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Theory of the Magnetic Susceptibility of Bloch Electrons*

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It has heretofore always been assumed that the magnetic susceptibility of a crystal could be written $\chi = \chi_{\rm dia}^{\rm core} + \chi_{\rm dia}^{\rm val} + \chi_{\rm spin}^{\rm val}$, where $\chi_{\rm dia}^{\rm core}$ is the contribution of the core electrons, $\chi_{\rm dia}^{\rm val}$ is the contribution of the orbital motion of Bloch valence or conduction electrons completely neglecting spin, and $\chi_{\rm spin}^{\rm val}$ is the Pauli spin paramagnetism but with the free-electron g factor replaced by the effective g factor. The entire effect of spin-orbit coupling is assumed to be included in the effective g factor. We show that this is not the case and that there is a large fourth contribution to χ, the effect of the spin-orbit coupling on the orbital motion of the Bloch electrons $\chi_{\rm so}$. We construct a many-band Hamiltonian using the Bloch representation and derive the susceptibility directly from this Hamiltonian avoiding the ambiguity of the usual decoupling transformations. Our result agrees with the expression derived by Roth but is in a much more transparent form.

I. INTRODUCTION

The pioneering work on the quantum theory of diamagnetic susceptibility of free electrons was done by Landau[1] who showed that for a degenerate electron gas the diamagnetic susceptibility per unit volume is

$$\chi_L = -e^2 k_0 / 12\pi^2 mc^2 \,, \tag{1.1}$$

where k_0 is the wave number at the top of the Fermi surface. The expression for the spin susceptibility of free electrons obtained by Pauli[2] is three times larger than Landau diamagnetism and is of opposite sign. Therefore, a degenerate electron gas is always paramagnetic. However, the periodic potential in a solid changes the magnitudes of the diamagnetic and paramagnetic effects and also causes a coupling of the two effects through spin-orbit interaction.

The first step in understanding the diamagnetism of Bloch electrons was made by Peierls.[3] He constructed an effective Hamiltonian using wave functions obtained in a tight-binding approximation and obtained three terms for the magnetic susceptibility, the leading term of which reduces to the Landau formula in the case of free electrons and is called the Landau-Peierls susceptibility. However, in this theory, both the interband effect and the many-body effect had been ignored. Further, the tight-binding approximation is not valid for many solids.

Adams[4] stressed the importance of the interband terms in the effective Hamiltonian when the energy gaps are small. He gave a general treatment of the interband effect and then examined a simple example of two bands separated by a small energy gap produced by the Bragg reflection of a weak one-dimensional cosine potential. He considered two particular cases. The first is the case where the number of electrons in the upper band is small and so all of these electrons are influenced by Bragg reflection. The second case is that where the upper band contains a large number of electrons and so only a smaller fraction of the electrons are affected by the periodic potential. However, Adams's expression for the second case has the defect that in the limit of a vanishing periodic potential it gives a divergent result.

Wilson[5] obtained the density matrix directly as a power series in the magnetic field in terms of the solutions of the Schrödinger equation when the field is zero. The calculation of the susceptibility then becomes a computational problem, but in practice the computation becomes so intractable by the appearance of large numbers of complicated interband matrix elements that no satisfiactory expression could be derived by him. However, Hebborn and Sondheimer[6] have calculated, in a complicated way, an expression for the orbital magnetic susceptibility by using the density-matrix method.

Kjeldaas and Kohn[7] have applied a generalization of the Luttinger and Kohn[8] version of the effective-mass theory to the orbital susceptibility by taking into account fourth-order terms in $\vec{k}-\vec{k}_0$ (they put $\vec{k}_0=0$). However, their result has only limited usefulness since it is valid only for parabolic energy bands.

Blount[9] and Roth[10] have independently derived expressions for the magnetic susceptibility of Bloch electrons (including spin) by essentially equivalent methods. In their theories, the Hamiltonian of the Bloch electron in a magnetic field is first transformed into an effective many-band Hamiltonian by extremely complicated methods. The many-band Hamiltonian is then transformed into effective one-band Hamiltonians by successive similarity transformations through nonunitary operators. The Peierls effective Hamiltonian is obtained from their one-band Hamiltonian as the lowest-order approximation. However, apart from being very complicated, there are several difficulties in such decoupling procedures, which limit their usefulness. First, as Blount[9] has shown, this method of diagonalizing the Hamiltonian is only asymptotically convergent. In Roth's paper, the question of convergence was not answered. But Fishbeck[11] has shown that Roth's decoupling procedure is also asymptotically convergent. Therefore, as the magnetic field strength increases, these asymptotic solution methods gradually lose their validity. Second, there is reasonable doubt as to whether the interband matrix elements can be removed exactly in the case of bands whose energies are nearly equal and occasionally overlap (apart from the twofold degenerate bands due to spin which has been treated explicitly in the above decoupling procedures). Third, the decoupling procedure of the many-band Hamiltonian is not unique. A certain amount of arbitrariness is introduced in choosing the diagonal matrix elements of the nonunitary operator. Fourth, these theories require the use of a particular gauge and while the results must be gauge invariant, they have never been explicitly demonstrated to be so. Finally, there is no simple way to understand these results since they can be put in many apparently different but actually equivalent forms. There has been no attempt to calculate χ from these extremely long and involved results because of formidable computational difficulties.

Recently, there have been many attempts to calculate the magnetic susceptibility by making simplifying assumptions. Glasser[12] has derived an expression for the magnetic susceptibility in a nearly free-electron model. However, his result has the undesirable feature that the expression for susceptibility blows up when the Fermi surface touches the zone boundary. Fukuyama and Kubo[13] have calculated the magnetic susceptibility of bismuth by using the $\vec{k}\cdot\vec{p}$ perturbation model of Wolff,[14] which assumes two bands separated by a small energy gap. Buot[15] has calculated the magnetic susceptibility of bismuth-antimony alloys by using a similar two-band model.[16] These parametrized models have proved successful for bismuth and its alloys but it is desirable to have more complete calculations.

Due to the lack of a suitable theory of magnetic susceptibility with proper interpretation of the results, there exists some confusion[17] in the literature. Usually, it has been the practice to regard the total susceptibility as a sum of three terms:

$$\chi_{\text{tot}}=\chi_{\text{dia}}^{\text{core}}+\chi_{\text{dia}}^{\text{val}}+\chi_{\text{spin}}^{\text{val}}\,, \tag{1.2}$$

where val refers to valence or conduction electrons. It is well known[18,19] that due to spin-orbit interaction, the g factor of valence or conduction electrons can differ from the free-electron value of 2.0023. In fact, in the presence of very small band-gap energies, the effective g factor becomes orders of magnitude larger than that of free electrons. Therefore, it has become the usual practice to include the effect of spin-orbit interaction by substituting the effective g factor for the free-electron g factor in the Pauli spin susceptibility and to assume that the effect of spin-orbit interaction is completely accounted for. For example, the experimental orbital susceptibility[20] is determined by subtracting the theoretical value of the ionic susceptibility and the value of spin susceptibility (computed from the g factor which is obtained from electron-spin-resonance experiments) from the experimental value of the total susceptibility (measured directly). This value is then wrongly compared with the theoretical value which is calculated from the dynamics of the purely orbital motion of the Bloch electrons.[17] There is no reason to expect the spin-orbit interaction to affect only the g factor, and indeed we shall see that it yields an important additional contribution as well.

It is clear from the foregoing remarks that there remained a need for a theory of magnetic susceptibility of solids, which can be derived from first principles in a much simpler way than the present methods, would be free from the ambiguity of the usual diagonalization procedure, and would be valid for high magnetic fields. The present work was carried out as an attempt in this direction and we believe that we have been able to derive a satisfactory theory.

Our approach is different from the methods of Blount[9] and Roth[10] in the sense that we construct a many-band Hamiltonian in a simple way using the Bloch representation. Then, instead of constructing effective one-band Hamiltonians which are valid asymptotically in the magnetic field, we calculate the magnetic susceptibility directly from the many-band Hamiltonian. Thus, we contradict the usual notion[10] that the Bloch representation cannot be used to calculate the magnetic susceptibility. Our theory has the advantages that our derivation is much simpler, the ambiguity of the decoupling procedure is avoided in our method, and our results are valid for arbitrary magnetic fields since, as we shall see, when an expansion for different orders of magnetic field is made, it has infinite radius of convergence. Also, in our theory we do not use any particular gauge. Further, since our results can be interpreted clearly, the prevailing confusion in the theory of magnetic susceptibility has been clarified.

The expression for the magnetic susceptibility of Bloch electrons, which we shall derive, is of the form

$$\chi = \chi_0 + \chi_g + \chi_{so} , \qquad (1.3)$$

where χ_0 is the expression for diamagnetic susceptibility derived by Misra and Roth[21] by considering purely orbital motion of Bloch electrons, χ_g is the effective Pauli spin susceptibility which is obtained by replacing the free-electron g factor in the Pauli spin susceptibility[2] by the effective g factor,[19] and χ_{so} is an additional contribution of the spin-orbit interaction to the susceptibility.

However, we have adopted the Bloch picture of electrons in solids and thus we have not considered the electron-electron interaction terms except insofar as they can be approximated in a one-electron band calculation. However, the many-body effects can be shown to be small for χ_0 and χ_{so} as long as we do not have superconductivity.[22] For χ_g, the exchange enhancement effect will be very similar to that for free electrons except that the free-electron g factor is to be replaced by the effective g factor.

The planning of the paper is as follows. In Sec. II, we derive an effective many-band Hamiltonian using the Bloch representation. In Sec. III, we derive an expression for the magnetic susceptibility from our effective many-band Hamiltonian, in a form from which numerical calculations can be made. In Sec. IV, we summarize and discuss our results. In Appendixes A–C, we have proved certain identities which we have used in our derivation. In Appendix D, we show that certain terms in our expression for magnetic susceptibility can be lumped together to yield the effective Pauli susceptibility. In Appendix E, we show that our result is equivalent to Roth's result.[10]

II. MANY-BAND HAMILTONIAN

The Hamiltonian for an electron in a periodic potential $V(\vec{r})$ and an uniform magnetic field $\vec{B}$ is

$$H = \frac{1}{2m}\left(\vec{p} + \frac{e\vec{A}}{c}\right)^2 + V(\vec{r}) + \frac{\hbar}{4m^2c^2}\,\vec{\sigma}\cdot\left[\nabla V \times\left(\vec{p} + \frac{e\vec{A}}{c}\right)\right] + \frac{\hbar^2}{8m^2c^2}\,\nabla^2 V + \frac{1}{2} g_s \mu_B \vec{B}\cdot\vec{\sigma} , \qquad (2.1)$$

where $\vec{A}(\vec{r})$ is the vector potential, g_s is the free-electron g factor ($g_s = 2.0023$), μ_B is the Bohr magneton, $\vec{\sigma}$ is the Pauli spin operator, and the other symbols have their usual meanings. The eigenfunctions of the unperturbed Hamiltonian ($\vec{B} = 0$) are the Bloch functions

$$\phi_{n,\vec{k},\rho} = e^{i\vec{k}\cdot\vec{r}}\, U_{n,\vec{k},\rho} , \qquad (2.2)$$

where $U_{n,\vec{k},\rho}$ is a periodic two-component function, n is the band index, $\vec{k}$ is the reduced wave vector, and the index ρ, $\rho = 1$ or 2, distinguishes the two independent eigenfunctions $\phi_{n,\vec{k},1}$ and $\phi_{n,\vec{k},2}$ which belong to a general wave vector $\vec{k}$ and energy $E_n(\vec{k})$ if the crystal has inversion symmetry. Since the Bloch functions form a complete set, we can expand the wave function for an eigenstate of our problem as

$$\psi(\vec{r}) = \sum_{n,\vec{k},\rho} e^{i\vec{k}\cdot\vec{r}}\, U_{n,\vec{k},\rho}\, \psi_{n,\rho}(\vec{k}) , \qquad (2.3)$$

where $\psi_{n,\rho}(\vec{k})$ is periodic in $\vec{k}$. Substituting Eqs. (2.1) and (2.3) in the Schrödinger equation

$$H\psi(\vec{r}) = E\psi(\vec{r}) , \qquad (2.4)$$

we obtain

$$\sum_{n',\vec{k}',\rho'} \left\{ \frac{1}{2m}\left(\vec{p} + \frac{e\vec{A}(\vec{r})}{c}\right)^2 + V(\vec{r}) + \frac{\hbar}{4m^2c^2}\,\vec{\sigma}\cdot\left[\nabla V \times\left(\vec{p} + \frac{e\vec{A}(\vec{r})}{c}\right)\right] + \frac{\hbar^2}{8m^2c^2}\,\nabla^2 V + \frac{1}{2} g_s\, \mu_B \vec{B}\cdot\vec{\sigma} - E \right\} e^{i\vec{k}'\cdot\vec{r}}\, U_{n',\vec{k}',\rho'}\, \psi_{n',\rho'}(\vec{k}') = 0 . \qquad (2.5)$$

We have for the coordinate operator

$$\vec{r}\sum_{n',\vec{k}',\rho'} e^{i\vec{k}'\cdot\vec{r}}U_{n',\vec{k}',\rho'}\psi_{n',\rho'}(\vec{k}') = \sum_{n',\vec{k}',\rho'}[(-i\nabla_{k'})e^{i\vec{k}'\cdot\vec{r}}]U_{n',\vec{k}',\rho'}\psi_{n',\rho'}(\vec{k}') . \quad (2.6)$$

When we integrate by parts, the surface term vanishes since the expression is periodic in $\vec{k}$. Thus, we have

$$\vec{r}\sum_{n',\vec{k}',\rho'} e^{i\vec{k}\cdot\vec{r}}U_{n',\vec{k}',\rho'}\psi_{n',\rho'}(\vec{k}') = \sum_{n',\vec{k}',\rho'} e^{i\vec{k}'\cdot\vec{r}}\, i\nabla_{k'}U_{n',\vec{k}',\rho'}\psi_{n',\rho'}(\vec{k}') . \quad (2.7)$$

Therefore Eq. (2.5) can be written in the alternate form

$$\sum_{n',\vec{k}',\rho'} e^{i\vec{k}'\cdot\vec{r}}\left(\frac{1}{2m}(\vec{p}+\vec{\kappa}')^2+V(\vec{r})+\frac{\hbar}{4m^2c^2}\vec{\sigma}\cdot[\nabla V\times(\vec{p}+\vec{\kappa}')] + \frac{\hbar^2}{8m^2c^2}\nabla^2V+\frac{1}{2}g_s\mu_B\vec{B}\cdot\vec{\sigma}-E\right)U_{n',\vec{k}',\rho'}\psi_{n',\rho'}(\vec{k}')=0 , \quad (2.8)$$

where the operator $\vec{\kappa}$ is defined as

$$\vec{\kappa}=\hbar\vec{k}+(e/c)\vec{A}(i\nabla_k) , \quad (2.9)$$

and is the momentum space equivalent of the operator $\vec{p}+e\vec{A}/c$. Multiplying Eq. (2.8) on the left-hand side by $U^*_{n,\vec{k},\rho}e^{-i\vec{k}\cdot\vec{r}}$ and integrating over the crystal, we obtain

$$\int d\vec{r}\sum_{n',\vec{k}',\rho'} U^*_{n,\vec{k},\rho}(\vec{r})e^{i(\vec{k}'-\vec{k})\cdot\vec{r}}[H(\vec{r},\vec{p}+\vec{\kappa}')-E] \times U_{n',\vec{k}',\rho'}(\vec{r})\psi_{n',\rho'}(\vec{k}')=0 , \quad (2.10)$$

where

$$H(\vec{r},\vec{p}+\vec{\kappa})=\frac{1}{2m}(\vec{p}+\vec{\kappa})^2+V(\vec{r})+\frac{\hbar}{4m^2c^2}\vec{\sigma}\cdot[\nabla V\times(\vec{p}+\vec{\kappa})] +\frac{\hbar^2}{8m^2c^2}\nabla^2V+\frac{1}{2}g_s\mu_B\vec{B}\cdot\vec{\sigma} . \quad (2.11)$$

Since $U_{n,\vec{k},\rho}(\vec{r})$ and $H(\vec{r},\vec{p}+\vec{\kappa})$ are periodic in $\vec{r}$, we can break the integral in Eq. (10) into integrals over the unit cell and we obtain

$$\sum_{n',\vec{k}',\rho'}\int_{\text{cell}} d\vec{r}\,U^*_{n,\vec{k},\rho}e^{i(\vec{k}'-\vec{k})\cdot\vec{r}}\sum_{\vec{R}} e^{i(\vec{k}'-\vec{k})\cdot\vec{R}} \times [H(\vec{r},\vec{p}+\vec{\kappa}')-E]U_{n',\vec{k}',\rho'}(\vec{r})\psi_{n',\rho'}(\vec{k}')=0 . \quad (2.12)$$

Since the $\vec{k}$'s form a discrete set of points in the first zone, we have

$$\sum_{\vec{R}} e^{i(\vec{k}'-\vec{k})\cdot\vec{R}}=N\delta_{\vec{k},\vec{k}'} . \quad (2.13)$$

From Eqs. (2.12) and (2.13), we obtain

$$\sum_{n',\rho'}\int d\vec{r}\,U^*_{n,\vec{k},\rho}[H(\vec{r},\vec{p}+\vec{\kappa})-E]U_{n',\vec{k},\rho'}\psi_{n',\rho'}(\vec{k})=0 , \quad (2.14)$$

where the integration is over the crystal. This is a many-band Schrödinger equation which can be written in the alternative form

$$H(\vec{\kappa})\psi(\vec{k})=E\psi(\vec{k}) , \quad (2.15)$$

where $H(\vec{\kappa})$ is the effective Hamiltonian defined by

$$H_{n\rho,n'\rho'}(\vec{\kappa})=\int d\vec{r}\,U^*_{n,\vec{k},\rho}H(\vec{r},\vec{p}+\vec{\kappa})U_{n',\vec{k},\rho'} . \quad (2.16)$$

This is an effective many-band Hamiltonian. The usual procedure[9, 10] is to diagonalize the effective Hamiltonian by successive similarity transformations using nonunitary operators. However, this procedure, apart from being very complicated, is only asymptotically convergent in the magnetic field. Further, the decoupling procedure is not unique since there is ambiguity in choosing the diagonal matrix elements of the nonunitary operator. Therefore, we shall derive an expression for the magnetic susceptibility from the many-band Hamiltonian.

III. MAGNETIC SUSCEPTIBILITY OF BLOCH ELECTRONS

We shall now derive from first principles an expression for the magnetic susceptibility of Bloch electrons. The magnetic susceptibility is determined from the free energy by the relation

$$\chi=-\frac{\partial^2F}{\partial H^2} , \quad (3.1)$$

where F is the free energy

$$F=N\zeta-\frac{1}{\beta}\sum_i\ln(1+e^{-\beta(E_i-\zeta)}); \quad (3.2)$$

ζ is the chemical potential which can be regarded as a constant to the second order in magnetic field; $\beta=1/kT$; where k is the Boltzmann constant; T is the temperature; and H is the external magnetic field.

Let

$$\bar{F}(\beta)=F-N\zeta=-\frac{1}{\beta}\sum_i\ln(1+e^{-\beta(E_i-\zeta)}) . \quad (3.3)$$

If we expand $\bar{F}$ in different orders in H, it has only a small radius of convergence which vanishes at $T=0$. Therefore we make use of the Laplace transform of $\bar{F}$ since $e^{sH(\vec{\kappa})}$ has an infinite radius of convergence. $\bar{F}(\beta)$ can be related to the classical partition function by the method of Wilson and Sondheimer.[5, 23] For our purposes, this can be expressed in the form

$$\bar{F}(\beta)=\sum_i \bar{F}_i(\beta)=\sum_i T_\beta(s)\, e^{-sE_i}=T_\beta(s)\,\mathrm{Tr}\Phi(\vec{\kappa}) , \quad (3.4)$$

where $T_\beta(s)$ represents the inverse Laplace transform and

$$\Phi(\vec{\kappa})=e^{-sH(\vec{\kappa})} . \quad (3.5)$$

Let $\Phi(\vec{k})$ be the operator such that $\Phi(\vec{\kappa})$ can be formed from it by replacing $\hbar\vec{k}$ with $\vec{\kappa}$ in a symmetric manner. [$\Phi(\vec{k})$ would of course depend explicitly on the magnetic field.] We show in Appendix A that[24]

$$\mathrm{Tr}\Phi(\vec{\kappa})=\mathrm{Tr}\Phi(\vec{k}) . \quad (3.6)$$

We shall now evaluate $\Phi(\vec{k})$. From Eq. (3.5), we have

$$\frac{d\Phi(\vec{\kappa})}{ds}=-H(\vec{\kappa})\,\Phi(\vec{\kappa}) . \quad (3.7)$$

Using the multiplication theorem of Roth,[10] we obtain

$$\frac{d\Phi(\vec{k})}{ds}=-e^{-i\vec{h}\cdot\nabla_k\times\nabla_{k'}}\,H(\vec{k})\,\Phi(\vec{k}')\big|_{\vec{k}'=\vec{k}} , \quad (3.8)$$

where

$$\vec{h}=\frac{e\vec{B}}{2\hbar c}=\frac{eB}{2\hbar c}\,\vec{\lambda} , \quad (3.9)$$

$\vec{\lambda}$ is a unit vector in the direction of the magnetic field and $H(\vec{k})$ is the operator from which $H(\vec{\kappa})$ can be formed by replacing $\hbar\vec{k}$ with $\vec{\kappa}$ symmetrically, i.e.,

$$H(\vec{k})=\frac{1}{2m}(\vec{p}+\hbar\vec{k})^2+V(\vec{r})+\frac{\hbar}{4m^2c^2}\,\vec{\sigma}\cdot[\nabla V\times(\vec{p}+\hbar\vec{k})]+\frac{\hbar^2}{8m^2c^2}\nabla^2V+\frac{1}{2}g_s\mu_B\vec{B}\cdot\vec{\sigma} . \quad (3.10)$$

We write $H(\vec{k})$ in different orders of the magnetic field

$$H(\vec{k})=H_0(\vec{k})+H_1 , \quad (3.11)$$

where

$$H_0(\vec{k})=\frac{1}{2m}(\vec{p}+\hbar\vec{k})^2+V(\vec{r})+\frac{\hbar}{4m^2c^2}\,\vec{\sigma}\cdot[\nabla V\times(\vec{p}+\hbar\vec{k})]+\frac{\hbar^2}{8m^2c^2}\nabla^2V \quad (3.12)$$

and

$$H_1=\tfrac{1}{2}g_s\mu_B\vec{B}\cdot\vec{\sigma} . \quad (3.13)$$

Let

$$\Phi(\vec{k})=\Phi_0(\vec{k})+\Phi_1(\vec{k}) , \quad (3.14)$$

where

$$\Phi_0(\vec{k})=e^{-sH_0(\vec{k})} . \quad (3.15)$$

From Eqs. (3.8), (3.11), (3.14), and (3.15) we have

$$\frac{d\Phi_0(\vec{k})}{ds}+\frac{d\Phi_1(\vec{k})}{ds}=-H_0(\vec{k})\,\Phi_0(\vec{k})-H_0(\vec{k})\,\Phi_1(\vec{k})-H_1[\Phi_0(\vec{k})+\Phi_1(\vec{k})]+ih_{\alpha\beta}\,\nabla_k^\alpha H_0(\vec{k})\,\nabla_k^\beta[\Phi_0(\vec{k})+\Phi_1(\vec{k})]+\tfrac{1}{2}h_{\alpha\beta}h_{\gamma\delta}[\nabla_k^\alpha\nabla_k^\gamma H_0(\vec{k})]\nabla_k^\beta\nabla_k^\delta[\Phi_0(\vec{k})+\Phi_1(\vec{k})]+\cdots , \quad (3.16)$$

where

$$h_{\alpha\beta}=\epsilon_{\alpha\beta\gamma}h^\gamma , \quad (3.17)$$

$\epsilon_{\alpha\beta\gamma}$ is the complete antisymmetric tensor of the third rank, and we follow the Einstein summation convention. We note that the expansion in Eq. (3.8) and, consequently, the expansion in Eq. (3.16) has an infinite radius of convergence. However, in Eq. (3.16) we have neglected all terms higher than second-order terms in the magnetic field since for steady magnetic susceptibility (zero field) we need terms up to the second order. From Eqs. (3.15) and (3.16), we have

$$\frac{d\Phi_1(\vec{k})}{ds}=-H_0(k)\,\Phi_1(\vec{k})+\{ih_{\alpha\beta}[\nabla_k^\alpha H_0(\vec{k})]\nabla_k^\beta-H_1+\tfrac{1}{2}h_{\alpha\beta}h_{\gamma\delta}[\nabla_k^\alpha\nabla_k^\gamma H_0(\vec{k})]\nabla_k^\beta\nabla_k^\delta\}[\Phi_0(\vec{k})+\Phi_1(\vec{k})] . \quad (3.18)$$

Treating the second term on the right-hand side as an inhomogeneous term, this equation can be solved for Φ_1:

$$\Phi_1=e^{-sH_0}\int_0^s ds'\,e^{s'H_0}[ih_{\alpha\beta}(\nabla_k^\alpha H_0)\nabla_k^\beta-H_1+\tfrac{1}{2}h_{\alpha\beta}h_{\gamma\delta}(\nabla_k^\alpha\nabla_k^\gamma H_0)\nabla_k^\beta\nabla_k^\delta](\Phi_0+\Phi_1) . \quad (3.19)$$

We can now iterate this expression to obtain, up to the second order in the magnetic field,

$$\Phi_1\cong e^{-sH_0}\int_0^s ds'\,e^{s'H_0}[ih_{\alpha\beta}(\nabla_k^\alpha H_0)\nabla_k^\beta-H_1+\tfrac{1}{2}h_{\alpha\beta}h_{\gamma\delta}(\nabla_k^\alpha\nabla_k^\gamma H_0)\nabla_k^\beta\nabla_k^\delta]e^{-s'H_0}+e^{-sH_0}\int_0^s ds'\,e^{s'H_0}[ih_{\alpha\beta}(\nabla_k^\alpha H_0)\nabla_k^\beta-H_1]e^{-s'H_0}\times\int_0^{s'}ds''\,e^{s''H_0}[ih_{\gamma\delta}(\nabla_k^\gamma H_0)\nabla_k^\delta-H_1]e^{-s''H_0} . \quad (3.20)$$

In order to evaluate $\mathrm{Tr}\Phi_1(\vec{k})$, we simplify Eq. (3.20) by using the following operator expansion theorems [Eq. (5.18) and (5.19) of Misra and Roth[21]]:

$$\nabla_k^\alpha e^{-sH_0}=-e^{-sH_0}\int_0^s ds'\,e^{s'H_0}(\nabla_k^\alpha H_0)e^{-s'H_0} \quad (3.21)$$

and

$$\nabla_k^\alpha \nabla_k^\gamma e^{-sH_0} = -e^{-sH_0}\int_0^s ds'\, e^{s'H_0}(\nabla_k^\alpha \nabla_k^\gamma H_0)\, e^{-s'H_0} + e^{-sH_0}\int_0^s ds'\, e^{s'H_0}(\nabla_k^\alpha H_0)\, e^{-s'H_0}\int_0^{s'} ds''\, e^{s''H_0}(\nabla_k^\gamma H_0)\, e^{-s''H_0}$$

$$+ e^{-sH_0}\int_0^s ds'\, e^{s'H_0}(\nabla_k^\gamma H_0)\, e^{-s'H_0}\int_0^{s'} ds''\, e^{s''H_0}(\nabla_k^\alpha H_0)\, e^{-s''H_0} \, . \quad (3.22)$$

We shall now take the trace of Eq. (3.20), with the help of Eqs. (3.21) and (3.22), with the function $U_{n,\vec{k},\rho}(\vec{r})$. We first introduce some notations and some preliminary algebra. We write

$$\vec{\pi}_{n,\rho,m\rho'} = \int d\vec{r}\; U^*_{n,\vec{k},\rho}(\vec{r})(\nabla_k H_0)\, U_{m,\vec{k},\rho'}(\vec{r})$$

$$= \hbar\int d\vec{r}\; U^*_{n,\vec{k},\rho}(\vec{r})\left(\frac{(\vec{p}+\hbar\vec{k})}{m} + \frac{\hbar}{4m^2c^2}\,\vec{\sigma}\times\nabla V\right) \times U_{m,\vec{k},\rho'}(\vec{r}) \, , \quad (3.23)$$

where $\vec{\pi}/\hbar$ is the velocity operator without the field and

$$\vec{\sigma}_{n\rho,m\rho'} = \int d\vec{r}\; U^*_{n,\vec{k},\rho}(\vec{r})\,\vec{\sigma}\, U_{m,\vec{k},\rho'}(\vec{r}) \, . \quad (3.24)$$

It can be easily shown that[25]

$$\vec{\pi}_{n\rho,n\bar{\rho}} = 0 \, , \quad (3.25)$$

where $\bar{\rho}$ is the state conjugate to ρ. We also write

$$\Phi_0(E_n) = e^{-sE_n} \, , \quad (3.26)$$

$$\Phi_0'(E_n) = -se^{-sE_n} \, , \quad (3.27)$$

$$\Phi_0''(E_n) = s^2 e^{-sE_n} \, , \quad (3.28)$$

and

$$\Phi_0'''(E_n) = -s^3 e^{-sE_n} \, . \quad (3.29)$$

We also use $\nabla_k^\alpha \nabla_k^\beta H = (\hbar^2/m)\,\delta_{\alpha\beta}$ and the identity which can easily be proved[26]

$$h_{\alpha\beta}\, h_{\gamma\delta}(M_1^\alpha M_2^\beta M_3^\gamma M_4^\delta + M_1^\alpha M_2^\gamma M_3^\delta M_4^\beta) = h_{\alpha\beta}\, h_{\gamma\delta} M_1^\alpha M_2^\gamma M_3^\beta M_4^\delta \, , \quad (3.30)$$

where M_1, M_2, M_3, and M_4 are any matrix elements. We now take the trace of Φ_1 with the functions $U_{n,\vec{k},\rho}(\vec{r})$, which for a fixed $\vec{k}$ form a complete set for periodic functions. We note that $\Phi_1(\vec{k})$ is periodic in $\vec{r}$ since $H_0(\vec{k})$ and H_1 are periodic in $\vec{r}$. We also adopt the convention that any running index means that the sum over all the bands and all the spin indices shall be taken, except that all band terms equal to n have been explicitly separated out. Then after considerable algebra, we obtain

$$\mathrm{Tr}\Phi_1(\vec{k}) = \sum_{\vec{k}} ih_{\alpha\beta}\, \pi^\alpha_{n\rho,m\rho'}\pi^\beta_{m\rho',n\rho}\left(\frac{2\Phi_0(E_n)}{E_{mn}^2} + \frac{\Phi_0'(E_n)}{E_{mn}}\right) + \frac{1}{2} g_s \mu_B B\lambda^\gamma \sigma^\gamma_{n\rho,n\rho}\;\Phi_0'(E_n)$$

$$+ h_{\alpha\beta}h_{\gamma\delta}\left[-\frac{\hbar^2 \pi^\alpha_{n\rho,n\rho}\pi^\gamma_{n\rho,n\rho}}{6m}\,\delta_{\beta\delta}\,\Phi_0'''(E_n) - \frac{\hbar^4\Phi_0''(E_n)}{4m^2}\,\delta_{\alpha\gamma}\delta_{\beta\delta}\right.$$

$$+ \pi^\alpha_{n\rho,n\rho}\pi^\gamma_{n\rho,n\rho}\pi^\beta_{n\rho,m\rho'}\pi^\delta_{m\rho',n\rho}\left(\frac{\Phi_0'''(E_n)}{3E_{mn}} + \frac{2\Phi_0''(E_n)}{E_{mn}^2} + \frac{6\Phi_0'(E_n)}{E_{mn}^3} + \frac{8\Phi_0(E_n)}{E_{mn}^4}\right)$$

$$+ \pi^\alpha_{n\rho,m\rho'}\pi^\gamma_{m\rho',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}} + \frac{4\Phi_0'(E_n)}{E_{mn}^2} + \frac{8\Phi_0(E_n)}{E_{mn}^3}\right)\frac{\hbar^2\delta_{\beta\delta}}{m}$$

$$- \pi^\alpha_{n\rho,m\rho'}\pi^\gamma_{m\rho',n\rho''}\pi^\beta_{n\rho'',q\rho'''}\pi^\delta_{q\rho''',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}E_{qn}} + \frac{6\Phi_0'(E_n)}{E_{mn}^2E_{qn}} + \frac{8\Phi_0(E_n)}{E_{mn}^3E_{qn}} + \frac{4\Phi_0(E_n)}{E_{mn}^2E_{qn}^2}\right)$$

$$+ \pi^\alpha_{n\rho,m\rho'}\pi^\gamma_{m\rho',q\rho''}\pi^\beta_{q\rho'',l\rho'''}\pi^\delta_{l\rho''',n\rho}\left(\frac{2\Phi_0'(E_n)}{E_{ln}E_{qn}E_{mn}} + \frac{4\Phi_0(E_n)}{E_{mn}^2E_{ln}E_{qn}} + \frac{4\Phi_0(E_n)}{E_{ln}^2E_{mn}E_{qn}}\right)$$

$$\left.+ \pi^\alpha_{n\rho,n\rho}\pi^\gamma_{n\rho,m\rho'}\pi^\beta_{m\rho',q\rho''}\pi^\delta_{q\rho'',n\rho}\left(-\frac{2\Phi_0'(E_n)}{E_{mn}E_{qn}^2} + \frac{2\Phi_0'(E_n)}{E_{mn}^2E_{qn}} - \frac{4\Phi_0(E_n)}{E_{mn}E_{qn}^3} + \frac{4\Phi_0(E_n)}{E_{mn}^3E_{qn}}\right)\right]$$

$$+ \lambda^\gamma\lambda^\delta\left(\frac{g_s\mu_B B}{2}\right)^2\left(\frac{\sigma^\gamma_{n\rho,n\rho'}\sigma^\delta_{n\rho',n\rho}}{2}\,\Phi_0''(E_n) - \frac{\sigma^\gamma_{n\rho,m\rho'}\sigma^\delta_{m\rho',n\rho}}{E_{mn}}\,\Phi_0'(E_n)\right)$$

$$+ \frac{1}{2}\, ih_{\alpha\beta}g_s\mu_B B\lambda^\gamma\left[\sigma^\gamma_{n\rho,n\rho'}\pi^\alpha_{n\rho',m\rho''}\pi^\beta_{m\rho'',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}} + \frac{3\Phi_0'(E_n)}{E_{mn}^2} + \frac{4\Phi_0(E_n)}{E_{mn}^3}\right)\right.$$

$$- \pi^\alpha_{n\rho,m\rho'}\pi^\beta_{m\rho',q\rho''}\sigma^\gamma_{q\rho'',n\rho}\left(\frac{\Phi_0'(E_n)}{E_{qn}E_{mn}} + \frac{2\Phi_0(E_n)}{E_{mn}^2E_{qn}}\right) - \pi^\alpha_{n\rho,m\rho'}\sigma^\gamma_{m\rho',q\rho''}\pi^\beta_{q\rho'',n\rho}\left(\frac{\Phi_0'(E_n)}{E_{qn}E_{mn}} + \frac{2\Phi_0(E_n)}{E_{mn}^2E_{qn}} + \frac{2\Phi_0(E_n)}{E_{qn}^2E_{mn}}\right)$$

$$-\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}\left(\frac{\Phi_0'(E_n)}{E_{qn}E_{mn}}+\frac{2\Phi_0(E_n)}{E_{mn}E_{qn}^2}\right)+\pi^{\alpha}_{n\rho,n\rho}\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}\left(\frac{\Phi_0'(E_n)}{E_{mn}^2}+\frac{2\Phi_0(E_n)}{E_{mn}^3}\right)$$
$$-\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}\left(\frac{\Phi_0'(E_n)}{E_{mn}^2}+\frac{2\Phi_0(E_n)}{E_{mn}^3}\right)\Bigg]\ , \quad (3.31)$$

where, as indicated earlier, sums will be taken over all indices n, m, q, l, ρ, and ρ', but $n\neq m$, q, l. In the above, we have also used the notation

$$E_{mn}\equiv E_m(\vec{k})-E_n(\vec{k})\ . \quad (3.32)$$

In Appendix B we derive the following identity:

$$\sum_{\vec{k}}\Bigg[h_{\alpha\beta}h_{\gamma\delta}\left(2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}-4\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E_{mn}^3}\right)$$
$$+\frac{1}{2}ih_{\alpha\beta}g_s\mu_B B\lambda^{\gamma}\left(2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}}{E_{mn}^2}-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}}{E_{mn}^2}\right)\Bigg]\Phi_0'(E_n)$$
$$=\sum_{\vec{k}}\Bigg\{h_{\alpha\beta}h_{\gamma\delta}\Bigg[8\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E_{mn}^4}+8\,\frac{\hbar^2}{m}\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{E_{mn}^3}\,\delta_{\beta\delta}$$
$$-4\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}\left(\frac{2}{E_{mn}^3E_{qn}}+\frac{1}{E_{mn}^2E_{qn}^2}\right)$$
$$+4\,\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}\left(\frac{1}{E_{mn}^2E_{ln}E_{qn}}+\frac{1}{E_{ln}^2E_{mn}E_{qn}}\right)$$
$$-4\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}\left(\frac{1}{E_{mn}E_{qn}^3}-\frac{1}{E_{mn}^3E_{qn}}\right)\Bigg]$$
$$+\frac{1}{2}ih_{\alpha\beta}g_s\mu_B B\lambda^{\gamma}\Bigg[4\,\frac{\sigma^{\gamma}_{n\rho,n\rho'}\pi^{\alpha}_{n\rho',m\rho''}\pi^{\beta}_{m\rho'',n\rho}}{E_{mn}^3}-2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\sigma^{\gamma}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}$$
$$-2\pi^{\alpha}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}\left(\frac{1}{E_{mn}^2E_{qn}}+\frac{1}{E_{qn}^2E_{mn}}\right)-2\,\frac{\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}$$
$$+2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}}{E_{mn}^3}-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}}{E_{mn}^3}\Bigg]\Bigg\}\ \Phi_0(E_n)\ . \quad (3.33)$$

It can be easily shown from time reversal symmetry that[27]

$$\vec{\pi}_{n\rho,m\rho'}(\vec{k})=\pm\vec{\pi}_{m\bar{\rho}',n\bar{\rho}}(-\vec{k}) \quad (3.34)$$

and[19]

$$\vec{\sigma}_{n\rho,n\rho}(\vec{k})=-\sigma_{n\bar{\rho},n\bar{\rho}}(-\vec{k})\ . \quad (3.35)$$

Using $h_{\alpha\beta}=-h_{\beta\alpha}$ and the above we have for nonferromagnetic crystals

$$\sum_{\substack{n,m,\rho,\rho',\vec{k}\\ n\neq m}} ih_{\alpha\beta}\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}\left(\frac{2\Phi_0(E_n)}{E_{mn}^2}+\frac{\Phi_0'(E_n)}{E_{mn}}\right)+\sum_{n,\rho,\vec{k}}\tfrac{1}{2}g_s\mu_B B\lambda^{\gamma}\sigma^{\gamma}_{n\rho,n\rho}\Phi_0'(E_n)=0\ . \quad (3.36)$$

From Eqs. (3.31), (3.33), and (3.36) we obtain

$$\mathrm{Tr}\Phi_1(\vec{k})=h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\Bigg[-\frac{\hbar^2\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}}{6m}\,\delta_{\beta\delta}\Phi_0'''(E_n)-\frac{\hbar^4\Phi_0''(E_n)}{4m^2}\,\delta_{\alpha\gamma}\delta_{\beta\delta}$$
$$+\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}\left(\frac{\Phi_0'''(E_n)}{3E_{mn}}+\frac{2\Phi_0''(E_n)}{E_{mn}^2}+\frac{2\Phi_0'(E_n)}{E_{mn}^3}\right)+\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}}+\frac{4\Phi_0'(E_n)}{E_{mn}^2}\right)\frac{\hbar^2\delta_{\beta\delta}}{m}$$
$$-\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}E_{qn}}+\frac{6\Phi_0'(E_n)}{E_{mn}^2E_{qn}}\right)+2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}\Phi_0'(E_n)$$

$$-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',q\rho''}\,\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}\,\Phi_0'(E_n)+2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\gamma}_{n\rho,m\rho'}\,\pi^{\delta}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}\,\Phi_0'(E_n)\Bigg]$$

$$+\lambda^{\gamma}\lambda^{\delta}\left(\frac{g_s\mu_B B}{2}\right)^2\left(\frac{\sigma^{\gamma}_{n\rho,n\rho'}\,\sigma^{\delta}_{n\rho',n\rho}}{2}\,\Phi_0''(E_n)-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\sigma^{\delta}_{m\rho',n\rho}}{E_{mn}}\,\Phi_0'(E_n)\right)$$

$$+\frac{1}{2}\,ih_{\alpha\beta}\,g_s\,\mu_B\,B\lambda^{\gamma}\Bigg[\sigma^{\gamma}_{n\rho,n\rho'}\,\pi^{\alpha}_{n\rho',m\rho''}\,\pi^{\beta}_{m\rho'',n\rho}\left(\frac{\Phi_0''(E_n)}{E_{mn}}+3\,\frac{\Phi_0'(E_n)}{E_{mn}^2}\right)-\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',q\rho''}\,\sigma^{\gamma}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,\Phi_0'(E_n)$$

$$-\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,\Phi_0'(E_n)-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\alpha}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,\Phi_0'(E_n)$$

$$-\frac{\pi^{\alpha}_{n\rho,n\rho}\,\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',n\rho}}{E_{mn}^2}\,\Phi_0'(E_n)+\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',n\rho}}{E_{mn}^2}\,\Phi_0'(E_n)\Bigg]\ . \qquad (3.37)$$

We note from Eq. (3.4) that

$$\frac{\partial\overline{F}_i}{\partial E_i}=-T_\beta s e^{-sE_i}=T_\beta\,\Phi_0'(E_i)\ ,$$

and from Eq. (3.3) that

$$\frac{\partial\overline{F}_i}{\partial E_i}=f(E_i)\ ,$$

the Fermi function. Thus we obtain $\overline{F}(\beta)$ by operating on Eq. (3.37) with T_β which causes every factor $\Phi_0'(E_n)$, $\Phi_0''(E_n)$, $\Phi_0'''(E_n)$ to be replaced by $f(E_n)$, $f'(E_n)$, $f''(E_n)$.

Then from Eq. (3.1) we obtain the expression for the magnetic susceptibility of Bloch electrons[28]

$$\chi=\frac{2h_{\alpha\beta}h_{\gamma\delta}}{H^2}\sum_{\vec{k}}\Bigg[\frac{\hbar^2\,\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\gamma}_{n\rho,n\rho}}{6m}\,\delta_{\beta\delta}f''(E_n)+\frac{\hbar^4\,\delta_{\alpha\gamma}\,\delta_{\beta\delta}}{4m^2}f'(E_n)-\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\gamma}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\pi^{\delta}_{m\rho',n\rho}\left(\frac{f''(E_n)}{3E_{mn}}+\frac{2f'(E_n)}{E_{mn}^2}+\frac{2f(E_n)}{E_{mn}^3}\right)$$

$$-\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',n\rho}\left(\frac{f'(E_n)}{E_{mn}}+\frac{4f(E_n)}{E_{mn}^2}\right)\frac{\hbar^2\,\delta_{\beta\delta}}{m}+\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',n\rho''}\,\pi^{\beta}_{n\rho'',q\rho'''}\,\pi^{\delta}_{q\rho''',n\rho}\left(\frac{f'(E_n)}{E_{mn}E_{qn}}+\frac{6f(E_n)}{E_{mn}^2E_{qn}}\right)$$

$$-2\,\frac{\pi^{\alpha}_{n\rho,m\rho'},\,\pi^{\gamma}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',l\rho'''}\,\pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}\,f(E_n)+2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',q\rho''}\,\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}\,f(E_n)$$

$$-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\gamma}_{n\rho,m\rho'}\,\pi^{\delta}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}f(E_n)\Bigg]-\frac{\lambda^{\gamma}\lambda^{\delta}}{2}\,(g_s\mu_B)^2\left(\frac{\sigma^{\gamma}_{n\rho,n\rho'}\,\sigma^{\delta}_{n\rho',n\rho}}{2}\,f'(E_n)-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\sigma^{\delta}_{m\rho',n\rho}}{E_{mn}}\,f(E_n)\right)$$

$$-i\,\frac{h_{\alpha\beta}}{H}\,g_s\,\mu_B\lambda^{\gamma}\Bigg[\sigma^{\gamma}_{n\rho,n\rho'}\,\pi^{\alpha}_{n\rho',m\rho''}\,\pi^{\beta}_{m\rho'',n\rho}\left(\frac{f'(E_n)}{E_{mn}}+\frac{3f(E_n)}{E_{mn}^2}\right)-\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',q\rho''}\,\sigma^{\gamma}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,f(E_n)$$

$$-\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,f(E_n)-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\alpha}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\,f(E_n)$$

$$-\frac{\pi^{\alpha}_{n\rho,n\rho}\,\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',n\rho}}{E_{mn}^2}\,f(E_n)+\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',n\rho}}{E_{mn}^2}\,f(E_n)\Bigg]\ . \qquad (3.38)$$

This is the general expression for magnetic susceptibility of Bloch electrons. As mentioned earlier, the summation is over all bands except that $m,\ q,\ l\neq n$, since such terms have been explicitly considered. Thus there are no divergent terms in the expression for χ. We shall show in Appendix E that this expression is equivalent to the expression obtained by Roth.[10] However, we note that this expression can be written in many different ways by making use of partial integrations. Equation (3.38) is in a suitable form to make numerical calculations except that f'' terms should be converted to f' terms by partial integrations. However, it does not lead to an understanding of the different terms nor is it expressed in a familiar form. We shall now express χ in a form in which the various terms will have clear physical meaning. This will also clarify the prevailing confusion in the literature.[17]

We show in Appendix C that

$$h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \left(-\frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{3E_{mn}} + \frac{\hbar^2 \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho}}{6m} \delta_{\beta\delta} \right) f''(E_n)$$

$$+ \tfrac{1}{2} \left(\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\beta}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho} + \pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\delta}_{n\rho'',q\rho'''} \pi^{\beta}_{q\rho''',n\rho} \right) f'(E_n)(E_{mn}E_{qn})^{-1}$$

$$- \frac{\hbar^2 \pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho}}{mE_{mn}} \delta_{\beta\delta} f'(E_n) + \frac{\hbar^4 \delta_{\alpha\gamma} \delta_{\beta\delta}}{4m^2} f'(E_n) \Big] = \frac{h_{\alpha\beta} h_{\gamma\delta}}{12} \sum_{\vec{k}} \nabla^{\alpha}_{k} \nabla^{\gamma}_{k} E_n \nabla^{\beta}_{k} \nabla^{\delta}_{k} E_n f'(E_n) , \quad (3.39)$$

and in Appendix D that

$$\frac{h_{\alpha\beta} h_{\gamma\delta}}{H^2} \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho''} \pi^{\gamma}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E_{mn}E_{qn}} f'(E_n) - \lambda^{\gamma}\lambda^{\delta} \left(\frac{g_s \mu_B}{2} \right)^2 \sigma^{\gamma}_{n\rho,n\rho'} \sigma^{\delta}_{n\rho',n\rho} f'(E_n)$$

$$- i \frac{h_{\alpha\beta}}{H} g_s \mu_B \lambda^{\gamma} \frac{\sigma^{\gamma}_{n\rho,n\rho'} \pi^{\alpha}_{n\rho',m\rho''} \pi^{\beta}_{m\rho'',n\rho}}{E_{mn}} f'(E_n) = -\frac{1}{2} g^2 \mu_B^2 f'(E_n) , \quad (3.40)$$

where g is the effective g factor. It can also be shown by a partial integration similar to that shown in Appendix B for Eq. (3.33) that

$$2h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{E^2_{mn}} f'(E_n)$$

$$= h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \Big[2 \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\beta}_{n\rho,q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}} + 2 \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\delta}_{n\rho,q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}$$

$$- \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho} \left(\frac{1}{E^2_{mn}E_{qn}} - \frac{1}{E^2_{qn}E_{mn}} \right)$$

$$- \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}} + \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E^2_{qn}E_{mn}}$$

$$- 2 \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{E^3_{mn}} - 2 \frac{\hbar^2 \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{mE^2_{mn}} \delta_{\alpha\gamma} \Big] f(E_n). \quad (3.41)$$

From Eqs. (3.38), (3.39), (3.40), and (3.41), we obtain the expression for magnetic susceptibility of Bloch electrons:

$$\chi = \frac{h_{\alpha\beta} h_{\gamma\delta}}{6H^2} \sum_{\vec{k}} \nabla^{\alpha}_{k} \nabla^{\gamma}_{k} E_n \nabla^{\beta}_{k} \nabla^{\delta}_{k} E_n f'(E_n) - \sum_{\vec{k}} \frac{1}{2} g^2 \mu_B^2 f'(E_n)$$

$$+ \sum_{\vec{k}} \Big[\frac{2h_{\alpha\beta} h_{\gamma\delta}}{H^2} \Big(\frac{2\hbar^2 \pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho}}{mE^2_{mn}} \delta_{\beta\delta} + 2 \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\beta}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}} + 2 \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho''} \pi^{\gamma}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}$$

$$- 2 \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',l\rho'''} \pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}} - \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^2_{qn}} + \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}} \Big)$$

$$+ \tfrac{1}{2} \lambda^{\gamma}\lambda^{\delta} (g_s \mu_B)^2 \frac{\sigma^{\gamma}_{n\rho,m\rho'} \sigma^{\delta}_{m\rho',n\rho}}{E_{mn}} - i \frac{h_{\alpha\beta}}{H} g_s \mu_B \lambda^{\gamma} \Big(3 \frac{\sigma^{\gamma}_{n\rho,n\rho'} \pi^{\alpha}_{n\rho',m\rho''} \pi^{\beta}_{m\rho'',n\rho}}{E^2_{mn}} - \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',q\rho''} \sigma^{\gamma}_{q\rho'',n\rho}}{E_{qn}E_{mn}}$$

$$- \frac{\pi^{\alpha}_{n\rho,m\rho'} \sigma^{\gamma}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}} - \frac{\sigma^{\gamma}_{n\rho,m\rho'} \pi^{\alpha}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E_{mn}} - \frac{\pi^{\alpha}_{n\rho,n\rho} \sigma^{\gamma}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho}}{E^2_{mn}} + \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \sigma^{\gamma}_{m\rho',n\rho}}{E^2_{mn}} \Big) \Big] f(E_n) . \quad (3.42)$$

This is the general expression for magnetic susceptibility of Bloch electrons. The first term is the Landau-Peierls susceptibility[3] which would be the expression for the orbital magnetic susceptibility in an effective-mass formalism. The second term is the Pauli spin susceptibility, except that the free-electron g factor g_s has been replaced by the effective g factor g. This term then can be referred to as the effective Pauli spin susceptibility. The contribution of this term for crystals having large effective g factors (like Bi) would be very large but it would be always positive. This then is the "paramagnetic" contribution to magnetic susceptibility. The other terms have been written in terms of Fermi functions for convenience of numerical calculations.

We can write Eq. (3.42) in the alternate form

$$\chi = \chi_0 + \chi_g + \chi_{so} \; , \tag{3.43}$$

where χ_0 is the orbital contribution to the susceptibility

$$\chi_0 = \frac{h_{\alpha\beta}h_{\gamma\delta}}{6H^2} \sum_{\vec{k}} \nabla_k^\alpha \nabla_k^\gamma E_n \nabla_k^\beta \nabla_k^\delta E_n f'(E_n) + 2\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2} \sum_{\vec{k}} \left(-2\frac{\hbar^2 \pi^\alpha_{n\rho,m\rho'} \pi^\gamma_{m\rho',n\rho}}{mE^2_{mn}} \delta_{\beta\delta} + 2\frac{\pi^\alpha_{n\rho,m\rho'} \pi^\gamma_{m\rho',n\rho''} \pi^\beta_{n\rho'',q\rho'''} \pi^\delta_{q\rho''',n\rho}}{E^2_{mn} E_{qn}} \right.$$
$$\left. - 2\frac{\pi^\alpha_{n\rho,m\rho'} \pi^\gamma_{m\rho',q\rho''} \pi^\beta_{q\rho'',l\rho'''} \pi^\delta_{l\rho''',n\rho}}{E_{ln} E_{qn} E_{mn}} - \frac{\pi^\alpha_{n\rho,n\rho} \pi^\gamma_{n\rho,m\rho'} \pi^\delta_{m\rho',q\rho''} \pi^\beta_{q\rho'',n\rho}}{E_{mn} E^2_{qn}} + \frac{\pi^\alpha_{n\rho,n\rho} \pi^\beta_{n\rho,m\rho'} \pi^\gamma_{m\rho',q\rho''} \pi^\delta_{q\rho'',n\rho}}{E^2_{mn} E_{qn}} \right) f(E_n) \; , \tag{3.44}$$

χ_g is the effective Pauli spin susceptibility

$$\chi_g = -\textstyle\sum_{\vec{k}} \frac{1}{2} g^2 \mu_B^2 f'(E_n) \; , \tag{3.45}$$

and χ_{so} is the additional spin-orbit contribution to the magnetic susceptibility,

$$\chi_{so} = \sum_{\vec{k}} \left[-i\frac{h_{\alpha\beta}}{H} g_s \mu_B \lambda^\gamma \left(3\frac{\sigma^\gamma_{n\rho,n\rho'} \pi^\alpha_{n\rho',m\rho''} \pi^\beta_{m\rho'',n\rho}}{E^2_{mn}} - \frac{\pi^\alpha_{n\rho,n\rho'} \pi^\beta_{m\rho',q\rho''} \sigma^\gamma_{q\rho'',n\rho}}{E_{qn}E_{mn}} - \frac{\pi^\alpha_{n\rho,m\rho'} \sigma^\gamma_{m\rho',q\rho''} \pi^\beta_{q\rho'',n\rho}}{E_{qn}E_{mn}} \right. \right.$$
$$\left. - \frac{\sigma^\gamma_{n\rho,m\rho'} \pi^\alpha_{m\rho',q\rho''} \pi^\beta_{q\rho'',n\rho}}{E_{qn}E_{mn}} - \frac{\pi^\alpha_{n\rho,n\rho} \sigma^\gamma_{n\rho,m\rho'} \pi^\beta_{m\rho',n\rho}}{E^2_{mn}} + \frac{\pi^\alpha_{n\rho,n\rho} \pi^\beta_{n\rho,m\rho'} \sigma^\gamma_{m\rho',n\rho}}{E^2_{mn}} \right)$$
$$\left. + 4\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2} \frac{\pi^\alpha_{n\rho,m\rho'} \pi^\beta_{m\rho',n\rho''} \pi^\gamma_{n\rho'',q\rho'''} \pi^\delta_{q\rho''',n\rho}}{E^2_{mn}E_{qn}} + \frac{1}{2}\lambda^\gamma \lambda^\delta (g_s \mu_B)^2 \frac{\sigma^\gamma_{n\rho,m\rho'} \sigma^\delta_{m\rho',n\rho}}{E_{mn}} \right] f(E_n) \; . \tag{3.46}$$

It can be easily shown that our expression for χ_0 is the same as the general expression for diamagnetic susceptibility obtained by Misra and Roth[21, 29] [their Eq. (5.36)] if we replace $\vec{\pi}$ with $\vec{P}/m$.

IV. CONCLUSION

In this paper we have derived, in a reasonably simple fashion, an expression for the magnetic susceptibility of Bloch electrons. We first constructed an effective many-band Hamiltonian using the Bloch representation. Then we derived an expression for the magnetic susceptibility directly from the many-band Hamiltonian. Thus our method of derivation is simpler and we have avoided the ambiguity of the usual decoupling procedures and have not worked in a specific gauge. We made an expansion in different orders of magnetic field in the course of our calculation, but this expansion has infinite radius of convergence. We have thus contradicted the usual notion[10] that Bloch representation cannot be used to calculate the magnetic susceptibility. We have also shown that our result agrees with the earlier results[10,21] for the cases which they have treated.

Our result, which is expressed in a form such that numerical calculations become practical, can be expressed as the sum of three terms. The first term is the susceptibility obtained by considering the purely orbital motion of Bloch electrons.[21] The second term is the effective Pauli spin susceptibility[30] which is obtained by replacing the free-electron g factor in the Pauli susceptibility[2] with the effective g factor.[19] The third term is the additional spin-orbit contribution to the susceptibility. Although this term may contain contributions of either sign (as indeed does χ_o, the purely orbital term[20]), it should be considered a spin-orbit correction to χ_o and distinguished from the spin-orbit contribution to the effective g factor for the following reason. There are two types of contributions to the magnetic energy of a one-electron eigenstate, terms linear in $\vec{B}$ which split the spin degeneracy and terms quadratic in $\vec{B}$ which do not. (Both terms, of course, contribute quadratically to the free energy.) The linear terms are all included in the g factor[19] and are always paramagnetic independent of the sign of the g factor, i.e., independent of the sign of the splitting of the spin degeneracy. The quadratic terms which arise from a perturbation of the one-electron wave functions by the magnetic field are generally diamagnetic[20] and are responsible for both χ_o and χ_{so}.

It is easy to see that in the absence of spin-orbit coupling every term except the $h_{\alpha\beta}h_{\gamma\delta}$ term of χ_{so} vanishes. [Every σ_{nm} term vanishes because of the orthogonality of the orbital functions. If one chooses $\vec{H}$ to lie in the z direction, one has $\sum \sigma^z_{n\rho,n\rho'} = \sigma^z_{n\uparrow n\uparrow} + \sigma^z_{n\downarrow n\downarrow} = 0$. This, coupled with the fact that in the absence of spin-orbit coupling $\vec{\pi}_{n\uparrow,m\uparrow} = \vec{\pi}_{n\downarrow,m\downarrow}$ and $\vec{\pi}_{n\uparrow,m\downarrow} = 0$, makes the first term of Eq. (3.46) vanish.] If, in addition to the absence of spin-orbit coupling, the crystal has inversion symmetry, then $\pi^{\alpha}_{n\rho,m\rho} = \pi^{\alpha}_{m\rho,n\rho}$ and the $h_{\alpha\beta}h_{\gamma\delta}$ term also vanishes. Therefore, in a crystal with inversion symmetry this term properly belongs in χ_{so}, but in a crystal lacking inversion symmetry it belongs in χ_0.[31] A similar term occurs in the g factor where it causes g to differ from its free-electron value even in the absence of spin-orbit coupling if the crystal lacks an inversion center. These terms arise from an imperfect quenching of orbital angular momentum which can be caused either by the spin-orbit interaction or by a lack of inversion symmetry in the crystal.[32]

In a later paper we shall give numerical calculations for PbTe which, like bismuth, has large spin-orbit coupling and small energy gaps. For such cases the dominant term in χ_g is the first term of Eq. (3.40) and the dominant term in χ_{so} is the (next to last) term containing four π's in Eq. (3.46). In a two-band effective-mass model one can show, using these dominant terms only, that

$$\chi^c_{so} \approx (8E_F/3E_G)\chi_g ,$$

$$\chi^v_{so} \approx -\tfrac{1}{2}\pi(E_G/E_F)^{1/2}\chi_g , \qquad (4.1)$$

where E_g is the band gap, χ^{so}_v is the contribution of the filled valence band to χ_{so} and χ^c_{so} is the contribution of the carriers, either electrons or holes, to χ_{so}. χ_g is, of course, due only to carriers since it involves an integration over the Fermi surface. We note that both electrons and holes give a paramagnetic contribution to χ_{so} but that the filled valence band gives a larger diamagnetic contribution and that this contribution is several times larger than the g-factor paramagnetic susceptibility. Thus, systems with a large effective Pauli paramagnetism are actually diamagnetic.

APPENDIX A

We shall prove that[24]

$$\mathrm{Tr} f(\vec{\kappa}) = \mathrm{Tr} f(\vec{k}) , \qquad (A1)$$

where $f(\vec{\kappa})$ is a symmetric function of $\vec{\kappa}$ formed from $f(\vec{k})$ by replacing $\hbar\vec{k}$ with $\vec{\kappa}$ symmetrically. Since $f(\vec{k})$ is periodic in $\vec{k}$ space, we can expand it in a Fourier series

$$f(\vec{k}) = \sum_{\vec{R}} f_{\vec{R}} e^{i\vec{k}\cdot\vec{R}} . \qquad (A2)$$

Since $f(\vec{\kappa})$ is obtained from $f(\vec{k})$ by replacing $\hbar\vec{k}$ with $\vec{\kappa}$ symmetrically, one way of defining $f(\vec{\kappa})$ is

$$f(\vec{\kappa}) = \sum_{\vec{R}} f_{\vec{R}} e^{i\vec{\kappa}\cdot\vec{R}/\hbar} . \qquad (A3)$$

To take the trace, we use a complete set of plane waves $(1/\sqrt{N})e^{i\vec{k}\cdot\vec{R}'}$ over the Brillouin zone. (The factor of $1/\sqrt{N}$ is for normalization over the Brillouin zone.) So we have

$$\mathrm{Tr} f(\vec{\kappa}) = \frac{1}{N} \sum_{\vec{k},\vec{R}'} e^{-i\vec{k}\cdot\vec{R}'} f(\vec{\kappa}) e^{i\vec{k}\cdot\vec{R}'} . \qquad (A4)$$

We now consider one term in the expansion (A3), $f_{\vec{R}} e^{i\vec{\kappa}\cdot\vec{R}/\hbar}$, which operates on $e^{i\vec{k}\cdot\vec{R}'}$. We use the well-known identity

$$e^{A+B} = e^A e^B e^{[B,A]/2} , \qquad (A5)$$

which holds if $[B, A]$ commutes with both A and B. It can be shown by expanding $\vec{A}(i\nabla_k)$ in a Taylor series that

$$[(ie/\hbar c)\vec{A}(i\nabla_k)\cdot\vec{R},\ i\vec{k}\cdot\vec{R}] = -(ie/\hbar c) R_i A_{ij}(i\nabla_k) R_j , \qquad (A6)$$

where $A_{ij}(i\nabla_k) \equiv \partial A_j/\partial r_i (i\nabla_k)$ is evaluated by first differentiating A_j with respect to r_i and then substituting $i\nabla_k$ for $\vec{r}$. Using Eqs. (2.9), (A5), and (A6) we obtain

$$e^{i\vec{\kappa}\cdot\vec{R}/\hbar} = e^{i\vec{k}\cdot\vec{R}} e^{i(e/\hbar c)\vec{A}(i\nabla_k)\cdot\vec{R}} e^{-i(e/2\hbar c)R_i A_{ij}(i\nabla_k)R_j} . \qquad (A7)$$

We also use the identity

$$e^A e^B = e^B e^A e^{[A,B]} \qquad (A8)$$

to write

$$e^{i(e/\hbar c)\vec{A}(i\nabla_k)\cdot\vec{R}} e^{i\vec{k}\cdot\vec{R}'} = e^{i\vec{k}\cdot\vec{R}'} e^{i(e/\hbar c)\vec{A}(i\nabla_k)\cdot\vec{R}} \times e^{i(e/\hbar c)\vec{R}'_i A_{ij}(i\nabla_k)R_j} . \qquad (A9)$$

From Eqs. (A3), (A4), (A7), and (A9) we obtain

$$\mathrm{Tr} f(\vec{\kappa}) = \frac{1}{N} \sum_{\vec{R},\vec{R}',\vec{k}} f_{\vec{R}} e^{i\vec{k}\cdot\vec{R}} e^{i(e/\hbar c)\vec{A}(i\nabla_k)\cdot\vec{R}} \times e^{i(e/2\hbar c)(2R'_i - R_i)A_{ij}(i\nabla_k)R_j} . \qquad (A10)$$

We note that because there are no $\vec{k}$'s for them to operate on, $\vec{A}(i\nabla_k)$ and $A_{ij}(i\nabla_k)$ are constants and

$$\sum_{\vec{k}} e^{i\vec{k}\cdot\vec{R}} = 0 , \qquad \vec{R} \neq 0$$

$$= N , \qquad \vec{R} = 0 \qquad (A11)$$

so all phase factors vanish in Eq. (A10) and we obtain

$$\mathrm{Tr} f(\vec{\kappa}) = N f_0 . \qquad (A12)$$

It is evident from Eq. (A2) that

$$\mathrm{Tr} f(\vec{k}) = N f_0 . \qquad (A13)$$

From Eqs. (A12) and (A13) we obtain the identity

$$\mathrm{Tr} f(\vec{\kappa}) = \mathrm{Tr} f(\vec{k}) . \qquad (A14)$$

APPENDIX B

In order to prove the partial integrations in Eq. (3.33), we first wish to obtain expressions for $\nabla^{\alpha}_k \pi^{\beta}_{n\rho,m\rho'}$ and $\nabla^{\alpha}_k \sigma^{\beta}_{n\rho,m\rho'}$. We have

$$\nabla^{\alpha}_k \pi^{\beta}_{n\rho,m\rho'} = \nabla^{\alpha}_k \int d\vec{r}\, U^*_{n,\vec{k},\rho} (\nabla^{\beta}_k H) U_{m,\vec{k},\rho'}$$

$$= \int d\vec{r}\,(\nabla_k^\alpha U^*_{n,\vec{k},\rho})(\nabla_k^\beta H)\,U_{m,\vec{k},\rho'} + \int d\vec{r}\,U^*_{n,\vec{k},\rho}(\nabla_k^\alpha \nabla_k^\beta H)\,U_{m,\vec{k},\rho'} + \int d\vec{r}\,U^*_{n,\vec{k},\rho}(\nabla_k^\beta H)\,\nabla_k^\alpha U_{m,\vec{k},\rho'}\,. \tag{B1}$$

Since the $U_{n,\vec{k},\rho}$ are a complete set for periodic functions, we can insert the identity $|U_{q,\vec{k},\rho''}\rangle \times\langle U_{q,\vec{k},\rho''}|$ in the first and third terms. Therefore, we have

$$\nabla_k^\alpha \pi^\beta_{n\rho,m\rho'} = \sum_{\substack{q,\rho'' \\ q\neq n}} \int d\vec{r}(\nabla_k^\alpha U^*_{n,\vec{k},\rho})\,U_{q,\vec{k},\rho''} \int d\vec{r}'U^*_{q,\vec{k},\rho''}(\nabla_k^\beta H)\,U_{m,\vec{k},\rho'} + \sum_{\rho''} \int d\vec{r}(\nabla_k^\alpha U^*_{n,\vec{k},\rho})\,U_{n,\vec{k},\rho''} \int d\vec{r}'\,U^*_{n,\vec{k},\rho''}(\nabla_k^\beta H)\,U_{m,\vec{k},\rho'}$$
$$+ \sum_{\substack{q,\rho'' \\ q\neq m}} \int d\vec{r}\,U^*_{n,\vec{k},\rho}(\nabla_k^\beta H)\,U_{q,\vec{k},\rho''} \int d\vec{r}'\,U^*_{q,\vec{k},\rho''}\nabla_k^\alpha U_{m,\vec{k},\rho'}$$
$$+ \sum_{\rho''} \int d\vec{r}\,U^*_{n,\vec{k},\rho}(\nabla_k^\beta H)\,U_{m,\vec{k},\rho''} \int d\vec{r}'\,U^*_{m,\vec{k},\rho''}\nabla_k^\alpha U_{m,\vec{k},\rho'} + (\hbar^2/m)\,\delta_{\alpha,\beta}\,\delta_{n\rho,m\rho'}\,. \tag{B2}$$

We also have

$$\nabla_k^\alpha \int d\vec{r}\,U^*_{n,\vec{k},\rho} H U_{q,\vec{k},\rho'} = 0\,, \tag{B3}$$

from which we obtain

$$E_q \int d\vec{r}(\nabla_k^\alpha U^*_{n,\vec{k},\rho})U_{q,\vec{k},\rho'} + E_n \int d\vec{r}\,U^*_{n,\vec{k},\rho}\nabla_k^\alpha U_{q,\vec{k},\rho'} + \pi^\alpha_{n\rho,q\rho'} = 0\,, \tag{B4}$$

and

$$\nabla_k^\alpha \int d\vec{r}\,U^*_{n,\vec{k},\rho} U_{q,\vec{k},\rho'} = 0\,, \tag{B5}$$

from which we obtain

$$\int d\vec{r}(\nabla_k^\alpha U^*_{n,\vec{k},\rho})\,U_{q,\vec{k},\rho'} = -\int d\vec{r}\,U^*_{n,\vec{k},\rho}\,\nabla_k^\alpha U_{q,\vec{k},\rho'}\,. \tag{B6}$$

From Eqs. (B4) and (B6) we have for $q\neq n$

$$\int d\vec{r} U^*_{n,\vec{k},\rho}\nabla_k^\alpha U_{q,\vec{k},\rho'} = \pi^\alpha_{n\rho,q\rho'}/E_{qn}\,. \tag{B7}$$

We define

$$\vec{D}_{n\rho,n\rho'} \equiv \int d\vec{r} U^*_{n,\vec{k},\rho}\nabla_k U_{n,\vec{k},\rho'}\,. \tag{B8}$$

From Eqs. (B2), (B7), and (B8) we obtain

$$\nabla_k^\alpha \pi^\beta_{n\rho,m\rho'} = \sum_{\substack{q,\rho'' \\ q\neq n}} \frac{\pi^\alpha_{n\rho,q\rho''}\pi^\beta_{q\rho'',m\rho'}}{E_{nq}} + \sum_{\substack{q,\rho'' \\ q\neq m}} \frac{\pi^\beta_{n\rho,q\rho''}\pi^\alpha_{q\rho'',m\rho'}}{E_{mq}} + \frac{\hbar^2}{m}\delta_{\alpha,\beta}\delta_{n\rho,m\rho'} - \sum_{\rho''}(D^\alpha_{n\rho,n\rho''}\pi^\beta_{n\rho'',m\rho'} - \pi^\beta_{n\rho,m\rho''}D^\alpha_{m\rho'',m\rho'})\,. \tag{B9}$$

We can prove in a similar fashion

$$\nabla_k^\alpha \sigma^\beta_{n\rho,n\rho'} = \sum_{\substack{q,\rho'' \\ q\neq n}} \frac{\pi^\alpha_{n\rho,q\rho''}\sigma^\beta_{q\rho'',m\rho'}}{E_{nq}} + \sum_{\substack{q,\rho'' \\ q\neq m}} \frac{\sigma^\beta_{n\rho,q\rho''}\pi^\alpha_{q\rho'',m\rho'}}{E_{mq}} - \sum_{\rho''}(D^\alpha_{n\rho,n\rho''}\sigma^\beta_{n\rho'',m\rho'} - \sigma^\beta_{n\rho,m\rho''}D^\alpha_{m\rho'',m\rho'})\,. \tag{B10}$$

The partial integrations can be done in the following way. We first differentiate

$$h_{\alpha\beta}h_{\gamma\delta}\,\nabla_k^\alpha\left(\sum_{\substack{m,\rho',q,\rho'' \\ m,q\neq n}} \frac{\pi^\beta_{n\rho,m\rho'}\pi^\gamma_{m\rho',q\rho''}\pi^\delta_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}e^{-sE_n}\right)\,. \tag{B11}$$

When we differentiate the $q\neq m$ terms, we obtain the following (where the $l=m$ and $l=q$ terms are displayed explicitly):

$$h_{\alpha\beta}h_{\gamma\delta}\Bigg[\Bigg(\frac{\pi^\alpha_{n\rho,l\rho'''}\pi^\beta_{l\rho''',m\rho'}}{E_{nl}} + \frac{\pi^\alpha_{n\rho,m\rho'}\pi^\beta_{m\rho',m\rho'}}{E_{nm}} + \frac{\pi^\alpha_{n\rho,q\rho'''}\pi^\beta_{q\rho''',m\rho'}}{E_{nq}} + \frac{\pi^\beta_{n\rho,l\rho'''}\pi^\alpha_{l\rho''',m\rho'}}{E_{ml}} + \frac{\pi^\beta_{n\rho,n\rho}\pi^\alpha_{n\rho,m\rho'}}{E_{mn}} + \frac{\pi^\beta_{n\rho,q\rho'''}\pi^\alpha_{q\rho''',m\rho'}}{E_{mq}}$$
$$- D^\alpha_{n\rho,n\rho'''}\pi^\beta_{n\rho''',m\rho'} + \pi^\beta_{n\rho,m\rho'''}D^\alpha_{m\rho''',m\rho'}\Bigg)\frac{\pi^\gamma_{m\rho',q\rho''}\pi^\delta_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}e^{-sE_n}$$
$$+ \frac{\pi^\beta_{n\rho,m\rho'}}{E^2_{mn}E_{qn}}\Bigg(\frac{\pi^\alpha_{m\rho',l\rho'''}\pi^\gamma_{l\rho''',q\rho''}}{E_{ml}} + \frac{\pi^\alpha_{m\rho',q\rho''}\pi^\gamma_{q\rho'',q\rho''}}{E_{mq}} + \frac{\pi^\alpha_{m\rho',n\rho'''}\pi^\gamma_{n\rho''',q\rho''}}{E_{mn}} + \frac{\pi^\gamma_{m\rho',l\rho'''}\pi^\alpha_{l\rho''',q\rho''}}{E_{ql}} + \frac{\pi^\gamma_{m\rho',m\rho'}\pi^\alpha_{m\rho',q\rho''}}{E_{qm}}$$
$$+ \frac{\pi^\gamma_{m\rho',n\rho'''}\pi^\alpha_{n\rho''',q\rho''}}{E_{qn}} - D^\alpha_{m\rho',m\rho'''}\pi^\gamma_{m\rho''',q\rho''} + \pi^\gamma_{m\rho',q\rho'''}D^\alpha_{q\rho''',q\rho''}\Bigg)\pi^\delta_{q\rho'',n\rho}e^{-sE_n}$$
$$+ \frac{\pi^\beta_{n\rho,m\rho'}\pi^\gamma_{m\rho',q\rho''}}{E^2_{mn}E_{qn}}\Bigg(\frac{\pi^\alpha_{q\rho'',l\rho'''}\pi^\delta_{l\rho''',n\rho}}{E_{ql}} + \frac{\pi^\alpha_{q\rho'',n\rho'''}\pi^\delta_{n\rho''',n\rho}}{E_{qn}}$$
$$+ \frac{\pi^\alpha_{q\rho'',m\rho'''}\pi^\delta_{m\rho''',n\rho}}{E_{qm}} + \frac{\pi^\delta_{q\rho'',l\rho'''}\pi^\alpha_{l\rho''',n\rho}}{E_{nl}} + \frac{\pi^\delta_{q\rho'',q\rho''}\pi^\alpha_{q\rho'',n\rho}}{E_{nq}}$$

$$+\frac{\pi^{\delta}_{q\rho'',m\rho'''}\pi^{\alpha}_{m\rho''',n\rho}}{E_{nm}}-D^{\alpha}_{q\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}+\pi^{\delta}_{q\rho'',n\rho'''}D^{\alpha}_{n\rho''',n\rho}\Bigg)e^{-sE_n}$$

$$-2\,\frac{\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}\pi^{\alpha}_{n\rho,n\rho}}{E^3_{nm}E_{qn}}e^{-sE_n}+2\,\frac{\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}\pi^{\alpha}_{m\rho',m\rho'}}{E^3_{nm}E_{qn}}e^{-sE_n}$$

$$+\frac{\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}\pi^{\alpha}_{n\rho,n\rho}}{E^2_{mn}E^2_{qn}}e^{-sE_n}-\frac{\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}\pi^{\alpha}_{n\rho,n\rho'}}{E^2_{nm}E^2_{qn}}e^{-sE_n}\Bigg], \qquad \text{(B12)}$$

where we sum over all the band indices but $n\neq m\neq q\neq l$. We note that all the terms proportional to the matrix elements D cancel out. We similarly differentiate the $q=m$ terms in Eq. (B11) and then add these terms to the terms in Eq. (B12). We simplify the sum by interchanging band indices (except n) wherever necessary and by using the identity in Eq. (3.30). Then we lump together the diagonal terms in the band indices l, q, and m with the nondiagonal terms. Finally, since we have a summation over $\vec{k}$, it can be changed to an integration. In that case, the volume integral over the $\vec{k}$ space can be changed to a surface integral, and since the integrand is periodic in $\vec{k}$ the surface integral vanishes. Thus the sum is zero and so the term proportional to $se^{-sE_n}[=-\Phi_0'(E_n)]$ will be equal and opposite in sign to all the terms proportional to $e^{-sE_n}[=\Phi_0(E_n)]$. So finally we obtain

$$h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}s\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}e^{-sE_n}=h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\Bigg(2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E^2_{mn}E_{qn}E_{ln}}$$

$$+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^3_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{nm}E^2_{qn}}-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\pi^{\gamma}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^3_{mn}E_{qn}}$$

$$-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E^2_{qn}}+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E^2_{qn}}+\frac{\hbar^2\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{mE^3_{mn}}\delta_{\beta\delta}\Bigg)e^{-sE_n}, \qquad \text{(B13)}$$

where the sums are over $m,\rho',q,\rho'',l,\rho'''$ but $m,q,\ l\neq n$.

Similarly, we obtain

$$h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}s\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^2_{qn}}e^{-sE_n}$$

$$=h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\Bigg(-2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E^2_{ln}E_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E^2_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\pi^{\gamma}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^3_{qn}E_{mn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E^2_{qn}}+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn}E^2_{qn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^3_{qn}}-\frac{\hbar^2\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{mE^3_{mn}}\delta_{\beta\delta}\Bigg)e^{-sE_n}, \qquad \text{(B14)}$$

$$h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}s\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E^3_{nm}}e^{-sE_n}$$

$$=h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\Bigg(\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E^3_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\delta}_{n\rho,q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^3_{qn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{qn}E^3_{mn}}$$

$$-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E^2_{qn}E^2_{mn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^3_{mn}E_{qn}}$$

$$-2\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E^4_{mn}}-\frac{\hbar^2\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{mE^3_{mn}}\delta_{\beta\delta}\Bigg)e^{-sE_n}, \qquad \text{(B15)}$$

$$h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}s\,\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\delta}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}}{E^3_{nm}}e^{-sE_n}$$

$$= h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \left(\frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\delta}_{n\rho,q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E_{mn} E^3_{qn}} + \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\beta}_{n\rho,q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E_{mn} E^3_{qn}} + \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E_{mn} E^3_{qn}} \right.$$

$$+ \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E_{mn} E^3_{qn}} + \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn} E^2_{qn}}$$

$$\left. - 2 \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\delta}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho}}{E^4_{mn}} - \frac{\hbar^2}{m} \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho}}{E^3_{mn}} \delta_{\beta\delta} \right) e^{-sE_n} , \quad \text{(B16)}$$

$$h_{\alpha\beta} \lambda^{\gamma} \sum_{\vec{k}} s \frac{\pi^{\alpha}_{n\rho,m\rho'} \sigma^{\gamma}_{m\rho',n\rho} \pi^{\beta}_{n\rho,n\rho}}{E^2_{mn}} e^{-sE_n} = h_{\alpha\beta} \lambda^{\gamma} \sum_{\vec{k}} \left(- \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',q\rho''} \sigma^{\gamma}_{q\rho'',n\rho}}{E^2_{mn} E_{qn}} + \frac{\pi^{\alpha}_{n\rho,m\rho'} \sigma^{\gamma}_{m\rho',n\rho} \pi^{\beta}_{n\rho,n\rho}}{E^3_{mn}} \right.$$

$$\left. + \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho''} \sigma^{\gamma}_{n\rho'',n\rho}}{E^3_{mn}} \frac{- \pi^{\alpha}_{n\rho,m\rho'} \sigma^{\gamma}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn} E_{qn}} \right) e^{-sE_n} \quad \text{(B17)}$$

and

$$h_{\alpha\beta} \lambda^{\gamma} \sum_{\vec{k}} s \frac{\sigma^{\gamma}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho} \pi^{\alpha}_{n\rho,n\rho}}{E^2_{mn}} e^{-sE_n} = h_{\alpha\beta} \lambda^{\gamma} \sum_{\vec{k}} \left(- \frac{\pi^{\alpha}_{n\rho,q\rho''} \sigma^{\gamma}_{q\rho'',m\rho'} \pi^{\beta}_{m\rho',n\rho}}{E_{qn} E^2_{mn}} + \frac{\sigma^{\gamma}_{n\rho,n\rho'} \pi^{\alpha}_{n\rho',m\rho''} \pi^{\beta}_{m\rho'',n\rho}}{E^3_{mn}} \right.$$

$$\left. - \frac{\sigma^{\gamma}_{n\rho,m\rho'} \pi^{\alpha}_{m\rho',q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E^2_{qn} E_{nm}} + \frac{\sigma^{\gamma}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho} \pi^{\alpha}_{n\rho,n\rho}}{E^3_{mn}} \right) e^{-sE_n} . \quad \text{(B18)}$$

Using Eqs. (B13)–(B18) and the identities

$$h_{\alpha\beta} h_{\gamma\delta} \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\beta}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E^3_{mn} E_{qn}}$$

$$= h_{\alpha\beta} h_{\gamma\delta} \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\beta}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E_{mn} E^3_{qn}} , \quad \text{(B19)}$$

which is obtained by interchanging m and q and ρ and ρ'' in the summation, and

$$h_{\alpha\beta} h_{\gamma\delta} \sum_{\substack{m,q,\rho',\rho'',\rho''' \\ \rho''\neq\rho}} \left(\frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\beta}_{n\rho'',q\rho'''} \pi^{\delta}_{q\rho''',n\rho}}{E_{mn} E_{qn}} \right.$$

$$\left. + \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho''} \pi^{\delta}_{n\rho'',q\rho'''} \pi^{\beta}_{q\rho''',n\rho}}{E_{mn} E_{qn}} \right) = 0 , \quad \text{(B20)}$$

which has been essentially proved in Appendix E, we obtain the desired result of Eq. (3.33).

APPENDIX C

We shall now prove Eq. (3.39). We can write

$$h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \left(- \frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{3 E_{mn}} \right.$$

$$\left. + \frac{\hbar^2 \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho}}{6m} \delta_{\beta\delta} \right) f''(E_n)$$

$$= \frac{h_{\alpha\beta} h_{\gamma\delta}}{6} \sum_{\vec{k}} \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \left(\frac{\pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{E_{nm}} + \frac{\pi^{\delta}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho}}{E_{nm}} \right.$$

$$\left. + \frac{\hbar^2 \delta_{\beta\delta}}{m} \right) f''(E_n) . \quad \text{(C1)}$$

Also

$$\nabla_k E_n = \vec{\pi}_{n\rho,n\rho} , \quad \text{(C2)}$$

from which using (B9) we obtain

$$\nabla^{\beta}_k \nabla^{\delta}_k E_n = \left(\frac{\pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{E_{nm}} + \frac{\pi^{\delta}_{n\rho,m\rho'} \pi^{\beta}_{m\rho',n\rho}}{E_{nm}} + \frac{\hbar^2 \delta_{\beta\delta}}{m} \right) . \quad \text{(C3)}$$

Thus, the right-hand side of Eq. (C1) can be written in the alternate form

$$\tfrac{1}{6} h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \nabla^{\beta}_k \nabla^{\delta}_k E_n f''(E_n) , \quad \text{(C4)}$$

which can be shown by partial integration to be equal to

$$- \sum_{\vec{k}} \tfrac{1}{6} h_{\alpha\beta} h_{\gamma\delta} \nabla^{\alpha}_k \nabla^{\gamma}_k E_n \nabla^{\beta}_k \nabla^{\delta}_k E_n f'(E_n) . \quad \text{(C5)}$$

From Eqs. (C1) and (C5), we obtain

$$h_{\alpha\beta} h_{\gamma\delta} \sum_{\vec{k}} \left(\frac{\pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho} \pi^{\beta}_{n\rho,m\rho'} \pi^{\delta}_{m\rho',n\rho}}{3E_{nm}} \right.$$

$$\left. + \frac{\hbar^2 \pi^{\alpha}_{n\rho,n\rho} \pi^{\gamma}_{n\rho,n\rho}}{6m} \delta_{\beta\delta} \right) f''(E_n)$$

$$= - \frac{h_{\alpha\beta} h_{\gamma\delta}}{6} \sum_{\vec{k}} \nabla^{\alpha}_k \nabla^{\gamma}_k E_n \nabla^{\beta}_k \nabla^{\delta}_k E_n f'(E_n) . \quad \text{(C6)}$$

We can write

$$\frac{h_{\alpha\beta} h_{\gamma\delta}}{2} \left(\frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\beta}_{n\rho,q\rho''} \pi^{\delta}_{q\rho'',n\rho}}{E_{mn} E_{qn}} + \frac{\pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho} \pi^{\delta}_{n\rho,q\rho''} \pi^{\beta}_{q\rho'',n\rho}}{E_{mn} E_{qn}} + 2 \frac{\hbar^2 \pi^{\alpha}_{n\rho,m\rho'} \pi^{\gamma}_{m\rho',n\rho}}{m E_{nm}} \delta_{\beta\delta} + \frac{1}{2} \frac{\hbar^4 \delta_{\alpha\gamma} \delta_{\beta\delta}}{m^2} \right)$$

$$=\frac{h_{\alpha\beta}h_{\gamma\delta}}{4}\left(\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{E_{nm}}+\frac{\pi^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',n\rho}}{E_{nm}}+\frac{\hbar^2\delta_{\alpha\gamma}}{m}\right)\left(\frac{\pi^{\beta}_{n\rho,q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{nq}}+\frac{\pi^{\delta}_{n\rho,q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{nq}}+\frac{\hbar^2\delta_{\beta\delta}}{m}\right). \quad (C7)$$

From Eqs. (C3) and (C7) we have

$$\frac{h_{\alpha\beta}h_{\gamma\delta}}{2}\sum_{\vec{k}}\left[\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\delta}_{n\rho,q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}+2\frac{\hbar^2\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{mE_{nm}}\delta_{\beta\delta}+\frac{1}{2}\frac{\hbar^4\delta_{\alpha\gamma}\delta_{\beta\delta}}{m^2}\right]f'(E_n)$$

$$=\frac{h_{\alpha\beta}h_{\gamma\delta}}{4}\sum_{\vec{k}}\nabla^{\alpha}_{k}\nabla^{\gamma}_{k}E_n\nabla^{\beta}_{k}\nabla^{\delta}_{k}E_n f'(E_n)\,. \quad (C8)$$

Adding Eqs. (C6) and (C8) and using (B20), we obtain the desired result in Eq. (3.39).

APPENDIX D

We here prove Eq. (3.40). From Eqs. (4.6), (4.11), and (4.18) of Yafet[19] and noting that

$$\pi^{\beta}_{n\rho,m\rho''}\pi^{\alpha}_{m\rho'',n\rho'}h_{\alpha\beta}=-\pi^{\alpha}_{n\rho,m\rho''}\pi^{\beta}_{m\rho'',n\rho'}h_{\alpha\beta}\,,$$

we obtain

$$-\lambda^{\alpha}\lambda^{\beta}\mu_B^2(L_\alpha+\tfrac{1}{2}g_s\sigma_\alpha)_{n\rho,n\rho'}(L_\beta+\tfrac{1}{2}g_s\sigma_\beta)_{n\rho',n\rho}$$

$$=\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2}\frac{\pi^{\alpha}_{n\rho,m\rho''}\pi^{\beta}_{m\rho'',n\rho'}\pi^{\gamma}_{n\rho',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E_{mn}E_{qn}}$$

$$-\lambda^{\gamma}\lambda^{\delta}(\tfrac{1}{2}g_s\mu_B)^2\,\sigma^{\gamma}_{n\rho,n\rho'}\sigma^{\delta}_{n\rho',n\rho}$$

$$-i\frac{h_{\alpha\beta}}{H}\lambda^{\gamma}g_s\mu_B\frac{\sigma^{\gamma}_{n\rho,n\rho'}\pi^{\alpha}_{n\rho',m\rho''}\pi^{\beta}_{m\rho'',n\rho}}{E_{mn}}\,, \quad (D1)$$

where all repeated indices (including ρ) except n are summed over. (Note that our $\vec{\pi}=\hbar/m$ times Yafet's $\vec{\pi}$.) However, the left-hand side of Eq. (D1) is just

$$-(\tfrac{1}{2}\mu_B)^2\lambda^{\alpha}\lambda^{\beta}[(\tilde{G}_\alpha\tilde{G}_\beta)_{\uparrow\uparrow}+(\tilde{G}_\alpha\tilde{G}_\beta)_{\downarrow\downarrow}]\,, \quad (D2)$$

where

$$\tilde{G}_\alpha=\begin{pmatrix}G_{\alpha z} & G_{\alpha x}-iG_{\alpha y}\\ G_{\alpha x}+iG_{\alpha y} & -G_{\alpha z}\end{pmatrix} \quad (D3)$$

and the $G_{\alpha j}$ are defined in Yafet's equation (4.20). Inserting (D3) into (D2) one finds

$$\lambda^{\alpha}\lambda^{\beta}(\tilde{G}_\alpha\tilde{G}_\beta)_{\uparrow\uparrow}=\lambda^{\alpha}\lambda^{\beta}(\tilde{G}_\alpha\tilde{G}_\beta)_{\downarrow\downarrow}=\lambda^{\alpha}\lambda^{\beta}G_{\alpha j}G_{\beta j}\,, \quad (D4)$$

so that the left-hand side of (D1) becomes

$$-\tfrac{1}{2}\mu_B^2\lambda^{\alpha}\lambda^{\beta}G_{\alpha j}G_{\beta j}=-\tfrac{1}{2}\mu_B^2 g^2\,, \quad (D5)$$

where g^2 is defined in Yafet's equation (4.23). (Note the misplaced brackets therein.)

APPENDIX E

We shall now show that our expression for magnetic susceptibility [Eq. (3.42)] is equivalent to Roth's expression [Eq. (102) in Ref. 10]. Our first term is the same as χ_a of Roth. Our second term, as written on the right-hand side of Eq. (D1), is identical to Roth's χ_b. Roth's χ_c can be written in our notation as

$$\chi_c=\sum_{\vec{k}}\left[2\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2}\left(\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E_{nm}^2}+\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}-\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}\right)\right.$$

$$\left.+i\frac{h_{\alpha\beta}}{2H}g_s\mu_B\lambda^{\gamma}\left(\frac{\pi^{\alpha}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,n\rho}}{E_{mn}}-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',n\rho}\pi^{\beta}_{n\rho,n\rho}}{E_{mn}}\right)\right]f'(E_n)\,. \quad (E1)$$

It can be shown by partial integration (in a way outlined in Appendix B) that

$$h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\left(\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}-\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}\right)f'(E_n)$$

$$=h_{\alpha\beta}h_{\gamma\delta}\sum_{\vec{k}}\left(-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',l\rho'''}\pi^{\beta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E_{mn}^2E_{qn}}\right.$$

$$+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\pi^{\gamma}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E_{mn}^2E_{qn}}-\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}+\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}$$

$$\left.-\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{mn}E_{qn}^2}+\frac{1}{2}\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}^2E_{qn}}-\frac{\hbar^2\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}}{mE_{mn}^2}\delta_{\beta\delta}\right)f(E_n)\,, \quad (E2)$$

and

$$\tfrac{1}{2}h_{\alpha\beta}\lambda^{\gamma}\sum_{\vec{k}}\left(\frac{\pi^{\alpha}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,n\rho}}{E_{mn}}-\frac{\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',n\rho}\pi^{\beta}_{n\rho,n\rho}}{E_{mn}}\right)f'(E_n)$$

$$=-h_{\alpha\beta}\lambda^{\gamma}\sum_{\vec{k}}\left(\frac{\pi^{\beta}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',q\rho''}\pi^{\alpha}_{q\rho'',n\rho}}{E_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\sigma^{\gamma}_{n\rho'',n\rho}}{E^2_{mn}}\right)f(E_n)\ . \qquad \text{(E3)}$$

Therefore, Roth's χ_c can be written in the alternate form [with the help of Eq. (3.41)]

$$\chi_c=\sum_{\vec{k}}\Bigg[2\,\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2}\Bigg(-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',l\rho'''}\pi^{\beta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\delta}_{n\rho,q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\pi^{\gamma}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E^3_{mn}}-2\,\frac{\hbar^2\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho'n\rho}}{mE^2_{mn}}\,\delta_{\alpha\gamma}\Bigg)$$

$$+i\frac{h_{\alpha\beta}}{H}g_s\mu_B\lambda^{\gamma}\left(\frac{\pi^{\alpha}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho'q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\sigma^{\gamma}_{n\rho'',n\rho}}{E^2_{mn}}\right)\Bigg]f(E_n)\ . \qquad \text{(E4)}$$

Roth's χ_d can be written in the simplified form

$$\chi_d=\sum_{\vec{k}}\Bigg[2\,\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2}\Bigg(-\frac{1}{2}\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E_{qn}E^2_{mn}}-\frac{1}{2}\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\delta}_{n\rho,q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',n\rho}}{E^3_{mn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',q\rho''}\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\gamma}_{n\rho,m\rho'}\pi^{\delta}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^2_{qn}}-\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\pi^{\gamma}_{q\rho'',l\rho'''}\pi^{\delta}_{l\rho''',n\rho}}{E_{mn}E_{ln}E_{qn}}$$

$$+\frac{3}{2}\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\pi^{\gamma}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}\Bigg)+i\,\frac{h_{\alpha\beta}}{H}\,g_s\mu_B\lambda^{\gamma}\Bigg(\frac{\pi^{\alpha}_{n\rho,n\rho}\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho}}{E^2_{mn}}+\frac{\sigma^{\gamma}_{n\rho,m\rho'}\pi^{\alpha}_{m\rho',q\rho''}\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}$$

$$-\frac{\pi^{\alpha}_{n\rho,n\rho}\pi^{\beta}_{n\rho,m\rho'}\sigma^{\gamma}_{m\rho',n\rho}}{E^2_{mn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',q\rho''}\sigma^{\gamma}_{q\rho'',n\rho}}{E_{qn}E_{mn}}-2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\beta}_{m\rho',n\rho''}\sigma^{\gamma}_{n\rho'',n\rho}}{E^2_{mn}}\Bigg)$$

$$+\frac{1}{2}\,\lambda^{\gamma}\lambda^{\delta}(g_s\mu_B)^2\,\frac{\sigma^{\gamma}_{n\rho,m\rho'}\sigma^{\delta}_{m\rho',n\rho}}{E_{mn}}\Bigg]f(E_n)\ . \qquad \text{(E5)}$$

We now consider the terms

$$\sum_{\substack{m,q,\rho',\rho'',\rho'''\\ \rho''\neq\rho}}\left(\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\delta}_{n\rho'',q\rho'''}\pi^{\beta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}\right), \qquad \text{(E6)}$$

which can be written in the alternate form

$$\sum_{\substack{m,\rho',\rho''\\ \rho''\neq\rho}}\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}}{E^2_{mn}}\left[\sum_{q,\rho'''}\left(\frac{\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E_{qn}}+\frac{\pi^{\delta}_{n\rho'',q\rho'''}\pi^{\beta}_{q\rho''',n\rho}}{E_{qn}}\right)\right]\ . \qquad \text{(E7)}$$

Using Eq. (B9), this can be written

$$\sum_{\substack{m,\rho',\rho''\\ \rho''\neq\rho}}\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}}{E^2_{mn}}\nabla^{\beta}_{k}\pi^{\delta}_{n\rho'',n\rho} \qquad \text{(E8)}$$

which is zero from Eq. (3.25). Therefore, we have

$$\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\beta}_{n\rho,q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho}\pi^{\delta}_{n\rho,q\rho'''}\pi^{\beta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}$$

$$=\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\beta}_{n\rho'',q\rho'''}\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\pi^{\gamma}_{m\rho',n\rho''}\pi^{\delta}_{n\rho'',q\rho'''}\pi^{\beta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}\ . \qquad \text{(E9)}$$

We now add χ_c and χ_d in Eqs. (E4) and (E5) and with the help of Eqs. (3.30) and (E9), we obtain

$$\chi_c+\chi_d=\sum_{\vec{k}}\Bigg[\,2\frac{h_{\alpha\beta}h_{\gamma\delta}}{H^2}\Bigg(-2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',l\rho'''}\,\pi^{\delta}_{l\rho''',n\rho}}{E_{ln}E_{qn}E_{mn}}+2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',n\rho''}\,\pi^{\beta}_{n\rho'',q\rho'''}\,\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}$$

$$+2\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',n\rho''}\,\pi^{\gamma}_{n\rho'',q\rho'''}\,\pi^{\delta}_{q\rho''',n\rho}}{E^2_{mn}E_{qn}}+\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\pi^{\gamma}_{m\rho',q\rho''}\,\pi^{\delta}_{q\rho'',n\rho}}{E^2_{mn}E_{qn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\gamma}_{n\rho,m\rho'}\,\pi^{\delta}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E^2_{qn}}$$

$$-2\,\frac{\hbar^2\,\pi^{\beta}_{n\rho,m\rho'}\,\pi^{\delta}_{m\rho',n\rho}}{m\,E^2_{mn}}\,\delta_{\alpha\gamma}\Bigg)+i\,\frac{h_{\alpha\beta}}{H}\,g_S\,\mu_B\,\lambda^{\gamma}\Bigg(\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}-3\,\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',n\rho''}\,\sigma^{\gamma}_{n\rho'',n\rho}}{E^2_{mn}}$$

$$+\frac{\pi^{\alpha}_{n\rho,n\rho}\,\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',n\rho}}{E^2_{mn}}+\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\pi^{\alpha}_{m\rho',q\rho''}\,\pi^{\beta}_{q\rho'',n\rho}}{E_{mn}E_{qn}}-\frac{\pi^{\alpha}_{n\rho,n\rho}\,\pi^{\beta}_{n\rho,m\rho'}\,\sigma^{\gamma}_{m\rho',n\rho}}{E^2_{mn}}+\frac{\pi^{\alpha}_{n\rho,m\rho'}\,\pi^{\beta}_{m\rho',q\rho''}\,\sigma^{\gamma}_{q\rho'',n\rho}}{E_{qn}E_{mn}}\Bigg)$$

$$+\tfrac{1}{2}\lambda^{\gamma}\lambda^{\delta}(g_s\,\mu_B)^2\,\frac{\sigma^{\gamma}_{n\rho,m\rho'}\,\sigma^{\delta}_{m\rho',n\rho}}{E_{mn}}\Bigg]f(E_n)\quad. \tag{E10}$$

Comparing Eq. (E 10) with (3.42) we find that they are equal except for the first two terms of Eq. (3.42), which have been shown to be equal to χ_a and χ_b of Roth.[10]

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[17]There are many examples of this confusion in the literature. However, we give reference to the work of (a) W. D. Knight, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1956), Vol. 2, p. 93; (b) R. T. Schumacher and C. P. Slichter, Phys. Rev. 101, 58 (1956); (c) T. Kjeldaas and W. Kohn, *ibid.* 105, 806 (1957); (d) P. K. Misra and L. M. Roth, *ibid.* 177, 1089 (1969); (e) K. Chhotray and P. K. Misra, Phys. Letters 32A, 217 (1970); (f) 33A, 69 (1970); (g) P. Jena, T. P. Das, and S. D. Mahanti, Phys. Rev. B 1, 432 (1970); (h) R. Dupree and D. J. W. Geldart, Solid State Commun. 9, 145 (1971).

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[20]The orbital magnetic susceptibility, although it is over-all diamagnetic, does contain paramagnetic terms. See Ref. 6 for a discussion of Van Vleck paramagnetic contributions to the orbital susceptibility.

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[24]We are grateful to Dr. Laura M. Roth for sending us her unpublished proof which we have generalized to be applicable in an arbitrary gauge.

[25]This follows immediately from the orthogonality and degeneracy of the ρ and $\bar{\rho}$ states or from Yafet's (Ref. 19) theorem (3.4). If the crystal lacks a center of inversion, the spin degeneracy does not occur and the ρ and $\bar{\rho}$ subscripts should be absorbed into the n and m subscripts.

[26]Without loss of generality one can choose the z axis along the direction of $\vec{B}$. Then $h_{xy}=-h_{yx}=eB/2\hbar c$ and all other $h_{ij}=0$ whence follows Eq. (3.30) immediately.

[27]$\vec{\pi}_{n\rho,m\rho'}(\vec{k})=(\psi_{n\vec{k}\rho},\,\vec{\pi}\psi_{m\vec{k}\rho'})=(\vec{\pi}\psi_{n\vec{k}\rho},\,\psi_{m\vec{k}\rho'})=(K\psi_{m\vec{k}\rho'},\,K\vec{\pi}\psi_{n\vec{k}\rho})$, where K is the antiunitary time-reversal operator and we have used the hermiticity of $\vec{\pi}$. Now $K\vec{\pi}=-\vec{\pi}K$ and $K\psi_{m,\vec{k},\rho}=\pm\psi_{m,-\vec{k},\bar{\rho}}$ which yields Eq. (3.34), the positive sign holding if $\rho=\bar{\rho}'$ and the negative if $\rho=\rho'$.

[28]Since $\chi\sim10^{-6}$, at this point we ignore the difference between $\vec{B}$ and $\vec{H}$.

[29]There is an error of factor of 2 in the last four terms in the expression for χ of Misra and Roth [Ref. 17(d)]. This is evident if we compare their Eqs. (5.35) and (5.36). However, this error has been corrected in their subsequent derivations.

[30]This is the term determined from electron spin resonance and usually called the paramagnetic susceptibility.

[31]Misra and Roth [Ref. 17(d)], because they implicitly assumed inversion symmetry, failed to obtain this term.

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