# Field dependence of magnetic susceptibility of crystals under conditions of degeneracy of their electron energy bands

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Field dependences of the magnetic susceptibility of crystals are investigated theoretically in two cases of degeneracy of their electron energy bands in which these dependences are strong. In the first case, the degeneracy is lifted linearly in the wave vector **k** measured from the degeneracy point in the Brillouin zone. In the second case, the degeneracy takes place along a line, and the degeneracy is lifted linearly in  $\mathbf{k}$  in the directions perpendicular to this line. In both cases, the electron energy spectrum in the magnetic field **H** is obtained exactly. With the spectrum, we analyze the magnetic susceptibility  $\chi$  for an arbitrary **H**. It is shown that in strong magnetic fields for a chemical potential close to the degeneracy energy, the value of  $\chi$  is proportional to  $\log H$  in the first case and to  $H^{-1/4}$  in the second case of the band degeneracy.

### **INTRODUCTION**

If the chemical potential  $\zeta$  of electrons in a crystal lies far away from the critical points of their energy spectrum, the smooth component of the electron magnetic susceptibility (which does not oscillate with the magnetic field  $\mathbf{H}$ ) is virtually independent of  $\mathbf{H}^{1}$ . see Refs. 1-6. When the chemical potential is close to the critical energy  $(|\zeta - \varepsilon_{cr}| \sim \delta \varepsilon_H)$ , the field dependence of magnetic susceptibility  $\chi$  is determined by the relation between the temperature T and  $\delta \varepsilon_H$ . In weak fields  $\delta \varepsilon_H \ll T$ , this dependence is still weak (the field corrections are of the order of  $(\delta \varepsilon_H/T)^2$ ). In strong fields,  $\delta \varepsilon_H >> T$ , the value of  $\chi$  generally depends on H significantly, and it becomes impossible to present the magnetic susceptibility as the sum of the smooth and oscillating components.

Critical points of the electron spectrum in crystals can be of two types. The first type includes energy values for which the  $2\frac{1}{2}$ order electron topological transitions occur.<sup>7</sup> The second type of critical points in the spectrum is associated with degeneracy of electron energy bands. The field dependences of magnetic susceptibility for values of  $\zeta$  close to points of the  $2\frac{1}{2}$  order electron topological transition were analyzed theoretically by Nedorezov.<sup>3,4</sup> For the second type of critical points, H-dependences of  $\chi$  have not yet been investigated. The dependence  $\chi(H)$  was calculated only for one model of the spectrum<sup>8</sup> in which the band degeneracy of a special type can take place.

It was shown in Ref. 9 that giant anomalies in the magnetic susceptibility can take place for certain types of energy-band degeneracy. In other words, the absolute value of  $\chi$  calculated at a low temperature  $(T \to 0)$  in the limit of weak field  $(H \to 0)$ increases unlimitedly as the chemical potential approaches the degeneracy energy  $\varepsilon_d$ . In the same paper, all such types of the band degeneracy were indicated. In all these cases, electron states with energies close to  $\varepsilon_d$  are responsible for the giant anomaly in the magnetic susceptibility. Since the same states also determine field dependences of the magnetic susceptibility for  $\zeta \to \varepsilon_d$ , its field dependences must be strong in the cases when the giant anomalies in  $\chi$  are possible. Aluminum, beryllium, graphite, and the alloys  $Bi_{0.96}Sb_{0.04}$  and  $Pb_{0.65}Sn_{0.35}$  Te are examples of the crystals in which the band-degeneracy points of the appropriate types exist near the Fermi level. It should be noted that strong field dependences of  $\chi$  were indeed observed for Bi and its alloys with  $\mathrm{Sb.}^{10}$ 

In this research, field dependences of the magnetic susceptibility of electrons are investigated in all the cases of the band degeneracy in which the giant anomalies in  $\chi$  are possible. In order to calculate these dependences, we must first determine the electron spectrum in a magnetic field. Therefore, we will proceed as follows. In the first section, a model spectrum describing all the cases of the band degeneracy of interest will be given. In Sec. 2, this spectrum will be used for obtaining the exact electron energy levels in a magnetic field. In Sec. 3, field dependences of the magnetic susceptibility of crystals with different types of the band degeneracy will be calculated on the basis of the obtained spectrum.

#### 1. MODEL SPECTRUM

According to Ref. 9, the giant anomalies in magnetic susceptibility are possible for the following cases of the band degeneracy.

Case I. Band degeneracy takes place at a certain point of the Brillouin zone and is lifted linearly in the wave vector  $\mathbf{k}$  in all directions (the value of  $\mathbf{k}$  is measured from the degeneracy point in all cases). Such a situation can take place both for a symmetry-<sup>1</sup>The relative value of field corrections to magnetic susceptibility is of the order of  $\delta \varepsilon_{H}^{2}/(\zeta - \varepsilon_{cr})^{2}$  where  $\delta \varepsilon_{H}$  is the characteristic separation between electron energy levels in a magnetic field and  $\varepsilon_{cr}$  is the critical point in the spectrum, which is the closest to  $\zeta$ .

enforced and an accidental band degeneracy if the spin-orbit interaction is not weak ( $\rm Bi_{0.96}Sb_{0.04}$  and  $\rm Pb_{0.65}Sn_{0.35}Te$ ). Besides, accidental band degeneracy of this type can be observed in crystals without an inversion center even if the spin-orbit interaction can be neglected.

Case II. Band degeneracy occurs along a certain line in the Brillouin zone and is lifted linearly in  $\mathbf{k}$  in directions perpendicular to this line. Such degeneracy can be realized along the symmetry axes of the Brillouin zone (graphite). Besides, if we neglect spin-orbit interaction in crystals with an inversion center, the accidental band degeneracy practically always occurs just on the lines (aluminum and beryllium). The degeneracy of this type is also possible in a symmetry plane of crystals without an inversion center. The giant anomaly in  $\chi$  is determined only by points on the band degeneracy line, at which the energy  $\varepsilon_d$  of these bands attains extremal values. Henceforth, we will be interested only in these points when analyzing the case II.

One more type of the band degeneracy for which the giant anomaly in  $\chi$  can be observed was indicated in Ref. 9. However, this type is probably rare in occurrence. At any rate, we do not know a crystal with this type of the band degeneracy. For this reason, this case will not be considered here.

For all the cases listed above, electron energy spectra in the vicinity of the degeneracy points of interest can be described by a universal model of band structure. This model was formulated in Ref. 9 with the use of the Luttinger—Kohn representation for describing the electron states in a crystal.<sup>11</sup> In this model, the electron Hamiltonian for the two bands under investigation (denoted by c and v) has the form ( $\hbar = 1$ )

$$\hat{H} = \begin{pmatrix} \Delta + K_c & R & 0 & S \\ R^* & -\Delta + K_v & -S & 0 \\ 0 & -S^* & \Delta + K_c & R^* \\ S^* & 0 & R & -\Delta + K_v \end{pmatrix}$$
(1)

where

$$K_{c,v} = \mathbf{v}_{c,v}\mathbf{k} + q_{c,v}k_3^2$$

$$R = \mathbf{rk} + Q_rk_3^2$$

$$S = \mathbf{sk} + Q_sk_3^2$$
(2)

Here  $\mathbf{v}_{c,v}$  are the intraband and  $\mathbf{r}$  and  $\mathbf{s}$  the interband matrix elements of the velocity operator, which were calculated for  $\mathbf{k} = 0$ . The terms with the coefficients  $q_{c,v}$ ,  $Q_r$  and  $Q_s$  take into account the effect of other bands on the spectrum. In formulas (2), the quantities  $v_{c,v}$  and  $q_{c,v}$  are real, while  $Q_r$  and  $Q_s$  are generally complex. The energy (and subsequently the chemical potential also) will be measured from the degeneracy energy  $\varepsilon_d(\mathbf{k} = 0)$ .

In order to be able to analyze the effect of a small gap in the spectrum on final results, we have introduced in (1) the band splitting equal to  $2\Delta$  at the point  $\mathbf{k} = 0$ . In case I, such a gap can emerge due to a slight difference between the concentration of the alloy (e.g.,  $\operatorname{Bi}_{1-x}\operatorname{Sb}_x$ ) and that in the case of the band degeneracy. Such gap also necessarily exists in crystals with an inversion center, for which the accidental band degeneracy on the line takes place only if we neglect the spin-orbit interaction. In formula (1), it is assumed that the crystal has an inversion center so that each of the bands c and v is doubly degenerate. Here we take into account the symmetry of matrix elements, which follows from the properties of the time inversion operator. However, the Hamiltonian (1) and (2) can be used for calculating the susceptibility for crystals without the inversion center as well. In this case, we must put S = 0, and the final result for  $\chi$  should be divided by two since the true Hamiltonian now coincides with any of the two-dimensional blocks on the principal diagonal of (1).

If the spin-orbit interaction is small and can be neglected, one should set S = 0. In this case, expression (1) takes into account the twofold band degeneracy in spin. Besides, in the presence of the inversion center, the quantities  $\mathbf{r}$  and  $Q_r$  are real.

Using relations (1) and (2), we derive the energy–momentum relations for the bands in the vicinity of the degeneracy point:

$$\varepsilon_{c,v} = \mathbf{a}\mathbf{k} + Qk_3^2 + E_{c,v}(\mathbf{k})$$
$$E_{c,v}(\mathbf{k}) = \pm \sqrt{(\Delta + \mathbf{a}'\mathbf{k} + Q'k_3^2)^2 + |R|^2 + |S|^2}.$$
(3)

In these relations, the following notations are used:

$$\mathbf{a} = \frac{\mathbf{v}_c + \mathbf{v}_v}{2}; \qquad \mathbf{a}' = \frac{\mathbf{v}_c - \mathbf{v}_v}{2}; Q = \frac{q_c + q_v}{2}; \qquad Q' = \frac{q_c - q_v}{2}.$$

Henceforth, we choose the coordinate axes along the principal directions of the quadratic form in the components of vector  $\mathbf{k}$ which appears in  $E_{c,v}^2(\Delta = 0)$ . Let  $b_{ii}$  be its principal values (which can be expressed in terms of the components of the vectors  $\mathbf{a}'$ ,  $\mathbf{r}$ , and  $\mathbf{s}$ ), i.e.,

$$E_{c,v}^2(\Delta=0)=b_{11}^2k_1^2+b_{22}^2k_2^2+b_{33}^2k_3^2$$

If one of the principal values is equal to zero as this occurs in the case II, we will assume that it coincides with  $b_{33}$ . For this reason, we have retained only the terms of the type  $k_3^2$  among the terms quadratic in  $k_i$  in (2). Other quadratic terms are relatively small. In the case I, all  $b_{ii}$  are nonzero, and the terms proportional to  $k_3^2$  can be neglected in (2).

## 2. ELECTRON SPECTRUM IN A MAGNETIC FIELD

Let us consider the electron energy levels in a magnetic field  $\mathbf{H}$  whose direction is determined by the unit vector  $\mathbf{n}$  for the model spectrum described above. The Hamiltonian of an electron in the magnetic field in the coordinate representation can be obtained from (1) and (2) with the substitution<sup>11</sup>

$$k_i \to -i \frac{\partial}{\partial k_i} + \frac{e}{c} A_i,$$
 (4)

where **A** is the vector potential of the magnetic field and *e* the absolute value of the electron charge. We will neglect the direct interaction of the electron spin with the magnetic field since the giant anomaly in  $\chi$  has an orbital nature, and the purely spin contribution to magnetic susceptibility is relatively small. Let the magnetic field be directed along the axis **x**<sub>3</sub>, i.e., **n** = (0,0,1). We choose the vector potential in the form

$$\mathbf{A} = (-Hx_2, 0, 0),$$

and search the solution of the eigenvalue equation

$$\hat{H}\vec{\psi}_{\mu} = \varepsilon_{\mu}\vec{\psi}_{\mu} \tag{5}$$

in the form

$$\vec{\psi}_{\mu}(\mathbf{r}) = \exp\{i(k_1x_1 + k_3x_3)\}\mathbf{f}_l(x_2),\tag{6}$$

where  $\vec{\psi}_{\mu}$ ,  $\mathbf{f}_{l}$  are the four-dimensional column vectors of the functions, and the index  $\mu$  indicates the set consisting of an integer l and real-valued  $k_{1}$  and  $k_{3}$ . In the case of a discrete spectrum, the vectors  $\mathbf{f}_{\mu}$ , must satisfy the boundary condition

$$\mathbf{f}_l(x_2) \to 0 \quad \text{for} \quad |x_2| \to \infty.$$

Such vectors do exist if

$$R_{\mathbf{x_3}} \equiv b_{11}b_{22}(1 - \tilde{a}_{\perp}^2) > 0, \tag{7}$$

where  $\tilde{a}_{\perp}^2 \equiv \tilde{a}_1^2 + \tilde{a}_2^2$ ,  $\tilde{a}_i \equiv a_i/\sqrt{b_{ii}}$ . The meaning of inequality (7) is the following: The cross sections of the constant-energy surface  $\varepsilon_{c,v}(\mathbf{k}) = \text{const}$  (with  $\varepsilon_{c,v}$  from (3)) by the planes  $k_3 = \text{const}$ are second-degree curves described by a quadratic form in  $k_1, k_2$ . The quantity  $\mathbf{R}_{\mathbf{x}_3}$  is an invariant of this form (the so-called discriminant of its higher terms).<sup>12</sup> A positive value of  $\mathbf{R}_{\mathbf{x}_3}$  indicates that the above mentioned second-degree curves are closed and are ellipses. When condition (7) is satisfied, we search the eigenfunctions  $\mathbf{f}_l$ , in the form

$$\mathbf{f}_{l}(x_{2}) = \exp(-\lambda_{1}x_{2}^{2} - \lambda_{2}x_{2}) [\mathbf{C}_{1}H_{l}\{\lambda_{3}(x_{2} - x_{20})\} + \mathbf{C}_{2}H_{l-1}\{\lambda_{3}(x_{2} - x_{20})\}],$$
(8)

where  $l \geq 0$ ,  $H_l(x)$  is an *l*-th degree Hermite polynomial,  $H_{-1} \equiv 0$ , and the parameters  $x_{20}$ ,  $\lambda_{1,2,3}$  as well as the constant four-dimensional vectors  $\mathbf{C_1}$  and  $\mathbf{C_2}$  are determined by the substitution of (6) and (8) into (5). It can be shown that such types of solutions exist, with each energy level corresponding to two sets of  $\mathbf{C_1}$ ,  $\mathbf{C_2}$  for l > 0 and one set for l = 0. The explicit form of the parameters and  $\mathbf{C_1}$ ,  $\mathbf{C_2}$  is not used in the further analysis and hence is not given here. The eigenvalues of energy  $\varepsilon_l(k_3)$  are determined from the equation

$$S(\varepsilon_l, k_3) = \frac{2\pi eH}{c}l,\tag{9}$$

where  $S(\varepsilon, k_3)$  is the cross-section area of the constant energy surface by the plane  $k_3 = \text{const.}$  It should be noted that the quantization condition (9) has a semiclassical form. In the given case, however, it determines the exact eigenvalues of the electron energy. The explicit form of these eigenvalues will be given below for each case of the band degeneracy. Since the Hamiltonian in the case I is linear in  $\mathbf{k}$ , the above results can easily be generalized to the case of an arbitrary directed magnetic field. In the case II, such generalization is impossible, but we can find approximate expressions for the energy levels of an electron in a magnetic field which direction is not too close to the plane  $(x_1, x_2)$ .

Case I. For an arbitrary direction of H, the condition for the existence of a discrete spectrum generalizing (7) has the form

$$R_{\mathbf{n}} = b_{11}b_{22}b_{33}\left(\widetilde{\mathbf{n}}^2 - [\widetilde{\mathbf{n}} \times \widetilde{\mathbf{a}}]^2\right) > 0, \tag{10}$$

where the vector  $\ \mathbf{\tilde{n}}$  is defined as

$$\tilde{n}_i \equiv n_i / \sqrt{b_{ii}}.$$

Condition (10) has the same meaning as (7). When (10) is satisfied, the cross sections of the constant-energy surfaces by the planes perpendicular to the magnetic field are ellipses.

It was shown in Ref. 9 that the giant anomaly in  $\chi$  exists if

$$\widetilde{\mathbf{a}}^2 \equiv \widetilde{a}_\perp^2 + \widetilde{a}_3^2 < 1 \quad . \tag{11}$$

This requirement is stronger than the condition for the existence of a discrete spectrum for a given direction of **H** (10). It can be proved that when condition (11) is satisfied, inequality (10) is valid for any **n**, i.e., the constant-energy surfaces are ellipsoids. In other words, condition (11) can be formulated as the requirement that the energy bands  $\varepsilon_c(\mathbf{k})$  and  $\varepsilon_v(\mathbf{k})$  must have points of minimum and maximum in **k**, respectively. If in this case  $\Delta \neq 0$  and  $\tilde{\mathbf{a}} \neq \mathbf{0}$ , then the band extrema occur at different points of the Brillouin zone, and the spectrum contains an indirect gap

$$2\Delta_{\min} = 2\Delta(1-\tilde{\mathbf{a}}^2)^{1/2}(1-(\tilde{\mathbf{a}}')^2)^{1/2}$$

$$\tilde{a}'_i \equiv \frac{a'_i}{\sqrt{b_{ii}}}$$

(the inequality  $(\tilde{\mathbf{a}}')^2 < 1$  follows from the definition of the quantities  $b_{ii}$ ). The minimum value of  $\varepsilon_c(\mathbf{k})$  is equal to  $\varepsilon_0 + \Delta_{\min}$ , while the maximum value  $\varepsilon_v(\mathbf{k})$  is  $\varepsilon_0 - \Delta_{\min}$ , where

$$\varepsilon_0 = -(\mathbf{\tilde{a}} \cdot \mathbf{\tilde{a}}')\Delta.$$

If condition (10) is satisfied, the energy levels of an electron in the magnetic field are

$$\varepsilon_l^{c,v}(\delta k_{\mathbf{n}}) = \varepsilon_0 + v \delta k_{\mathbf{n}}$$
$$\pm \sqrt{\frac{eH\alpha}{c}l + L(\delta k_{\mathbf{n}})^2 + \Delta_{\mathbf{n}}^2}, \qquad (12)$$

where

L

where

$$\delta k_{\mathbf{n}} = k_{\mathbf{n}} - k_{\mathbf{n}}^{0}, \quad k_{\mathbf{n}} = \mathbf{kn}, \quad k_{\mathbf{n}}^{0} = -\Delta(\widetilde{\mathbf{a}}'\widetilde{\mathbf{n}}),$$

$$v = \frac{(\widetilde{\mathbf{a}}\widetilde{\mathbf{n}})}{\widetilde{\mathbf{n}}^{2}}, \quad \alpha = \frac{2R_{\mathbf{n}}^{3/2}}{b_{11}b_{22}b_{33}\widetilde{\mathbf{n}}^{2}},$$

$$= \frac{R_{\mathbf{n}}}{b_{11}b_{22}b_{33}\widetilde{\mathbf{n}}^{4}}, \quad \Delta_{\mathbf{n}}^{2} = \Delta^{2}\frac{R_{\mathbf{n}}[1 - (\widetilde{\mathbf{a}}')^{2}]}{b_{11}b_{22}b_{33}\widetilde{\mathbf{n}}^{2}},$$
(13)

and l is an non-negative integer. The two signs in (12) correspond to the two spectral branches. Note that although  $\Delta_{\mathbf{n}} \geq \Delta_{\min}$ , under condition (11) one has  $L \geq v^2$ , and the minimum value of  $\varepsilon_0^c(\delta \mathbf{k_n})$  is still equal to  $\varepsilon_0 + \Delta_{\min}$ , while the maximum value of  $\varepsilon_0^v(\delta \mathbf{k_n})$  is equal to  $\varepsilon_0 - \Delta_{\min}$ .

Case II. In this case,  $b_{33} = 0$ . Let the magnetic field be directed along the axis  $x_3$ , that is, along the tangent to the line of the band degeneracy at the given point of the Brillouin zone. The giant anomaly in  $\chi$  is associated only with the points on this line at which, in addition to (7), the following condition is satisfied: <sup>9</sup>

$$a_3 = 0.$$

Using (9), we can obtain the following explicit expression for the eigenenergy values of electron states in the vicinity of these points:

$$\varepsilon_l^{c,v}(k_3) = Bk_3^2 \pm \sqrt{\frac{eH\alpha}{c}l},\tag{14}$$

where l is a non-negative integer, and the two signs correspond to the two spectral branches;

$$\alpha = \frac{2R_{\mathbf{x}_{\mathbf{3}}}^{3/2}}{b_{11}b_{22}};$$
$$B = Q - Q'(\widetilde{\mathbf{a}}_{\perp}\widetilde{\mathbf{a}}'_{\perp}) - Q_r(\widetilde{\mathbf{a}}_{\perp}\widetilde{\mathbf{r}}_{\perp}); \tag{15}$$

 $\mathbf{\tilde{r}}_{\perp}$  is the vector perpendicular to the axis  $x_3$ :

$$\mathbf{\tilde{r}}_{\perp}=\big(\frac{r_1}{\sqrt{b_{11}}},\frac{r_2}{\sqrt{b_{22}}},0\big)$$

and  $\tilde{\mathbf{a}}_{\perp}$ ,  $\tilde{\mathbf{a}}'_{\perp}$  are defined similarly. Another expression for the coefficient *B* (which is equivalent to (15)) is given in Ref. 9.

When the band degeneracy on a certain line in a crystal with an inversion center takes place only if one neglects the spin-orbit interaction, then this interaction leads to  $\Delta \neq 0$  in expression (1) and to changes in the values of the parameters appearing in (2). However, these changes are such that in the leading order in  $\Delta$ , the values of **a** and *Q* remain the same as for  $\Delta = 0$ , and

$$E_{c,v}^2(\Delta, \mathbf{k}) = \Delta^2 + E_{c,v}^2(\Delta = 0, \mathbf{k}).$$

The minimum indirect gap will be equal to

$$2\Delta_{\min} = 2\Delta(1 - \tilde{a}_{\perp}^2)^{1/2},$$

and the spectrum in a magnetic field is

$$\varepsilon_l^{c,v}(k_3) = Bk_3^2 \pm \sqrt{\frac{eH\alpha}{c}l + \Delta_{\min}^2} \tag{16}$$

with the previous values of the parameters B and  $\alpha$  .

Let us now suppose that the magnetic field is directed at the angle  $\theta \neq 0$  to the axis  $\mathbf{x_3}$ . In this case, the eigenvalue equation cannot be solved exactly in view of the presence in (2) of the terms quadratic in  $k_3$ . After the substitution (4) and simple transformations, these terms make contributions of the type  $q(eH/c)^2 \sin^2 \theta$  to the Hamiltonian, where q is a certain combination of  $q_{c,v}$ ,  $Q_r$ , and  $Q_s$ . However, these contributions are relatively small in the parameter  $\eta^2$ , where  $\eta = \tan \theta (\varepsilon/E_0)^{1/2}$ and  $E_0$  is the characteristic energy separation between the two bands under consideration and other bands that are not considered explicitly in (1). Dropping all the terms that are small in parameter  $\eta$ , we obtain formulas (14) and (16) in which H should be replaced by  $H \cos \theta$ . Note that the electron-band model (1), (2) itself is valid in the leading order in  $(\varepsilon/E_0)^{1/2}$ , which implies that this parameter is small. Hence the obtained approximate results can, in fact, describe  $\varepsilon_l^{c,v}(k_3)$  quite well for nearly all values of the angle  $\theta$  except the small neighborhood of  $\pi/2$ . Before concluding this section, we note that formulas (9), (12), and (16) obtained above are in complete agreement with the results obtained in Refs. 8, 13-15 in which electron energy levels were studied in a magnetic field for specific crystal structures.

# 3. COMPUTATION OF MAGNETIC SUSCEPTIBILITY

If the chemical potential is close to the energy of the band degeneracy, the magnetic susceptibility of a crystal can be represented

as the sum of a special contribution associated with the electron states that are close to the degeneracy point, and a background term associated with all other states. The special contribution determines the dependence of  $\chi$  on T,  $|\mathbf{H}|$ , and  $\zeta$ , while the background term is practically independent of the magnetic field and temperature, and remains practically unchanged as  $\zeta$  varies in the vicinity of the degeneracy energy. In order to compute the special contribution to the magnetic susceptibility of the crystal (which is the only quantity in which we shall be interested), we use the following expression for the  $\Omega$ -potential per unit volume:

$$\Omega(\mathbf{H}) = -\frac{eHT}{4\pi^2 c} \sum_{c,v} \sum_{l=0}^{\prime} \int d(\delta k_{\mathbf{n}}) \\ \times \ln\left\{1 + \exp\left(\frac{\zeta - \varepsilon_l^{c,v}(\delta k_{\mathbf{n}})}{T}\right)\right\}$$
(17)

In this expression, the limits of integration with respect to  $\delta k_{\mathbf{n}}$ and of summation over l are determined from the condition  $\varepsilon_l^{c,v}(\delta k_{\mathbf{n}}) \geq -\zeta_0$ , where it is assumed that the spectral cutoff parameter  $\zeta_0$  satisfies the condition

$$E_0 >> \zeta_0 >> T, \zeta, \delta \varepsilon_H, \Delta.$$

The prime on the summation over l indicates that terms with l > 0 must be doubled.

In weak magnetic fields ( $\delta \varepsilon_H \ll T$ ), integration of (17) by parts and the application of the Euler-Maclaurin summation formula give results for magnetic susceptibility that are in complete agreement with formulas obtained in Ref. 9. Hence we will consider only the case of strong magnetic fields,  $\delta \varepsilon_H \gg T$ , below. This case can be easily realized in the vicinity of band degeneracy points. Indeed, formulas (12) and (16) give  $\delta \varepsilon_H \approx \sqrt{eH\alpha/c}$ . Hence the condition for strong field is  $H \gg H_T \equiv (cT^2/e\alpha)$ . Assuming that T = 4 K and using typical values for matrix elements of the velocity operator ( $m\alpha \sim 1$  eV, where m is the electron mass), we obtain  $H_T \approx 10$  Oe.

In order to analyze formula (17), we integrate it by parts several times and use Poisson's summation formula and put T = 0(due to  $\delta \varepsilon_H >> T$ ). Eventually we obtain the following results:

Case I. Suppose that  $|\zeta - \varepsilon_0| < \Delta_{\min}$ , i.e., the chemical potential is in the gap of the spectra (3) and (12). In this case

$$\Omega(\mathbf{H}) = \frac{1}{24\pi^2} \left(\frac{e}{c}\right)^2 \frac{\alpha}{\sqrt{L}} H^2 \left\{ \ln\left(\frac{2\zeta_0}{\Delta_{\min}}\right) + \frac{3}{\pi^2} \sum_{l=1}^{\infty} \frac{1}{l^2} \left[\sin(\varphi_l) \operatorname{si}(\varphi_l) + \cos(\varphi_l) \operatorname{Ci}(\varphi_l)\right] \right\}, \quad (18)$$

where  $\varphi_l = 2\pi l (H_\Delta/H)$ ,

$$H_{\Delta} = \frac{c\Delta_{\mathbf{n}}^2}{e\alpha}$$

is the characteristic magnetic field for which  $\delta \varepsilon_H \sim \Delta_n$ ; si( $\varphi$ ) and Ci( $\varphi$ ) are the integral sine and cosine defined by the expressions<sup>16</sup>

$$\operatorname{si}(\varphi) = \int_{\infty}^{\varphi} \frac{\operatorname{sin}(t)}{t} dt, \quad \operatorname{Ci}(\varphi) = \int_{\infty}^{\varphi} \frac{\cos(t)}{t} dt.$$

In formula (18), we have dropped the contribution that does not depend on magnetic field, as well as terms that are small in the parameter  $(\delta \varepsilon_H / \zeta_0)$ .<sup>2</sup>

<sup>&</sup>lt;sup>2</sup>These terms also include the terms oscillating in (1/H) at a high frequency proportional to  $\zeta_0^2$ . Although these terms could make a large contribution to magnetic susceptibility, they appear here as a result of artificial introduction of a sharp energy boundary for electron states, which is equal to  $-\zeta_0$ .

Another equivalent representation of  $\Omega(H)$ ,

$$\begin{split} \Omega(\mathbf{H}) = & \frac{1}{24\pi^2} \left(\frac{e}{c}\right)^2 \frac{\alpha}{\sqrt{L}} H^2 \Big\{ \ln\left(\frac{2\zeta_0}{\Delta_{\min}}\right) \\ & -\frac{3}{\pi^2} \int_0^\infty \frac{tL_2(e^{-t})}{t^2 + \left(\frac{2\pi H_\Delta}{H}\right)^2} dt \Big\}, \end{split}$$

which is convenient for studying limiting cases, is obtained with the expression

$$\sin(\varphi)\operatorname{si}(\varphi) + \cos(\varphi)\operatorname{Ci}(\varphi) = \ln \varphi - \frac{1}{2} \int_0^\infty e^{-t} \ln(t^2 + \varphi^2) dt \quad (19)$$

where

$$L_2(z) = \sum_{n=1}^{\infty} \frac{z^n}{n^2}.$$

Using this representation, one can show that the magnetic susceptibility tensor defined as

$$\chi_{ij} = -\frac{\partial^2 \Omega}{\partial H_i \partial H_j}.$$

is a monotonically decreasing function of H. For  $H \ll H_{\Delta}$ , we obtain the following expression for it:

$$\chi_{ij} = -\frac{1}{6\pi^2} \left(\frac{e}{c}\right)^2 \frac{1}{(b_{11}b_{22}b_{33})^{1/2}} \left\{ \kappa^{ij} \ln\left(\frac{2\zeta_0}{\Delta_{\min}}\right) -\frac{\pi^2}{20} \frac{H^2}{H_\Delta^2} \varphi^{ij} \right\}$$
(20)

where

$$\varphi^{ij} = \sum_{k=1}^{3} \sum_{l=1}^{3} \frac{\kappa^{ij} \kappa^{kl} + 2\kappa^{ik} \kappa^{jl}}{3R_{\mathbf{n}}} n_k n_l,$$
  
$$\kappa^{ij} = \frac{b_{11} b_{22} b_{33}}{(b_{ii} b_{ij})^{1/2}} \left\{ \left(1 - \widetilde{\mathbf{a}}^2\right) \delta_{ij} + \widetilde{a}_i \widetilde{a}_j \right\}.$$

Here the tensor  $\kappa_{ij}$  introduced in Ref. 9 was obtained using Eq. (10) and the definition of  $\kappa_{ij}$ ,

$$R_{\mathbf{n}} \equiv \sum_{ij} \kappa_{ij} n_i n_j.$$

The first term in (20) coincides with the magnetic susceptibility in a weak magnetic field,<sup>9</sup> which is finite due to the existence of a gap in the spectrum, while the second term gives the first field correction to it. For  $H >> H_{\Delta}$ , we obtain for  $\chi_{ij}$ :

$$\chi_{ij} = -\frac{1}{6\pi^2} \left(\frac{e}{c}\right)^2 \frac{1}{(b_{11}b_{22}b_{33})^{1/2}} \left\{ \kappa^{ij} \ln\left[\frac{2\zeta_0}{\Delta_{\min}} \left(\frac{H_\Delta}{H}\right)^{1/2}\right] + A\kappa^{ij} - \frac{3}{4}\varphi^{ij} \right\}$$
(21)

where

$$A = \frac{1}{2} \left( \ln(2\pi) + C_{EM} \right) + \frac{3}{\pi^2} \sum_{l=1}^{\infty} \frac{\ln l}{l^2} \approx 1.50 \quad ,$$

and  $C_{EM} \approx 0.58$  is the Euler-Mascheroni constant. Using Eq. (13), we find that

$$\frac{\sqrt{H_{\Delta}}}{\Delta_{\min}} = \left(\frac{c}{2e\sqrt{R_{\mathbf{n}}}\left(1-\widetilde{\mathbf{a}}^2\right)}\right)^{1/2},\tag{22}$$

i.e., the value of  $\chi_{ij}$  in this field region does not depend on the gap in the spectrum. Formulas (21) and (22) describe the magnetic susceptibility of the crystal with a point of band degeneracy at T = 0. The general form of the dependence of  $\chi_{ij}$  on H is represented in Fig. 1. Note that, if  $T \neq 0$  and  $\Delta < T$  the form of the H-dependence of  $\chi_{ij}$  is approximately the same as for T = 0 and  $\Delta \neq 0$ . One must only replace  $H_{\Delta}$  by  $H_T$ . But  $\chi_{ij}$  at  $H \gg H_T$ will be still described by Eq. (21) with substitution (22).

Let us now consider the region  $|\zeta - \varepsilon_0| > \Delta_{\min}$ . In this case,  $\Omega$ -potential (17) can be represented as

$$\Omega(\mathbf{H},\zeta) = \Omega(\mathbf{H}) + \delta\Omega(\mathbf{H},\zeta) \tag{23}$$



FIG. 1. Schematic form of the magnetic field dependence of the magnetic susceptibility tensor  $\chi_{ij}$  of the crystal in the case I; T = 0, and  $\zeta$  is inside the gap in spectrum (3). The dashed curve corresponds to  $\Delta = 0$  and the solid curve to  $\Delta \neq 0$ .

where  $\Omega(\mathbf{H})$  is described by Eq. (18), while the correction  $\delta\Omega$  is the sum of a finite number of terms:

$$\delta\Omega(\mathbf{H},\zeta) = -\frac{1}{4\pi^2} \left(\frac{e}{c}\right)^2 \frac{\alpha H^2}{\sqrt{L}} \sum_{m=0}^{M} {}^{\prime} \left\{ x\sqrt{x^2 - x_m^2} - x_m^2 \ln\left[\frac{x + \sqrt{x^2 - x_m^2}}{x_m}\right] \right\}.$$
 (24)

Here

$$x_m^2 = m + H_{\Delta}/H; \quad x = \frac{|\zeta - \varepsilon_0|}{\Delta_{\min}} \left(\frac{H_{\Delta}}{H}\right)^{1/2}$$

M is the maximum integer for which  $x_M^2 \leq x^2$ . It follows from (24) that the magnetic susceptibility is a symmetric function of  $\zeta - \varepsilon_0$ . For large x, formulas (18), (23), and (24) lead to the well-known result according to which the magnetization of a crystal is the sum of  $\chi(H \to 0) \cdot H$  and an oscillating de Haas-van Alfen component. Qualitatively, this statement remains true even

for  $x \sim x_1$ . With decreasing x the absolute value of the smooth magnetization component increases, while the amplitude of the oscillating component decreases, the former being considerably larger than the latter for  $x \sim x_1$ . According to (23), we obtain the following expression for the magnetization **M** of the crystal in the gapless state (or at  $H \gg H_{\Delta}$ ) for  $0 < x < x_1$ :

$$\mathbf{M}(\mathbf{H}, \zeta) = \mathbf{M}(\mathbf{H}) + \delta \mathbf{M}(\mathbf{H}, \zeta).$$

Here  $\mathbf{M}(\mathbf{H})$  is independent of  $\zeta$  and is obtained from (18)  $[M_i(\mathbf{H}) \sim H \log H, \text{ see Eq. (21)}]$ . The additional term

$$\delta M_i(\mathbf{H},\zeta) = \frac{1}{4\pi^2} \frac{e}{c} \frac{\kappa^{ij} n_j}{(b_{11}b_{22}b_{33})^{1/2}} \frac{\zeta^2}{(1-\tilde{\mathbf{a}}^2)\sqrt{R_{\mathbf{n}}}}$$

describes the behavior of the magnetization upon variation of  $\zeta$  in the vicinity of the energy of the band degeneracy. If  $H << H_{\Delta}$ , this correction becomes proportional to  $(|\zeta - \varepsilon_0| - \Delta_{\min})^{3/2}$  for  $x_0 \leq x < x_1$  and is in accordance with the result obtained from formulas (41) and (42) of Ref. 3.

The chemical potential of electrons in a crystal is generally a function of the magnetic field. This dependence is obtained from the condition of conservation of the total number of electrons. If, in addition to the two bands under consideration, the crystal has other partially filled bands with a high density of states, i.e., if we are dealing with a metal, then the value of  $\zeta$  remains practically constant, and we can directly use the above formulas. Let us now consider the case when there are no other unfilled bands and hence the crystal is a gapless semiconductor (or a semiconductor with a narrow gap). In this case, the density of charge carriers (electrons or holes) is described by the expression

$$\nu = \frac{1}{2\pi^2} \left(\frac{e}{c}\right)^{3/2} \frac{\alpha^{1/2} H^{3/2}}{\sqrt{L - v^2}} \sum_{m=0}^{M} \sqrt{x^2 - x_m^2}$$
(25)

which gives the dependence  $x(\nu, H)$ . In order to compute the magnetization or the magnetic susceptibility of electrons, we must use the free energy instead of the  $\Omega$ -potential. As a result, we obtain

$$\begin{split} \mathbf{M}(\mathbf{H},\nu) = & \mathbf{M}(\mathbf{H},x) \Big|_{x=x(\nu,\mathbf{H})}, \\ \chi_{ij}(\mathbf{H},\nu) = & \left[ \chi_{ij}(\mathbf{H},x) - \frac{\partial\nu}{\partial H_i} \frac{\partial\nu}{\partial H_j} \left( \frac{\partial\nu}{\partial\zeta} \right)^{-1} \right]_{x=x(\nu,\mathbf{H})}. \end{split}$$

For an intrinsic semiconductor ( $\nu = 0$ ), we find from (25) that x = 0 for any **H**, while  $\chi_{ij}(\mathbf{H}, \nu)$  is still described by Eqs. (20) and (21). For  $\nu \neq 0$ , we restrict ourselves by an analysis of the high field case

$$H > H_0 = \frac{c}{e} (4\pi^4 \nu^2 (L - v^2) / \alpha)^{1/3}$$

In such fields,  $x < x_1$ , and the sums in (24) and (25) reduce to a single term. If there is no gap in the spectrum, or if  $H >> H_{\Delta}$ , the magnetic susceptibility  $\chi_{ij}(\mathbf{H}, \nu)$  is described by formula (21) in which the expression in curly braces is supplemented by the term

$$(3/2)(H_0/H)^3(9\varphi^{ij}-5\kappa^{ij}).$$

If, however,  $H_0 < H << H_{\Delta}$ , the additional term is proportional to  $(H_0/H)^4$ , in accordance with the results of Ref. 1.

Finally, it should be noted that, if  $-\zeta_0$  is taken as the value of the chemical potential for which we know (say, experimentally) the magnetic susceptibility tensor in a weak magnetic field, the above formulas directly give the value of  $\chi_{ij}(\mathbf{H}, \zeta) - \chi_{ij}(\mathbf{H}, -\zeta_0)$ . According to (20)-(24), the choice of another value of  $\zeta_0$  will lead to a change in the magnetic susceptibility by a constant value and does not affect the dependences of this quantity on H and  $\zeta$ .

Case II. In this case, we obtain the following expression for the  $\Omega$ -potential at T = 0 and H directed along the axis  $\mathbf{x_3}$ 

$$\Omega(\mathbf{H},\zeta) = \frac{1}{\pi^3} \left(\frac{e}{c}\right)^{7/4} |B|^{-1/2} H^{7/4} \alpha^{3/4} \sum_{l=1}^{\infty} \frac{1}{l} \\ \times \left\{ I_l + \int_{\max(|x|,(H_\Delta/H)^{1/2})}^{\infty} dt (t+x)^{1/2} \\ \times \sin\left[2\pi l \left(t^2 - H_\Delta/H\right)\right] \right\},$$
(26)

where  $x \equiv \operatorname{sgn}(B)\zeta(c/e\alpha H)^{1/2}$ ,  $H_{\Delta} \equiv (c\Delta_{\min}^2/e\alpha)$ ,  $\operatorname{sgn}(B) = \pm 1$ depending on the sign of B, and the term  $I_l$  differs from zero only for positive  $x > (H_{\Delta}/H)^{1/2}$  and is given by

$$I_{l} = \int_{(H_{\Delta}/H)^{1/2}}^{x} dt \left[ (x+t)^{1/2} - (x-t)^{1/2} \right] \\ \times \sin \left[ 2\pi l \left( t^{2} - H_{\Delta}/H \right) \right].$$

In view of convergence of all the integrals in (26), we omitted the contribution independent of H to the  $\Omega$ -potential and set  $|\zeta_0| \to \infty$ . Let us now analyze various limiting cases with Eq. (26).

At first consider the case of the gapless spectrum for which  $H_{\Delta} = 0$ . It follows from (26) that the special contribution to magnetization of the crystal is

$$M_3 = -\frac{1}{\pi^2} \left(\frac{e}{c}\right)^{7/4} |B|^{-1/2} \alpha^{3/4} H^{3/4} f(x), \qquad (27)$$

where the function of one argument f(x) does not depend on spectral parameters, i.e., it is universal for all crystals with given type of the band degeneracy. Evaluating the sum over l in (26) and differentiating with respect to H, we arrive at the following expression for f(x):

$$f(x) = \frac{1}{4} \int_{-x}^{\infty} dt \left(\frac{1}{2} - \{t^2\}\right) \operatorname{sgn}(t) \frac{7t + 6x}{\sqrt{x+t}},$$
 (28)

where  $\{t^2\}$  is the fractional part of the number  $t^2$ . It can be shown that for |x| >> 1, Eqs. (27), (28) describe a smooth contribution to magnetization, which coincides with that obtained in Ref. 9, and the oscillating component which can be found with the Lifshits-Kosevich formula<sup>17</sup> and expression (9). It is follows from (28) that in contrast to the case I, the magnetization is not symmetric in x, and the amplitude of oscillations of  $M_3$  is of the order of the smooth magnetization component for  $x \sim 1$  (this result also remains valid in the presence of a gap in the spectrum). In the region |x| << 1, we obtain the following expansion for f(x):

$$f(x) = f_0 + f_1 x - \frac{2}{3} x^{3/2} \sigma(x) + O(x^2), \qquad (29)$$

where  $\sigma(x) = 1$  for x > 0 and  $\sigma(x) = 0$  for x < 0, and the constants  $f_0$  and  $f_1$  are given by

$$f_0 = \frac{7\cos(\pi/8)\zeta(7/4)}{2^{13/4}\pi^{3/4}\Gamma(1/4)} \approx 0.156$$

and

$$f_1 = \frac{5\sin(\pi/8)\zeta(5/4)}{2^{15/4}\pi^{1/4}\Gamma(3/4)} \approx 0.40$$

Here  $\Gamma(y)$  is the gamma function, and  $\zeta(y)$  is the Riemann zeta function. The term proportional to  $x^{3/2}$  in (29) describes the singularity in magnetization associated with the point of the band degeneracy under consideration.

Formulas (27)-(29) give the dependence of the electron magnetization on the chemical potential for various values of the magnetic field H. These formulas can also be used for obtaining the dependence  $M_3(H)$  at fixed  $\zeta$ . For example, for |x| << 1, we obtain the following expression for the magnetic susceptibility  $\chi_{33}$ :

$$\chi_{33}(H) \approx -\frac{3}{4\pi^2} \left(\frac{e}{c}\right)^{7/4} \frac{\alpha^{3/4} f_0}{|B|^{1/2}} H^{-1/4}.$$
 (30)

It should be noted that the dependence  $\chi \sim H^{-1/4}$  for strong magnetic fields was also obtained earlier by Beneslayskii and Fal'kovskii,<sup>8</sup> although the spectrum considered by them does not correspond to the case II (but is close to it in a certain respect: the degeneracy is lifted linearly in k in two directions and quadratically in the third direction).

Let us now consider the influence of the presence of a gap in the spectrum on the obtained results. It follows from (26) that this influence is small for  $|\zeta| >> \Delta_{\min}$ . For this reason, we will analyze only the case when  $\zeta \sim \Delta_{\min}$ , i.e.,  $|x| \sim (H_{\Delta}/H)^{1/2}$ . This influence can be neglected for  $H >> H_{\Delta}$ , i.e. for |x| << 1 since the coefficients  $f_0$  and  $f_1$  in (29) change by quantities of the order of  $(H_{\Delta}/H)$ , while the term  $x^{3/2}\sigma(x)$  is replaced by

$$\frac{x_+^{3/2}\sigma(x_+) + x_-^{3/2}\sigma(x_-)}{2},$$

where

$$x_{\pm} = x \pm (H_{\Delta}/H)^{1/2}.$$

The results will differ noticeably from those presented above only for  $H \leq H_{\Delta}$  and  $|\zeta| \leq \Delta_{\min}$ . Indeed, for  $H \ll H_{\Delta}$ , the smooth component of magnetic susceptibility can be described by the same formulas as in weak fields  $(H \ll H_T)$  everywhere excepting the small neighborhood of the points  $\zeta = \pm \Delta_{\min}$ :

$$\chi_{33} - \chi_{33}^{os} = -\frac{1}{24\pi^2} \left(\frac{e}{c}\right)^{7/4} \alpha^{3/4} |B|^{-1/2} \frac{1}{H_{\Delta}^{1/4}} \\ \times \left\{ \zeta_+^{1/2} \sigma(\zeta_+) - \zeta_-^{1/2} \sigma(\zeta_-) \right\},$$
(31)

where  $\zeta_{\pm} = (\operatorname{sgn}(B)\zeta/\Delta_{\min}) \pm 1$ , and  $\chi_{33}^{os}$  is the oscillating component existing only for  $|\zeta| > \Delta_{\min}$  (we do not give the explicit form of this component here). It should be noted that, in contrast to the case I, the magnetic susceptibility in the region  $|\zeta| < \Delta_{\min}$ depends strongly on the chemical potential. As the magnetic field increases from  $H \ll H_{\Delta}$  to  $H \gg H_{\Delta}$ , a crossover from (31) to (30) occurs gradually. In the crossover region, the magnetic susceptibility can change monotonically (curve 1 in Fig. 2) or nonmonotonically (curve 2) depending on the value of  $\zeta$  in the interval  $|\zeta| < \Delta_{\min}$ . It should be recalled that in the case I the field dependence of  $\chi_{ij}$  is always monotonic for  $|\zeta| < \Delta_{\min}$ .



FIG. 2. Field dependences of  $\chi_{33}$  in the case II for T = 0. The dashed curve corresponds to  $\Delta = 0$  and the solid curves to  $\Delta \neq 0$  (curve 1 is plotted for  $\zeta = 0$  and curve 2 for  $\zeta = -0.75\Delta$ );  $\chi_{33}(0)$  is the value of  $\chi_{33}$  in a weak field  $(H \to 0)$  for  $\zeta = 0$  and  $\Delta \neq 0$ .

If the angle  $\theta$  between the magnetic field and the axis  $\mathbf{x_3}$ differs from zero, all the formulas presented above for the  $\Omega$ potential,  $M_3$ , and  $\chi_{33}$  remain valid if according to the results of Sec. 2, we replace H by  $H_3 = H \cos \theta$ . The expressions thus obtained determine the dependences of all these quantities on the direction of  $\mathbf{H}$  (under condition  $\eta \ll 1$ ). Other components of magnetization and magnetic susceptibility tensor are relatively small since they do not exhibit the giant anomaly.<sup>9</sup> As to the dependence of  $\zeta$  on the magnetic field, it can be neglected since the density of states is rather high for any  $\zeta$  in the case II.

Finally, it should be noted that the electron spectrum in the magnetic field and the magnetic susceptibility were calculated here in the one-electron approximation (no manifestations of many-particle effects in the orbital magnetization have been established experimentally so far). It is well known, however,<sup>18</sup> that the inclusion of electron-electron interaction can strongly modify the electron spectrum, and hence the magnetic susceptibility for certain types of the band degeneracy. This question will be analyzed in a separate publication.

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