a form for the Zeeman interaction in terms of an effective spin $\mathbf{s}$, which is no longer the free spin because of spin-orbit interaction. The result is the 2×2 effective Hamiltonian

$$\mathcal{K}_1(\mathbf{k}) = \mu_B \mathbf{s}\cdot\mathcal{g}(\mathbf{k})\cdot\mathbf{B}, \quad (46)$$

where $\mathcal{g}$ is a g tensor which is not necessarily symmetric.[17] The form of Eq. (46) is dictated by the fact that the linear term in B must change sign under the operation $\mathcal{g}\mathcal{K}$. The actual behavior of $\mathcal{K}$ under time inversion is altered when $\mathbf{k}$ is replaced by κ, but this does not change the form of Eq. (46).

The effect of the Zeeman interaction on a band electron can be described classically in a rather simple way. We can let $\frac{1}{2}\mathcal{g}(\mathbf{k})\cdot\mathbf{B}$ be an effective magnetic field acting on the electron. As the electron traverses the cyclotron orbit in the magnetic field its spin feels a time-varying magnetic field. Here the time is related to the wave vector $\mathbf{k}$ for a point on the orbit by Eq. (15). The problem is thus reduced to solving the equation of motion of a classical spin in a time varying magnetic field:

$$d\mathbf{s}/dt = \mu_B(\mathcal{g}(\mathbf{t})\cdot\mathbf{B})\times\mathbf{s}. \quad (47)$$

Quantum-mechanically there appear to be two methods to approach this problem. One is the equation-of-motion method of the last section, and the second is the finding of a unitary transformation which diagonalizes the Zeeman interaction. We shall discuss the equation-of-motion method first and then use the result to obtain the desired transformation.

For the spin problem we shall be interested in a raising operator D^+ which reverses the spin Let us assume the form

$$D^+ = \mathbf{\sigma}\cdot\mathbf{D}(\kappa) \quad (48)$$

where $\mathbf{D}(\kappa)$ is a symmetrized vector function of κ. Since the spin cannot be raised more than once, we must have $(D^+)^2 = 0$. If we are interested in the result to lowest order in B, we neglect noncommutivity of κ and this implies

$$\mathbf{D}\cdot\mathbf{D} = 0 \quad (49)$$

as a condition on $\mathbf{D}$. [Since D is complex, Eq. (49) does not imply that $\mathbf{D}$ vanishes.] Using Eq. (9) we have now to require that

$$[\mathcal{E} + \mu_B\mathbf{s}\cdot\mathcal{g}\cdot\mathbf{B}, \mathbf{\sigma}\cdot\mathbf{D}] = \mathbf{\sigma}\cdot\mathbf{D}\omega_1. \quad (50)$$

We shall evaluate this to lowest order in $\mathbf{B}$. Thus for

[16] A. Messiah, *Quantum Mechanics* (North-Holland Publishing Company, Amsterdam, 1963), Vol. II, pp. 667–676.

[17] E. I. Blount and M. Cohen, Phil. Mag. **5**, 115 (1960).

the $\mathcal{E}$ commutator we use the multiplication rule, Eq. (7), to first order, and for the Zeeman part we ignore the κ noncommutativity. Thus we have

$$-2ih\epsilon_{\alpha\beta}v_\alpha\nabla_\beta\boldsymbol{\sigma}\cdot\mathbf{D}+i\mu_B\boldsymbol{\sigma}\cdot(\mathbf{g}\cdot\mathbf{B})\times\mathbf{D}=\omega_1\boldsymbol{\sigma}\cdot\mathbf{D}. \quad (51)$$

The first term on the left, from Eqs. (27), (17), and (15) can be written as the "time" derivative $-i(\partial/\partial t)\boldsymbol{\sigma}\cdot\mathbf{D}$. We can also omit $\boldsymbol{\sigma}$, as the result must hold for all components of $\mathbf{D}$, giving

$$\frac{\partial\mathbf{D}}{\partial t}=\mu_B(\mathbf{g}\cdot\mathbf{B})\times\mathbf{D}+i\omega_1\mathbf{D}. \quad (52)$$

We see that the equation for D is almost the same as the classical equation of motion for $\mathbf{s}$, Eq. (47). In fact the equation for $\mathbf{D}e^{-i\omega_1 t}$ is identical except that t is not now *officially* the time. Thus a solution of the classical equations of motion is closely related to a solution for $\mathbf{D}$. Equation (52) is a coupled set of linear equations for which the boundary condition is single-valuedness of $\mathbf{D}$ or, in terms of t, periodicity with the cyclotron period. We also have the condition, Eq. (49). In regard to this, we can dot $\mathbf{D}$ into Eq. (52) to give

$$\frac{1}{2}\frac{\partial}{\partial t}\mathbf{D}\cdot\mathbf{D}=i\omega_1\mathbf{D}\cdot\mathbf{D}, \quad (53)$$

so that if Eq. (49) holds for $t=0$, it holds for all t. The eigenvalues ω_1 of Eq. (52) include the spin splitting, as we shall see below, or classically the precession frequency of the spin.

A special case of Eq. (52) which is readily solved is when $\mu_B\mathbf{g}\cdot\mathbf{B}\equiv\mathbf{W}$ is in the z direction, but may vary with t in a periodic fashion. We can then combine the x and y component equations of Eq. (52) and, defining $D_x\pm iD_y$ as $D_\pm$, we have

$$\partial D_\pm/\partial t=i(\omega_1\pm W_z)D_\pm, \quad (W_x=W_y=0). \quad (54)$$

Assuming W_z to be positive, we have the solution

$$D_\pm=D_\pm{}^0\exp\left[i\int^t dt'(\omega_1\pm W_z)\right], \quad (W_x=W_y=0), \quad (55)$$

In order for $D_\pm$ to be single-valued we must have
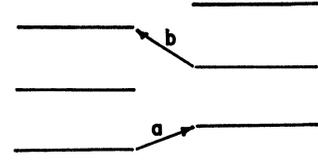
$$\omega_1=\mp\omega_0/2\pi\oint dt W_z+s\omega_0, \quad (W_x=W_y=0), \quad (56)$$

where s is an integer. Taking the lower sign and $s=0$, we find

$$\omega_1=\bar{\omega}_1\equiv\omega_0/2\pi\oint dt W_z, \quad (W_x=W_y=0), \quad (57)$$

We shall assume that $\bar{\omega}_1<\omega_0$ (if $\bar{\omega}_1>\omega_0$ we can choose s to make ω_1 between zero and ω_0). Our choice corresponds to taking $D_-{}^0$ finite and $D_+{}^0$ zero, and gives for D^+, the

raising operator,

$$D^+=\boldsymbol{\sigma}\cdot\mathbf{D}=\begin{pmatrix}0 & D^- \\ 0 & 0\end{pmatrix}, \quad (W_x=W_y=0), \quad (58)$$

which takes a spin-down state to a spin-up state. Note, however, that taking the upper sign and $s=1$ also gives a positive frequency (i.e., $\omega_0-\bar{\omega}_1$). Thus there are two basic choices of raising operators, which corresponds to the transitions a and b in Fig. 1. We can always multiply D^+ by $e^{is\varphi}=e^{is\omega_0 t}$ to obtain a combination of spin splitting and cyclotron frequencies, but we have already found out what we need about the level structure.

The spin splitting, however defined, comes from averaging W_z over the cyclotron orbit. This is entirely consistent with using Eqs. (1) and (42) for the energy quantization, since our case corresponds to having found a representation for which the Zeeman interaction is diagonal. For the general case for which the Zeeman interaction is not diagonal, we revert to Eq. (52) which cannot in general be reduced to quadratures. However, we can show that once a solution for D^+ has been found the Zeeman interaction can be diagonalized. For, knowing $D_\pm$ and D_z we can take the states

$$\psi_\uparrow=(1+yy^*)^{-1}\begin{pmatrix}1 \\ -y\end{pmatrix}; \quad \psi_\downarrow=(1+yy^*)^{-1}\begin{pmatrix}y^* \\ 1\end{pmatrix}, \quad (59)$$

where $y=D_z/D_-=-D^+/D_z$, the latter equality from Eq. (49). It is readily shown that $\boldsymbol{\sigma}\cdot\mathbf{D}\psi_\uparrow$ vanishes, and that $\boldsymbol{\sigma}\cdot\mathbf{D}\psi_\downarrow$ gives a multiple of $\psi_\uparrow$. Let us now transform to a system in which $\psi_\uparrow$ and $\psi_\downarrow$ become

$$\begin{pmatrix}1 \\ 0\end{pmatrix} \quad\text{and}\quad \begin{pmatrix}0 \\ 1\end{pmatrix},$$

respectively. The transformation matrix T is to be unitary, and we shall assume it to be a symmetrized function of κ. Let us write to first order in h

$$T=T_0+hT_1, \quad (60)$$

where

$$T_0=(1+yy^*)^{-1}\begin{pmatrix}1 & y^* \\ -y & 1\end{pmatrix}, \quad (61)$$

which is unitary ignoring noncommutativity, and where T_1 corrects this to first order in h, and need not be stated explicitly. We now transform $\mathcal{E}(\kappa)$ using the
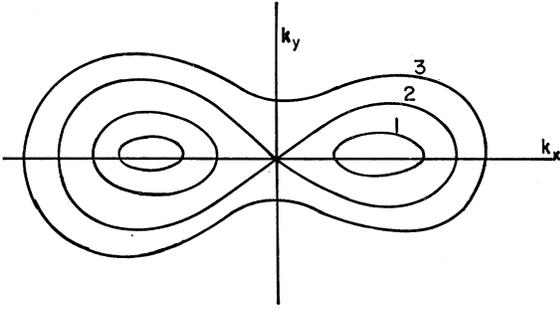
FIG. 2. Magnetic orbits in the vicinity of a self-intersecting "figure 8" orbit. B is in the z direction.

multiplication rule of Eq. (7), to first order in h. Thus

$$T_0^{-1}\mathcal{E}(\mathbf{\kappa})T_0 \equiv [\mathcal{E}(\mathbf{k})\{1 - ih\cdot\nabla_\mathbf{k}T_0^+\times\nabla_\mathbf{k}T_0\}$$
$$- \tfrac{1}{2}iT_0^{-1}(\partial T_0/\partial t) + \tfrac{1}{2}(i\partial T_0/\partial t)^{-1}T_0]_{\mathbf{k}\to\mathbf{\kappa}}, \quad (62)$$

where we have put in the t derivative as in Eq. (52). The second term in the curly brackets must be exactly canceled by T_1 if T is to be unitary, since the right side must give 1 if $\mathcal{E}(\mathbf{\kappa})$ is replaced by 1. The last two terms are equal since $(\partial/\partial t)(T_0^{-1}T_0) = 0$. We also transform $\mathbf{W}\cdot\mathbf{s}$, to lowest order in h, giving altogether a new Zeeman interaction

$$\mathbf{W}'\cdot\mathbf{s} = T_0^{-1}\mathbf{W}\cdot\mathbf{s}T_0 - iT_0^{-1}(\partial T_0/\partial t). \quad (63)$$

It is straightforward now to obtain

$$W_z' = W_z - \tfrac{1}{2}(W_-y + W_+y^*), \quad (64)$$

with $W_x' = W_y' = 0$. We can also transform $\boldsymbol{\sigma}\cdot\mathbf{D}$ into $\boldsymbol{\sigma}\cdot\mathbf{D}'$, where $\mathbf{D}'$ has only the component

$$D_-' = D_-(1 + yy^*). \quad (65)$$

Summing up, the Zeeman interaction in a crystal with inversion symmetry can be reduced to a diagonal form provided the equations of motion, Eq. (47) or (52), have been solved. Thus we are led again to Eqs. (1) and (42) for the levels. We should note however that the transformation of Eq. (63), should really be carried out to second order in h, as $\mathcal{3C}_2$, Eq. (38) will now be modified. We shall not carry this through but in the following section we shall calculate the susceptibility assuming that the correct $\mathcal{3C}_2$ has been obtained.

4. Self-Intersecting Orbits

In this section we shall discuss the generalization of the results of Sec. 1 to the case of orbits which are close to saddle points. For this case we must use another method, as our present scheme breaks down. The simplest example of such an orbit is the "figure-eight" orbit structure shown in Fig. 2, which has been considered by Azbel,[18] who used the extension of the WKB

[18] M. Ya. Azbel, Zh. Eksperim. i Teor. Fiz. **39**, 1276 (1960) [English transl.: Soviet Phys.—JETP **12**, 891 (1961)].

method for the case in which the energy is nearly equal to the height of the barrier.[19]

For energies close to $\mathcal{E}_2$, the energy of the self-intersecting orbit, the area has an infinite slope. In fact for energies slightly above $\mathcal{E}_2$, (assuming energy increases outward) we have

$$A \sim - (4eH/c)x\ln|x|, \quad (66)$$

where $x = (\mathcal{E} - \mathcal{E}_2)(c/2eH)(\mathcal{E}_{xx}\mathcal{E}_{yy})^{1/2}$, and the energy derivatives are evaluated at $\mathbf{k} = 0$ in Fig. 2 with axes chosen so that $\mathcal{E}_{xy} = 0$. For energies below $\mathcal{E}_2$, the singularity is equally divided between the two orbits. The cyclotron frequency goes to zero as $\mathcal{E} \to \mathcal{E}_2$, since $\partial A/\partial\mathcal{E}$ has a logarithmic divergence.

By the use of the proper connection formulas, Azbel obtained the quantization condition

$$\cos\left\{\frac{c}{2eB}(A_1 + A_2) + \varphi\right\}$$
$$= -[e^{4\pi x} + 1]^{-1/2}\cos\frac{c}{2eB}(A_1 - A_2), \quad (67)$$

where

$$\varphi = \varphi_0 = 2x\ln|x|/e + i\ln\frac{\Gamma(\tfrac{1}{4} + ix)}{\Gamma(\tfrac{1}{4} - ix)} - \tan^{-1}\tanh\pi x, \quad (68)$$

and where A_1 and A_2 are the areas to the left and right of the line $k_x = 0$, respectively. The area includes the singularity, which we might note is just subtracted off by the first term in φ. Equation (67) shows how the levels below the orbit $\mathcal{E}_2$ go smoothly into those above it over a range of energy, $(eH/c)(|\mathcal{E}_{xx}\mathcal{E}_{yy}|)^{1/2}$.

The interesting feature of this calculation for our present discussion is the fact that for large values of x, φ goes as $-1/48x$, which can be shown from the asymptotic expansion of the gamma function.[20] However, if we calculate Γ according to our methods, Eq. (38), we find that near the singularity

$$\begin{aligned}2\pi\Gamma_1h &\sim 1/96x \quad x < 0 \\ &\sim 1/48x \quad x < 0,\end{aligned} \quad (69)$$

provided that we take $\Gamma_1 = 0$. The difference in Γ_1 between the two signs of x comes in because of the change from two orbits to one. Upon examining the limiting values of Eq. (66) as $x \to \infty$ $(+\infty)$, we see that φ goes into $-4\pi\Gamma_1h$ $(-2\pi\Gamma_1h)$, with $\Gamma_0 = \tfrac{1}{2}$. Thus we see that it is consistent to take $\Gamma_1 = 0$ above as well as below the singularity.

If we combine the results of the present analysis with those of Azbel, the more general quantization condition can be obtained which is Eq. (67) with φ

[19] P. M. Morse and H. Feshback, *Methods of Theoretical Physics* (McGraw-Hill Book Company, Inc., New York, 1953), pp. 1103, 1399, 1567–8.

[20] E. Jahnke, F. Emde, and F. Losch, *Tables of Higher Functions* (McGraw-Hill Book Company, Inc., New York, 1960), p. 5.

replaced by

$$\varphi = \varphi_0 - \binom{4}{2}\pi\Gamma_1 h + \frac{1}{48x}, \quad x \to \pm\infty, \quad (70)$$

with φ_0 given by Eq. (68). Thus we include Γ_1 in our equation but, near the singularity, we replace the singular part of Γ_1 by Eq. (68).

Azbel also considers the case of a "dimple." Other analogous situations which can arise are the transition from electron to hole orbits, and from open to closed orbits. The latter case is discussed in the next section. The conclusion of the present section is that it is probably correct to take $\bar{\Gamma}_0 = \frac{1}{2}$ and $\bar{\Gamma}_1 = 0$ for all closed orbits except in the immediate vicinity of self-intersecting orbits.

5. Open Orbits

It is well known[8] that another type of cyclotron orbit is possible in a solid, namely an open orbit. If we extend $\mathcal{E}(\mathbf{k})$ as a periodic function in reciprocal space, such orbits are never closed, as can be seen by the example in Fig. 3 (orbit 3). The energy levels connected with such orbits are continuous, and the subject of this section is to exhibit such continuous levels in the present formalism.

Actually Fig. 3 represents the special case in which the magnetic field is parallel to a crystal axis. In this case for constant k_z the total orbit structure is doubly periodic in $\mathbf{k}$ space and the open orbit is periodic. A somewhat more general case occurs if the magnetic field is along any lattice vector; we shall refer to this as a rational direction. The constant k_z plane then tilts through several cells in reciprocal space before the orbit structure repeats itself. That is, the orbit structure is still doubly periodic but with a larger two-dimensional unit cell. The definition of k_z has now become a little ambiguous since through one Brillouin zone there will be a set of k_z planes. The treatment of Blount[3] helps to clarify the situation. Blount redefines the unit cell of the crystal so that the magnetic field is along one side of the unit cell. The corresponding reciprocal lattice
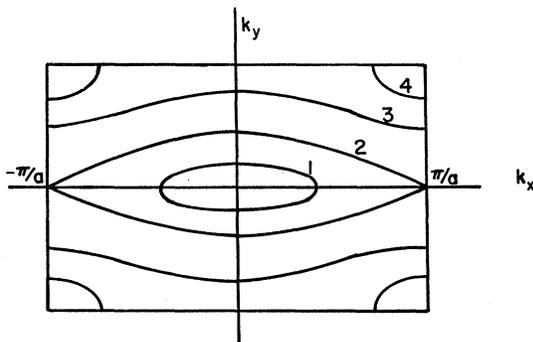

FIG. 3. Magnetic orbits for an orthorhombic crystal, with field in the z direction, showing closed-electron (1) and hole (4) orbits, an open orbit (3) and a self-intersecting orbit (2).
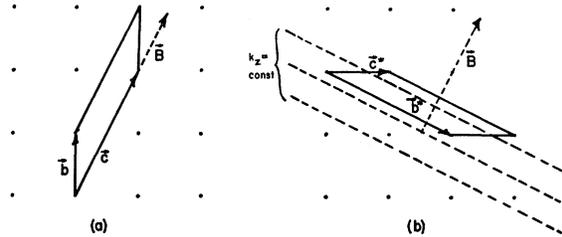

FIG. 4. Redefinition of unit cell according to Blount (Ref. 3) to give magnetic unit cell for a cubic lattice with **a** axis out of the page: (a) real lattice, (b) reciprocal lattice. A typical set of $k_z =$ const. planes is shown, and we see that the reciprocal lattice magnetic unit cell has only one of these planes through it, whereas the original cubic lattice has three.

unit cell can now be defined so that two of the axes are perpendicular to the magnetic field so that the repetition area is again the cross section of the unit cell as in Fig. 3. The scheme is illustrated in Fig. 4. If the primative lattice vector along the field is rather long, the new reciprocal-lattice unit cell becomes rather flat until in the limit of an irrational direction it becomes a plane or a strip.[3] We shall deal here with rational directions[21–23] and periodic open orbits, and shall regard the irrational directions as limiting cases.

We consider first a set of magnetic translation operators[22] which under certain circumstances commute with the Hamiltonian. These can be defined in terms of an operator $\mathbf{P}^0$, which is given for a gauge in which $\mathbf{A}$ is a linear function of $\mathbf{r}$ (i.e., $\mathbf{A} = \mathbf{r} \cdot \nabla \mathbf{A}$) by

$$\mathbf{P}^0 = \mathbf{p} + e/c\nabla\mathbf{A} \cdot \mathbf{r}, \quad (71)$$

where $\mathbf{p} = (1/i)\nabla$. The magnetic translation operator for a displacement $\boldsymbol{\varrho}$ is then defined as

$$T_\rho = \exp(i\boldsymbol{\varrho} \cdot \mathbf{P}^0). \quad (72)$$

The operators $\mathbf{P}^0$ and T_ρ can readily be shown to commute with the kinetic-momentum operator $\mathbf{P} = \mathbf{p} + e\mathbf{A}/c$ of Eq. (4). T_ρ thus commutes with the kinetic energy part of the Hamiltonian, but only commutes with the periodic potential when $\boldsymbol{\varrho}$ is a lattice vector $\mathbf{R}$.

The operator $\mathbf{P}^0$ can be related to "orbit-center" coordinates.[24,25] For, subtracting $\mathbf{P}^0$ from $\mathbf{P}$ we obtain

$$(\mathbf{P} - \mathbf{P}^0) = e/c\mathbf{B}\times\mathbf{r}, \quad (73)$$

[21] The introduction of special "rational" values of the magnitude of the magnetic field, for which a rational fraction of a flux quantum $(2\pi\hbar c/e)$ goes through a unit cell, as in the work of Brown (Ref. 22), is not necessary in our approximation. This is related to the fact that open orbits extend in one direction only. For two-dimensional orbit arrays (Ref. 23) sometimes involved in magnetic breakdown, the rationality of the magnitude of the field is important.

[22] E. Brown, Phys. Rev. **133**, A1038 (1964).

[23] A. B. Pippard, Phil. Trans. Roy. Soc. London **A256**, 317 (1964); W. G. Chambers, Phys. Rev. **140**, A135 (1965).

[24] R. Kubo, N. Hashitsume and S. J. Miyake, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1965), Vol. 17.

[25] M. H. Johnson and B. A. Lippman, Phys. Rev. **76**, 828 (1949).

and crossing this into $\mathbf{B}c/eB^2$, we have

$$r_{\perp} = (\mathbf{P} - \mathbf{P}^0) \times \mathbf{B}c/eB^2. \qquad (74)$$

The right-hand side of this equation can be interpreted as the sum of a relative coordinate $\mathbf{P} \times \mathbf{B}c/eB^2$ and an orbit center coordinate $\mathbf{r}_0 = -\mathbf{P}^0 \times \mathbf{B}c/eB^2$. The x and y components of the orbit-center coordinate act like a canonical coordinate and momentum pair with the commutation relations[25]

$$[x_0, y_0] = c/ieB. \qquad (75)$$

Because of the commutation relations, y_0 is proportional to the generator for a translation of x_0, and vice versa, so that our translation operator T_ρ translates the orbit-center coordinate by ϱ.

We should remark that in the case of free electrons for which x_0 and y_0 commute with the Hamiltonian, the degeneracy of levels can be obtained by confining x_0 and y_0 to the area $L_x L_y$ of the crystal, and noting that the number of levels in the $(eB/c)^{1/2}x_0$, $(eB/c)^{1/2}y_0$ phase space is given by $1/2\pi$ times the area, so that each level has a degeneracy $eBL_x L_y/2\pi c$.

Let us now consider the momentum-space operator corresponding to T_ρ,

$$T_\rho' = \exp(i\varrho \cdot \mathbf{\kappa}^0), \qquad (76)$$

where

$$\mathbf{\kappa}^0 = \mathbf{k} + e/c\nabla\mathbf{A} \cdot i\nabla_\mathbf{k}. \qquad (77)$$

The operators $\mathbf{\kappa}^0$ and T_ρ' commute with $\mathbf{\kappa}$. But since $\mathfrak{IC}$ is a function of $\mathbf{\kappa}$, T_ρ' commutes with $\mathfrak{IC}$, no matter what ϱ is. However, only for ϱ equal to a lattice vector is T_ρ' periodic in $\mathbf{k}$ space. Thus we see that we must impose periodicity in $\mathbf{k}$ space in order to obtain the same physical information we had before.

Since the operators $T_\mathbf{R}'$, where $\mathbf{R}$ is a lattice vector, commute with $\mathfrak{IC}$ and are periodic in $\mathbf{k}$, if we have an eigenfunction ψ, then $T_\mathbf{R}'\psi$ is also an eigenfunction with the same energy. We are interested however in raising operators C^+ for which $C^+\psi$ is an eigenfunction with a higher energy. We have previously assumed that C^+ was a function of $\mathbf{\kappa}$ alone. Let us now consider the more general function

$$C^+ = C_\rho^+(\mathbf{\kappa})T_\rho', \qquad (78)$$

in which we have a product of a magnetic translation operator and function of $\mathbf{\kappa}$. We now require that C^+ be periodic in $\mathbf{\kappa}$, that is,

$$C^+(\mathbf{\kappa} + \mathbf{K}) = C^+(\mathbf{\kappa}), \qquad (79)$$

where $\mathbf{K}$ is a vector of the reciprocal lattice. But since

$$T_\rho'(\mathbf{k} + \mathbf{K}) = \exp[i\mathbf{K} \cdot \varrho]T_\rho'(\mathbf{\kappa}), \qquad (80)$$

we must have

$$C_\rho^+(\mathbf{\kappa} + \mathbf{K}) = \exp[-i\mathbf{K} \cdot \varrho]C_\rho^+(\mathbf{\kappa}). \qquad (81)$$

The raising operator must again obey Eq. (9), but, since T_ρ' commutes with $\mathfrak{IC}$, we can remove it from the equation, giving

$$[\mathfrak{IC}, C_\rho^+(\mathbf{\kappa})] = C_\rho^+(\mathbf{\kappa})\omega_0(\mathfrak{IC}), \qquad (82)$$

which has the same form as Eq. (9). We can now use the same arguments to obtain, to lowest order in B,

$$C_\rho^+ = e^{i\psi}, \qquad (83)$$

$$\psi = (\omega_0 c/eH)\int^\mathbf{k} d\lambda/v_\perp. \qquad (84)$$

Suppose we consider the case of Fig. 3 in which we assume for simplicity that the unit cell axes are at right angles. For closed orbits, such as orbit 1 we can again use the condition that C^+ be single valued to obtain the quantization condition, Eq. (1). The condition of Eq. (82) for extending ψ to other zones can be met by simply defining C^+ to have a different phase factor in each unit cell. If we take $s = 0$ in Eq. (13), we have the result that C^+ effectively commutes with H for these levels, so that the levels have the same degeneracy as in the free electron case, at least in the semiclassical approximation.

For an open orbit like orbit 3 in Fig. 3, however, we see that Eq. (80) imposes the condition on ψ, for, e.g., the upper trajectory,

$$\psi(\pi/a) - \psi(-\pi/a) = 2\pi(s - \rho_x/a), \qquad (85)$$

so that

$$\omega_0 = (2\pi eH/c)\left(s - \frac{\rho_x}{a}\right)\left[\int_{-\pi/a}^{\pi/a} \frac{d\lambda}{v_\perp}\right]^{-1}. \qquad (86)$$

Since ρ_x can be arbitrarily small, we see that ω_0 can have any value, so that the levels are continuous in the semiclassical approximation.

We can now ask whether the argument of Sec. 1 can be continued to higher order in B for open orbits. In fact, it can. Suppose that we let $\psi = (s - \rho_x/a)\phi$ where

$$\phi = 2\pi\int^\mathbf{k} d\lambda/v_\perp\left[\int_{-\pi/a}^{\pi/a} d\lambda/v_\perp\right]^{-1}. \qquad (87)$$

Then, we let a be $1/4\pi$ times the area enclosed by the upper trajectory 3, the x axis, and sides of the zone. The desired extension of Eq. (19) is

$$[a - h\Gamma, C_\rho^+] = (s - \rho_x/a)hC_\rho^+. \qquad (88)$$

Equation (27) now becomes

$$i(\partial C_\rho^+/\partial\phi) + (s - \rho_x/a)C_\rho^+ = 0, \qquad (89)$$

and the left sides of the following equations are similar. By entirely analogous reasoning, we find $\Gamma_0 = \bar{\Gamma}_0 = $ const and

$$\Gamma_1 = \bar{\Gamma}_1 + \frac{1}{48\pi}\frac{\partial}{\partial\mathcal{E}}\int_{-\pi/a}^{\pi/a}(d\lambda/v_\perp)(\mathcal{E}_{11}\mathcal{E}_{22} - \mathcal{E}_{12}^2). \qquad (90)$$

We now have no direct way of evaluating $\bar{\Gamma}_0$ and $\bar{\Gamma}_1$. We can, however, construct a bridge between the open orbits and closed orbits by considering the region near the self-intersecting orbit 2 of Fig. 3. The calculation is of course interesting in itself, and is similar to that of the last section. The result has not to this author's knowledge been published, but some unpublished work by Kohn has been discussed by Blount,[3] and the result has been obtained in an unpublished calculation by Hohenberg and the author. The quantization condition for energies close to that of the self-intersecting orbit is given by

$$\cos\left(\frac{c}{2eB}A+\varphi\right)=[1+e^{-4\pi x}]^{-1/2}\cos(2\pi x_0/a), \quad (91)$$

where φ is given by Eq. (67) with x defined below Eq. (68) and with the energy derivatives evaluated again at the saddle point. x_0 is the x component of the "orbit-center" coordinate which is a good quantum number, and A is the area which goes into the area of orbit 1.

If we let $x \to -\infty$ in Eq. (91), the right side goes to zero and we obtain the generalized Onsager condition with $\bar{\Gamma}_0=\frac{1}{2}$, $\bar{\Gamma}_1=0$ and $2\Gamma_1 h \sim 1/48\pi x$, all as in Sec. 4. As x goes toward zero from below, the levels begin to broaden. If we let $x \to =\infty$, the gaps between the broadened levels close up and the levels become continuous. The quantization condition becomes

$$A/2=2\pi eB/c\left(n\pm\frac{x_0}{a}+\frac{1}{96\pi x}\right). \quad (92)$$

The appropriate area here is $A/2$, and the two signs refer to the upper and lower trajectories. The last term in the parentheses can be interpreted as $2\Gamma_1 h$, consistent with Eq. (90). The result shows that we should take $\bar{\Gamma}_0=\bar{\Gamma}_1=0$ for open orbits.

Part II: MAGNETIC SUSCEPTIBILITY OF BLOCH ELECTRONS

6. Basic Derivation

We now apply the results of part I to calculate the magnetic susceptibility and de Haas–van Alphen effect. We shall assume that we are dealing with a band, or region of a band, with closed orbits and for which the the energy is an increasing function of area, i.e., electron orbits. We also assume that the conditions for $\bar{\Gamma}_0=\frac{1}{2}$ and $\bar{\Gamma}_1=0$ Eq. (37) are met. In the next section we shall discuss the extension to holes states, and to more general orbits. We begin with the expression for the free energy

$$F=N\zeta-kT\sum_i\phi(\mathcal{E}_i);$$

$$\phi=\ln(1+\exp-(\mathcal{E}_i-\zeta)/kT), \quad (93)$$

where ζ is the Fermi energy. In terms of F, the magnetic moment is given by

$$M=-(\partial F/\partial B). \quad (94)$$

The degeneracy of the Landau levels is the same as in the free-electron case, as we have shown in Sec. 5, so that we have (for unit volume),

$$\sum_i \to (h/2\pi^2)\int dk_z \sum_{n\mu}. \quad (95)$$

Here, k_z is the wave vector in the magnetic-field direction, which we have thus far left suppressed, and μ is summed over the spin states. The levels are given by Eq. (1) which we can rewrite as

$$a(\mathcal{E}_{n\mu},k_z)=(n+\gamma_\mu(\mathcal{E}_{n\mu},k_z))h, \quad (96)$$

with γ given to first order in B by Eq. (42).

We now apply the Poisson summation formula[26] in the form

$$\sum_{n=0}^{\infty} f(n)=\sum_{r=-\infty}^{\infty}\int_{n_0}^{\infty} dn\, e^{2\pi i n r}f(n); \quad -1<n_0<0. \quad (97)$$

It is convenient to take $n_0=-\frac{1}{2}$. We then have

$$F-N\zeta=-\frac{kTh}{2\pi^2}\int dk_z \sum_\mu \int_{-1/2}^{\infty} dn\phi(\mathcal{E}_{n\mu})$$

$$-\frac{kTh}{\pi^2}\int dk_z \sum_{\mu,r>0} \mathrm{Re}\int_{-1/2}^{\infty} dn\, \phi(\mathcal{E}_{n\mu})e^{2\pi i n r}, \quad (98)$$

where we have written the $r=0$ term separately. The upper limit is taken to be ∞, as we assume that ϕ vanishes sufficiently far above the Fermi level. The second term can be integrated by parts to give

$$\frac{kTh}{\pi^2}\int dk_z\left\{\sum_{\mu,r>0}\mathrm{Re}\int_{-1/2}^{\infty}dn\frac{\partial\phi}{\partial\mathcal{E}_{n\mu}}\frac{\partial\mathcal{E}_{n\mu}}{\partial n}\frac{e^{2\pi i n r}}{2\pi i r}\right.$$

$$\left.-\sum_{\mu,r>0}\mathrm{Re}\phi(\mathcal{E}_{n\mu})\frac{e^{2\pi i n r}}{2\pi i r}\Big|_{n=-1/2}^{\infty}\right\}. \quad (99)$$

The boundary term here vanishes, and in the remaining term, we can use

$$\partial\phi/\partial\mathcal{E}=-(1/kT)f_0(\mathcal{E}), \quad (100)$$

where f_0 is the Fermi function. We now suppose that we can continue the integrand into the complex n plane, and that the only singularities are poles of f_0. Then we can add and subtract the integral along the positive imaginary axis, and so separate the result into a steady

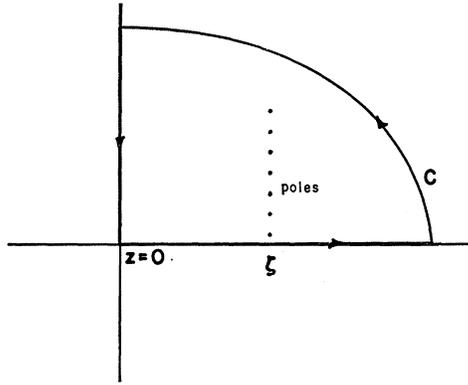[26] P. M. Morse and H. Feshbach, Ref. 19, p. 467.

FIG. 5. Contour integral for Eq. (103).

part F_1 and an oscillatory part F_2. The result is

$$F = N\zeta + F_1 + F_2;$$ (101)

$$F_1 = \frac{kTh}{2\pi^2} \sum_\mu \int dk_z \int_{1/2}^\infty dn\, \phi(\mathscr{E}_n)$$

$$-\frac{h}{\pi^2} \sum_{\mu,r>0} \mathrm{Re} \int dk_z \int_{-1/2}^{-1/2+i\infty} dn f_0 \frac{\partial \mathscr{E}_{n\mu}}{\partial n} \frac{e^{2\pi i n r}}{2\pi i r};$$ (102)

$$F_2 = -\frac{h}{\pi^2} \sum_{\mu,r>0} \mathrm{Re} \int dk_z \int_C dn f_0 \frac{\partial \mathscr{E}_{n\mu}}{\partial n} \frac{e^{2\pi i n r}}{2\pi i r}.$$ (103)

The contour C is shown in Fig. 5. It can be closed in the upper half-plane so that only the poles contribute. These are near $n(\zeta)$, so that F_2 is indeed oscillatory.

Let us first calculate F_1, to second order in h. We let $x = (n+\tfrac{1}{2})h$ and evaluate $\mathscr{E}(x)$ to second order in the form

$$a(\mathscr{E}) = x + \gamma_\mu' h,$$ (104)

where $\gamma_\mu' = \gamma_\mu - \tfrac{1}{2}$. The result for $\mathscr{E}$ is

$$\mathscr{E}_\mu = \mathscr{E}_\mu^0 + (\gamma_\mu'/a')h + d/d\mathscr{E}(\gamma_\mu'^2/2a')h^2.$$ (105)

Now for the first term of Eq. (102), we have

$$\int_0^\infty dx\phi(\mathscr{E}_\mu) = \int_0^\infty dx \left\{ \phi(\mathscr{E}_\mu^0) \right.$$

$$+ \frac{\partial \phi}{\partial \mathscr{E}_\mu^0} \left(\frac{\gamma_\mu'}{a'}h + \frac{1}{a'} \frac{\partial}{\partial \mathscr{E}} \frac{\gamma_\mu'^2}{2a'}h^2 \right)$$

$$\left. + \frac{1}{2} \frac{\partial^2 \phi}{\partial(\mathscr{E}_\mu^0)^2} \left(\frac{\gamma_\mu'}{a'}h \right)^2 \cdots \right\}$$ (106)

$$= \int_0^\infty dx\phi(\mathscr{E}_\mu^0) - \frac{1}{kT} \int_0^\infty d\mathscr{E} \left\{ f_0\gamma_\mu'h \right.$$

$$\left. + \frac{1}{2} \frac{\partial}{\partial \mathscr{E}} \left(f_0 \frac{\gamma_\mu'^2}{a'}h^2 \right) \right\},$$ (107)

where we have changed from the variable x (= a for zero field) to $\mathscr{E}$, and have used Eq. (100). In the second term of (102), we integrate by parts once, and keep only

the boundary terms as the remainder is higher order in h:

$$\sum_{r>0} \int_0^{i\infty} dx f_0 \frac{\partial \mathscr{E}}{\partial x} \frac{e^{2\pi i r}[x/h - 1/2]}{2\pi i r}$$

$$\cong \sum_{r=1}^\infty \frac{h(-1)^r}{(2\pi r)^2} f_0 \frac{d\mathscr{E}}{dx}\bigg|_{x=0} = -hf_0/48a'\bigg|_{\mathscr{E}=0},$$ (108)

where we have used the result[27]

$$\sum_1^\infty (-1)^{r+1}/r^2 = \pi^2/12$$

and converted from x to $\mathscr{E}$ as a variable. We now have for F_1

$$F_1 - (F_1)_{h=0} = 2\pi^2 \int dk_z \sum_\mu \left\{ \left[\int_0^\infty d\mathscr{E} f_0 \gamma_\mu' h \right. \right.$$

$$\left. \left. - \frac{\gamma_\mu'^2 h^2 f_0}{2a'} \right|_{\mathscr{E}=0} + \frac{1}{24} \frac{f_0 h^2}{a'} \right|_{\mathscr{E}=0} \right\}.$$ (109)

We now evaluate the γ's from Eq. (64), remembering to omit the $\tfrac{1}{2}$. The first term of Eq. (109) is then second order in h, and involves

$$\sum_\mu \int_0^\infty d\mathscr{E} f_0 \gamma_\mu' h = 2h^2 \int_0^\infty d\mathscr{E} f_0 \left[\oint \frac{d\lambda}{4\pi v_\perp} H_2 \right.$$

$$\left. + \frac{\partial}{\partial \mathscr{E}} \oint \frac{d\lambda}{4\pi v_\perp} \{ \tfrac{1}{6}(\mathscr{E}_{11}\mathscr{E}_{22} - \mathscr{E}_{12}{}^2) - \tfrac{1}{2}H_1{}^2 \} \right].$$ (110)

Now integrating the second term of this equation by parts, the derivative is transferred to the f_0, and we are left with a boundary term. It is easy to show that this boundary term exactly cancels the second two terms of Eq. (109). The remaining integrals can be changed into integrals over $\mathbf{k}$ by Eq. (16). Finally, we obtain

$$F_1 - (F_1)_{h=0} = \frac{h^2}{4\pi^3} \int d\mathbf{k} \left[+f_0 H_2 \right.$$

$$\left. + \frac{\partial f_0}{\partial \mathscr{E}} \{ -\tfrac{1}{6}(\mathscr{E}_{11}\mathscr{E}_{22} - \mathscr{E}_{12}{}^2) + \tfrac{1}{2}H_1{}^2 \} \right].$$ (111)

This result is in agreement with similar expressions obtained by other methods.[2–5]

We now turn to Eq. (103), which reduces to the sum of the residues of the poles shown in Fig. 5. These poles lie inside the contour as long as $\mathrm{Re}\, n > -\tfrac{1}{2}$. When all the poles are outside this region, the integral vanishes, so that this sets the limit on k_z. We shall assume we can merely take $n(\mathscr{E}_F) > -\tfrac{1}{2}$. The poles of f_0 are at $\mathscr{E} - \zeta = \pm(2m+1)\pi i kT$, with m a non-negative integer. Since $\mathscr{E}$ is an increasing function of n, the poles with plus signs lie within the contour, at least for small enough m.

[27] Jahnke et al., Ref. 20, p. 40.

Therefore

$$F_2 = \frac{kTh}{\pi^2} \sum_{\mu,r>0} \frac{1}{r} \text{Re} \int_{n(\zeta)>-(1/2)} dk_z$$

$$\times \sum_{m=0}^{\infty} \exp\{2\pi i r n_\mu(\zeta + [2m+1]\pi i k T)\}. \quad (112)$$

We now approximate by expanding n about ζ, and keeping the linear term in T. We expect the next order term to be of order kT/ζ, so that the assumption here is $kT/\zeta \ll 1$. Carrying out the sum over m, we have

$$F_2 \cong \frac{kTh}{\pi^2} \sum_{\mu,r>0} \frac{1}{r} \text{Re} \int_{n(\zeta)>-(1/2)} dk_z$$

$$\times \frac{\exp[2\pi i r n_\mu(\zeta)]}{2 \sinh(2\pi^2 r k T (\partial n/\partial \zeta))}. \quad (113)$$

Now the integral over k_z can be evaluated by noting that the exponential is a rapidly oscillating function since $n \sim a/h$, so that the major contribution comes from an extremal area.[1] Expanding n to second order in k_z and carrying out the Gaussian integral, we have

$$F_2 \cong \frac{kTh}{2\pi^2} \sum_{\mu,r>0} \frac{1}{r} \text{Re} \left| \frac{1}{r(d^2n/dk_z^2)} \right|_m^{1/2}$$

$$\times \frac{\exp[2\pi i r n_{\mu m}(\zeta) \mp \pi i/4]}{\sinh(2\pi^2 r k T (\partial n/\partial \zeta))}, \quad (114)$$

where the subscript m means extremal and the $\mp$ depends on the type of extremum, the upper sign applying to a maximum.

We can now use our expression Eq. (96) to evaluate n, giving finally.

$$F_2 = \frac{kT}{2\pi^2} \left(\frac{eB}{c}\right)^{3/2} \sum_{r>0} \frac{\cos\{[(\gamma_+ - \gamma_-)\pi r] \cos\{(rAc/eB) - \pi r(\gamma_+ + \gamma_-) \mp \pi/4\}}{r^{3/2} |(1/2\pi)(d^2A/dk_z^2)|_m^{1/2} \sinh(2\pi^2 r k T/\omega_0)}, \quad (115)$$

where we have reverted to the original notation, and have omitted γ except in the oscillatory part. This leads directly to the desired result for the de Haas–van Alphen effect, in essential agreement with previous work.[1] The de Haas–van Alphen period is $\Delta(1/B) = 2\pi e/Ac = e/2ac$. The first cosine factor is due to the g factor. For free electrons with spin, this would give a minus sign and simply reverse the phase of the effect. This is believed to occur for Bi electrons[17] where the spin splitting is equal to the Landau level spacing. The amplitude depends on temperature and field through the sinh factor. We should actually modify Eq. (115) to include lifetime broadening effects[24,28,29] by multiplying the rth component by the Dingle factor $\exp(-2\pi^2 n k T'/\omega_0)$, where T' is an effective temperature.

7. Change in de Haas–van Alphen Period with Field

Since we have calculated the dependence of γ on h to first order, it is appropriate to consider the change in de Haas–van Alphen period with magnetic field. As in Sec. 6, we limit ourselves to the case of closed electron orbits. It is important to include here the effect of the change in Fermi level with magnetic field, which will affect the area function. (We assume here that the number of carriers is constant.) We replace the usual area function by

$$\bar{a}(\zeta_0) = a(\zeta_0) + (\partial a/\partial \zeta)(\zeta - \zeta_0) - 2\gamma_1 h^2. \quad (116)$$

Here, γ_1 is the first order part of γ from Eq. (42). In order to calculate the change in Fermi level, we use the

relationship

$$dF/d\zeta = 0 \cong (d/d\zeta)[N\zeta + F_1] \quad (117)$$

to preserve the number of particles. Here we have omitted the oscillatory term in determining the Fermi level. The latter will affect only higher harmonics. Now, using the definition of the susceptibility, we have

$$N = -(d/d\zeta)[F_1(\zeta)|_{h=0} - \tfrac{1}{2}xB^2]$$
$$= N_0(\zeta) + \tfrac{1}{2}dx/d\zeta B^2 = N_0(\zeta_0). \quad (118)$$

Therefore, to lowest order in B,

$$\zeta - \zeta_0 = -\frac{\tfrac{1}{2}(dx/d\zeta)B^2}{dN_0/d\zeta}. \quad (119)$$

Using Eqs. (116) and (119), we have

$$\bar{a} - a = \frac{h^2 da/d\zeta}{(d/d\zeta) \int d\mathbf{k} f_0} \frac{d}{d\zeta} \int d\mathbf{k}$$

$$\times \left[f_0 \mathfrak{K}_2 - \frac{\partial f_0}{\partial \mathcal{E}} \{ \tfrac{1}{6}(\mathcal{E}_{11}\mathcal{E}_{22} - \mathcal{E}_{12}{}^2) - \tfrac{1}{2}\mathfrak{K}_1{}^2 \} \right]$$

$$- h^2 \left[\oint \frac{d\lambda}{4\pi v_\perp} \mathfrak{K}_2 + \frac{\partial}{\partial \zeta} \oint \frac{d\lambda}{4\pi v_\perp} \right.$$

$$\left. \times \{ \tfrac{1}{6}(\mathcal{E}_{11}\mathcal{E}_{22} - \mathcal{E}_{12}{}^2) - \tfrac{1}{2}\mathfrak{K}^2 \} \right]_m. \quad (120)$$

Several things can be noticed about this expression. It depends on the difference between quantities on the extremal orbit and on averages over k_z. If we have a constant H_2 term, this does not contribute. However, for the free-electron case, there is a contribution from

[28] R. B. Dingle, Proc. Roy. Soc. (London) **A211**, 517 (1952).
[29] L. M. Roth and P. N. Argyres, in *Semiconductors and Semimetals*, edited by R. Willardson and A. C. Beer (Academic Press Inc., New York, 1966), Vol. 1.

the first term, though not the second. Evaluating this we find

$$(\bar{a}-a)_{\text{free}} = \hbar^2/24a = a[1/24(n+\gamma)^2], \quad (121)$$

which gives the relative change in period, which is proportional to $1/a$. This result applies also to ellipsoids. We see that the correction term in Eq. (121) is unimportant except for small n, where the theory breaks down anyway. We can expect a similar situation to apply more generally.

We can also calculate an extra field dependence for the amplitude of the oscillations, which depends primarily upon the sinh term. The dependence is through the inverse cyclotron frequency, $\omega_c^{-1} = \hbar^{-1}\partial\bar{a}/\partial\varphi$, where $\bar{a}$ is given by Eq. (120).

8. Extension to More General Orbits and a Possible New Effect

We now discuss the extension of the susceptibility calculation to more general orbits. The discussion is necessarily rather qualitative as we shall find that questions are raised which would take some labor to answer precisely.

The simplest extension is that to the case of hole orbits in an almost filled band. For this case if we let $\mathcal{E}' = -\mathcal{E}$, $\zeta' = -\zeta$, we can rewrite F as

$$F = \sum_i \mathcal{E}_i + N_{\text{h}}\zeta'$$
$$\quad - kT \sum_i \ln(1+\exp[-(\mathcal{E}_i'-\zeta')/kT]), \quad (122)$$

where the sum i goes over the one band. The first term is the zero degree free energy for the filled band, and the last two give Eq. (93) but for holes, with N_{h} the hole density. The result from the second two terms will just give Eq. (115) again for the oscillations, if we keep $\omega_0 > 0$. The steady result from these two terms just subtracts the hole part from the filled band normal susceptibility as calculated by the usual methods. Thus the result agrees with previous work provided that we assume that the first term of Eq. (122) indeed gives rise to the usual filled-band normal susceptibility. This is not obvious since in going from the bottom of the band to the top we always encounter self-intersecting orbits and usually open orbits, as can be seen in Fig. 3, and our derivation of the susceptibility is not valid for these.

The calculation of the susceptibility in the vicinity of open orbits is considerably simplified by the fact that the levels are continuous. For the example of Sec. 5, the sum over states involves an integration over x_0, the orbit center x component, from Eq. (92). For one unit cell we can integrate x_0 from $-a/2$ to $+a/2$, the number of states per unit interval of x_0 being $eBL_y/4\pi^2c$ for 2 dimensions from the argument after Eq. (75) in Sec. 5. It is convenient simply to multiply this degeneracy by the number of unit cells in the x direction L_x/a, since otherwise we would have to

redefine n. The sum over states now including k_z and per unit volume is then

$$\sum \rightarrow (\hbar/2\pi^2)\int dk_z \sum_{n\mu} \int_{-1/2}^{1/2} d(x_0/a). \quad (123)$$

Now looking at Eq. (92) we see that the integration over x_0 simply turns the sum oven n into an integration so that we need keep only the $r=0$ term in the summation corresponding to Eq. (97). Thus there are, as expected, no de Haas–van Alphen oscillations for open orbits. Inclusion of the γ_1 term essentially as in Eq. (42) leads to the usual normal susceptibility, except for possible boundary terms which we have not investigated, but which are related to the discussion below.

Thus far we have found nothing unexpected. However, if we consider the case of self-intersecting orbits, there is certainly a possibility of extra contributions, even for filled bands. In the first place there will undoubtedly be nonanalytic contributions to the steady susceptibility. Since, however, the regions of k space involved are quite small for low fields these terms are probably small in an asymptotic sense. More interesting is the possibility of oscillatory contributions to the susceptibility. If we consider the "figure 8" situation of Sec. 4 we find that at the energy $\mathcal{E}_2$ two types of orbits cease to exist and a third type appears. This situation is quite analogous to there being Fermi-surface cross sections corresponding to the three areas. Thus we might expect to find oscillations with periods corresponding to A_1, A_2, and A_1+A_2. If we use the simple Onsager condition, Eq. (1), these show up in the susceptibility as additional boundary terms, e.g., in Eq. (99), since the several n summations now have finite limits. The areas of transition between open and closed orbits could also contribute periods.

In the more rigorous treatment of Sec. 4 we see from Eqs. (67)–(68) that the change over takes place over a small range of energy. Thus there is a built-in broadening which decreases the amplitude of the oscillations, and may wipe them out completely. A detailed investigation is being made to see whether there are conditions under which such oscillations could be observed. If so, they would have some unusual properties since they would not be related to the Fermi surface, and since their amplitudes would not depend on temperature. Since no experimental observations of such periods have been reported, it seems rather doubtful that they exist, but the matter deserves further study.

Finally the most general closed-electron or hole orbit is one which may have self-intersecting orbits within it (i.e., for different energies). The extension of the theory to this case gives the expected results for the de Haas–van Alphen effect and steady susceptibility, partly as a result of the expressions for $\bar{\Gamma}_0$ and $\bar{\Gamma}_1$, from Sec. 4, and with the possible addition of effects due to the self-intersecting orbits themselves as discussed above. Thus, the work of this paper essentially confirms

previous results for the susceptibility for the various types of bands.

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APPENDIX A. SYMMETRIZATION OF A FUNCTION OF $\mathcal{3C}(\kappa)$

Given a function $F(\mathcal{3C})$, we wish to obtain a symmetrized function $F_S(\kappa)$, as an expansion in powers of $h = eH/2c$. We shall obtain the result by induction, under the assumption that F can be expanded in a Taylor series in $\mathcal{3C}$. We first suppose that F_S can be expanded as follows:

$$F_S(\kappa) = F(\kappa) + hF_1(\kappa) + h^2F_2(\kappa) + \cdots . \quad (A1)$$

We can now obtain the expansion for $(FH)_S$ in terms of the coefficients of Eq. (A1) using the multiplication theorem of Eq. (7). Since $\mathcal{3C}$ commutes with F, we can use the symmetrical product $\{H,F\}_S$, and we obtain for the function of $\mathbf{k}$,

$$(F\mathcal{3C})_S = F_S\mathcal{E} + (h^2/2)\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}(F_S)_{\alpha\gamma}\mathcal{E}_{\beta\delta} + \cdots , \quad (A2)$$

using the notation of Eq. (21). Because of the symmetric product, all of the odd-order terms vanish here. Since we can take F successively as 1, $\mathcal{3C}$, $\mathcal{3C}^2$ we see that no odd order terms in h appear, so that $F_1 = 0$. For F_2 we have, from Eq. (A2),

$$\begin{aligned}(F\mathcal{3C})_2 &= F_2\mathcal{E} - \tfrac{1}{2}\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}F_{\alpha\gamma}\mathcal{E}_{\beta\delta}\\ &= F_2\mathcal{E} - \tfrac{1}{2}\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}[F'\mathcal{E}_{\alpha\gamma}\mathcal{E}_{\beta\delta} + F''v_\alpha v_\gamma \mathcal{E}_{\beta\delta}], \quad (A3)\end{aligned}$$

where the primes represent derivatives with respect to energy. We can rewrite these derivatives as

$$\begin{aligned}F' &= \tfrac{1}{2}[(F\mathcal{E})'' - F''\mathcal{E}];\\ F'' &= \tfrac{1}{3}[(F\mathcal{E})''' - F'''\mathcal{E}].\end{aligned} \quad (A4)$$

Suppose now that F_2 is given by

$$F_2 = -(1/12)\epsilon_{\alpha\beta}\epsilon_{\gamma\delta}[3F''\mathcal{E}_{\alpha\gamma}\mathcal{E}_{\beta\delta} + 2F'''v_\alpha v_\gamma\mathcal{E}_{\beta\delta}]. \quad (A5)$$

Equation (A5) is valid for $F = \mathcal{E}^2$, as can be readily verified. Also from Eqs. (A3) and (A4) if F_2 has the form (A5), $(F\mathcal{3C})_2$ has the same form. Thus the form of F_2, Eq. (A5), is established by induction. The expansion can be carried out to higher order in the same manner.

APPENDIX B

The object of this Appendix is to obtain the second term in Eq. (35) from the second term in Eq. (31). We

can write this term, except for a factor of 6π, as

$$K = \epsilon_{\alpha\beta}\epsilon_{\gamma\delta}\epsilon_{\mu\nu}\oint d\phi\, a_{\alpha\gamma\mu}$$

$$\times[-\phi_\beta\phi_{\delta\nu} - i(\phi_\beta\phi_\nu + \phi_\beta\phi_{\delta\nu} + \phi_\delta\phi_{\beta\nu}) + \phi_\beta\phi_\delta\phi_\nu]. \quad (B1)$$

In reducing this expression, we shall suppress the ϵ's, but shall use repeatedly the antisymmetry between the pairs $\alpha\beta$, $\gamma\delta$, and $\mu\nu$. Letting the first term of Eq. (B1) be K_1, we have, using Eq. (32),

$$K_1 = -\oint d\phi\, a_{\alpha\gamma\mu}\phi_\beta\phi_{\delta\nu} = -\frac{\partial}{\partial a}\oint d\phi\, a_{\alpha\gamma\mu}a_\beta\phi_{\delta\nu}. \quad (B2)$$

Differentiating by parts gives

$$K_1 = -\frac{\partial}{\partial a}\oint d\phi\{\nabla_\mu[a_{\alpha\gamma}a_\beta\phi_{\delta\nu}] - a_{\alpha\gamma}a_{\beta\mu}\phi_{\delta\nu}\}. \quad (B3)$$

The second term goes into itself if we interchange δ and ν, γ and μ and α and β. The first two of these give a plus sign from the ϵ's but the last gives a minus sign, so that the result vanishes. Let us call this "argument 1." Again applying Eq. (32) to the first term, we have

$$K_1 = -\frac{\partial^2}{\partial a^2}\oint d\phi\, a_\mu a_{\alpha\gamma}a_\beta\phi_{\delta\nu}. \quad (B4)$$

Consider the first and last factors in Eq. (B4); we have

$$a_\mu\phi_{\delta\nu} = \nabla_\delta(a_\mu\phi_\nu) - a_{\mu\delta}\phi_\nu. \quad (B5)$$

The first term (with $\epsilon_{\mu\nu}$) vanishes from Eq. (27). The second term has a minus sign which can be eliminated by interchanging μ and ν. Thus, we have merely interchanged the a and ϕ symbols in Eq. (B4). Let us call this "argument 2." Now we have

$$K_1 = -\frac{\partial^2}{\partial a^2}\oint d\phi\, \phi_\mu a_{\alpha\gamma}a_\beta a_{\delta\nu}. \quad (B6)$$

This is antisymmetric in μ and β, as we can see by interchanging μ and β, ν and α, which gives a plus sign, and γ and δ which gives a minus sign. Since it is also antisymmetric in μ and ν, we might ask whether we can interchange β and ν. In fact, we can interchange them if we also multiply by $\tfrac{1}{2}$, a fact that can be verified by writing out components. This we shall call "argument 3." Applying Eq. (27) as well, we obtain finally

$$K_1 = +\frac{\partial^2}{\partial a^2}\oint d\phi\, a_{\alpha\gamma}a_{\beta\delta}. \quad (B7)$$

Now, having amassed our arguments, we can dispose of the remaining terms. A typical middle term of

Eq. (B1) gives

$$K_2 = -i \oint d\phi \; a_{\alpha\gamma\mu}\phi_{\beta\delta}\phi_\nu$$

$$= -i \oint d\phi \{ \nabla_\alpha (a_{\gamma\mu}\phi_{\beta\delta}\phi_\nu) - a_{\gamma\mu}\phi_{\beta\delta}\phi_{\alpha\nu} \}. \quad \text{(B8)}$$

The second term vanishes from argument 1. We apply Eq. (32) to the first term, but first interchange the first a and the last ϕ in the bracket (argument 2), giving

$$K_2 = -\frac{\partial}{\partial a} i \oint d\phi \; a_{\alpha}\phi_{\gamma\mu}\phi_{\beta\delta}a_\nu, \quad \text{(B9)}$$

which also vanishes from argument 1. Now, for the last term in Eq. (B1), we have

$$K_3 = -\oint d\phi \; a_{\alpha\gamma\mu}\phi_\beta\phi_\delta\phi_\nu$$

$$= -\oint d\phi \{ \nabla_\alpha [a_{\gamma\mu}\phi_\beta\phi_\delta\phi_\nu] - 2a_{\gamma\mu}\phi_\beta\phi_{\alpha\delta}\phi_\nu \}. \quad \text{(B10)}$$

In both terms, we use argument 2 to interchange the first a and the last ϕ. We then use argument 3 in the second term to interchange α and ν. Meanwhile, integrating the first term, we have

$$K_3 = -\frac{\partial}{\partial a} \oint d\phi \; a_{\alpha}\phi_\beta\phi_{\gamma\mu}\phi_\delta a_\nu + \oint d\phi \; a_{\alpha}\phi_\beta\phi_{\gamma\mu}\phi_{\delta\nu}. \quad \text{(B11)}$$

We can now reduce both terms with Eq. (27), and then integrating the second term, we find that it cancels the first, so that $K_3 = 0$.

We are thus left with K_1, and restoring the ϵ's, we obtain for the second term of Eq. (31)

$$\frac{1}{6\pi} \frac{\partial^2}{\partial a^2} \oint d\phi \; \epsilon_{\alpha\beta}\epsilon_{\gamma\delta}a_{\alpha\gamma}a_{\beta\delta}. \quad \text{(B12)}$$

APPENDIX C

We wish to evaluate the constants $\bar{\Gamma}_0$ and $\bar{\Gamma}_1$ in the vicinity of a maximum or minimum. Choosing a minimum, we must evaluate Γ for $\mathscr{E} = 0$, measured from the minimum. We choose an origin so that the minimum is at $\mathbf{k} = 0$, and then transform to a coordinate system for which the energy contours are circles near the minimum. Using $k_\pm = (1/\sqrt{2})(k_x \pm i k_y)$ as variables, with indices $\mu\nu$ running over $+$ and $-$, we assume an expansion for $\mathscr{E}(\mathbf{k})$,

$$\mathscr{E}(\mathbf{k}) = \alpha k_+ k_- + \tfrac{1}{6}\beta_{\mu\nu\lambda}k_\mu k_\nu k_\lambda + (1/24)\gamma_{\mu\nu\lambda\sigma}k_\mu k_\nu k_\lambda k_\sigma. \quad \text{(C1)}$$

We first calculate the eigenvalues of $\mathscr{E}(\kappa)$ for small n and to second order in h. The eigenvalues of the first

term of Eq. (C1) (with $\mathbf{k} \to \kappa$) are $(2n+1)\alpha h$ as is well known. We then use second-order perturbation theory for the second term, and first-order perturbation theory for the third term, remembering to use the completely symmetrized products. The terms are readily evaluated using the creation and annihilation operators

$$\begin{pmatrix} a^\dagger \\ a \end{pmatrix} = (2h)^{-1}k_\pm,$$

giving

$$\mathscr{E}_n = (2n+1)\alpha h + h^2 [(n+\tfrac{1}{2})^2 + \tfrac{1}{4}]\gamma_{++--}$$

$$- \frac{(n+\tfrac{1}{2})^2 h^2}{3\alpha} (\beta_{+++}\beta_{---} + 9\beta_{++-}\beta_{--+})$$

$$- \frac{h^2}{36\alpha} (5\beta_{+++}\beta_{---} + 9\beta_{++-}\beta_{--+}). \quad \text{(C2)}$$

In order to calculate Γ, we must obtain the eigenvalues of the area function $a(\mathscr{K})$. The function $a(\mathscr{E})$ can be calculated by treating the second two terms of Eq. (C1) as small and using the expansion of Eq. (39). The result is

$$a = a_0 - \oint \frac{d\lambda}{4\pi v_\perp} \mathscr{E}_1 + \frac{1}{2}\frac{\partial}{\partial \mathscr{E}} \oint \frac{d\lambda}{4\pi v_\perp} \mathscr{E}_1^2$$

$$= \frac{\mathscr{E}}{2\alpha} - \frac{1}{8}\frac{\mathscr{E}^2}{\alpha^3}\gamma_{++--}$$

$$+ \frac{1}{24}\frac{\mathscr{E}^2}{\alpha^4}(\beta_{+++}\beta_{---} + 9\beta_{++-}\beta_{--+}). \quad \text{(C3)}$$

We now substitute Eq. (C2) into Eq. (C3), keeping terms up to second order in h, to obtain

$$a_n = (n + \Gamma_0(0) + 2\Gamma_1(0)h)h, \quad \text{(C4)}$$

where $\Gamma_0(0) = \tfrac{1}{2}$ as expected, and

$$2\Gamma_1(0) = \left[\frac{\gamma_{++--}}{8\alpha} - \frac{5\beta_{+++}\beta_{---} + 9\beta_{++-}\beta_{--+}}{72\alpha^2} \right]. \quad \text{(C5)}$$

However, we have another expression for Γ_1 in Eq. (37). Writing this in terms of k_+ and k_-, we have

$$\Gamma_1 - \bar{\Gamma}_1 = \frac{1}{48\pi} \frac{\partial}{\partial \mathscr{E}} \oint \frac{d\lambda}{v_\perp} (\mathscr{E}_{+-}^2 - \mathscr{E}_{++}\mathscr{E}_{--}). \quad \text{(C6)}$$

The right-hand side can also be evaluated at $\mathscr{E} = 0$ by using our expansion, Eq. (37), differentiated with respect to $\mathscr{E}$. It turns out to be exactly equal to the right-hand side of Eq. (C5), so that we have $\bar{\Gamma}_1 = 0$.