evidence that the ground state of the Hubbard Hamiltonian can ever by ferromagnetic. For any spin state, there are many opportunities for hopping without having an extra pair of two electrons on the same atom. Since a nonmagnetic state has the possibility of lowering the energy by allowing an optimum number of doubly occupied atoms, while the fully ferromagnetic state has no such opportunity, it is very unlikely that the exact ground state is ferromagnetic.

In the case of a linear chain of atoms with the hopping matrix elements $\epsilon_{RR'}$ nonzero only for nearest neighbors, Lieb and Mattis[10] have proved that the ground state is always a singlet or a doublet but never ferromagnetic. Hubbard[3] has calculated the ferromagnetic instability of a nonmagnetic state and shown that such instability can occur only if the density of states at the Fermi level is much higher than the average over the band.

These results incline one to suggest that the present model can never be ferromagnetic and the present treatment on the stability of an antiferromagnetic state against a nonmagnetic state will be justified even though the ferromagnetic instability is not considered. Ferromagnetism is expected to appear if the degeneracy of d electrons is explicitly taken into account, and then it becomes necessary to investigate the ferromagnetic instability of an antiferromagnetic solution. However, the spin polarization of bands which yields the antiferromagnetic stability against a nonmagnetic state is different from the splitting of spin-up and spin-down bands which leads to the ferromagnetic stability against the same nonmagnetic state. The ferromagnetic instability of an antiferromagnetic solution is not known, and we have to compare the energies of the two-distinct state to see which one of the solutions is more stable than the other.

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General Theory of Magnetic Pseudopotentials

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A general magnetic-pseudopotential theory has been developed for Bloch electrons in a magnetic field. This theory is a generalization of the earlier formulations of Misra and Roth, and Misra, and it includes the effects of spin and spin-orbit interaction. In this method, tight-binding and orthogonalized-plane-wave functions are constructed which have the symmetry of the magnetic Bloch functions and which form a complete set for the wave function of the Hamiltonian of the crystal in a magnetic field. These are used as basis states for the wave function of an eigenstate of the problem, and an effective Hamiltonian is obtained which includes the magnetic pseudopotential. The magnetic pseudopotentials due to Misra and Roth, and Misra, and zero-field pseudopotentials, are obtained from this general magnetic pseudopotential in appropriate limits. The expression for the magnetic pseudopotential has been obtained in a form such that it can be calculated to any order in the magnetic field. This expression is further simplified for metals. The immediate purpose of this formulation is to calculate the total magnetic susceptibility of metals and alloys.

I. INTRODUCTION

Recently, Misra and Roth[1] have introduced a modified pseudopotential method in the theory of Bloch electrons of simple metals in a magnetic field. Misra[2] has extended this method, which he referred to as the magnetic-pseudopotential method, so that it can be applied to metals with more than one atom per unit cell, to alloys, and to imperfect lattices. In this method, tight-binding and orthogonalized-plane-wave (OPW) functions are constructed which have the symmetry of the magnetic Bloch functions and which form a complete set for the wave function of the Hamiltonian of the crystal in a magnetic field. These are used as basis states for the wave function of an eigenstate of the problem and an effective Hamiltonian is obtained which differs from the effective Hamiltonians of Kohn,[3] Roth,[4] and Blount[5] in the sense that there is a magnetic pseudopotential in the Hamiltonian which can be calculated to any order in the magnetic field.

The magnetic-pseudopotential method is important in the theory of Bloch electrons in a magnetic field in two ways. Firstly, this method has proved to be very useful in the calculation of diamagnetic susceptibility of metals[1,6] since, in this approach, which is different from traditional band calculations, perturbation theory can be used. Secondly, it has been the usual practice to solve the problems of Bloch electrons in a magnetic field by using the asymptotic solution methods of Kohn,[3] Wannier and Fredkin,[7] Roth,[4] and Blount.[5] In these methods, which are essentially equivalent, the Hamiltonian of the Bloch electrons in a magnetic field is transformed into effective one-band Hamiltonians which are obtained in the form of an asymptotic expansion in powers of field strength. The solution of the general eigenvalue problem of a one-band Hamiltonian had been obtained by Zilberman[8] by application of the Wentzel-Kramers-Brillouin (WKB) method. He had found a close connection with the electron orbits of the semiclassical theory and confirmed Onsager's quantization rule.[9] Owing to this close relation between the problems with and without fields, experiments in magnetic fields are performed to obtain information about properties of solids in the absence of the field. However, as the experimental field strength is gradually increased this relation is gradually lost, since it is established by asymptotic solution methods which lose their validity. The only method available at present whose range of validity enlarges with increasing fields is the nearly-free-electron approximation. Therefore, it is necessary to reformulate the theory of Bloch electrons in a magnetic field in terms of the pseudopotential theory.

However, the magnetic-pseudopotential formulation of Misra and Roth,[1] and Misra[2] has only limited application since spin has not been included in their theory. Thus, as an example, it cannot be used to calculate the total magnetic susceptibility of metals and alloys. Further, in many problems of Bloch electrons in a magnetic field, the spin-orbit interaction plays an important role. An example is the diamagnetism of bismuth.[10] The magnetic-pseudopotential theory of Misra and Roth,[1] and Misra[2] cannot be applied to such problems.

In the present paper we formulate a general magnetic-pseudopotential theory for Bloch electrons in a magnetic field in which spin and spin-orbit interactions have been included. The magnetic pseudopotentials of Misra and Roth,[1] and Misra,[2] and the zero-field pseudopotentials[11-14] are obtained from this general pseudopotential in the appropriate limits. The immediate purpose of our formulation is to calculate the total magnetic susceptibility of metals using this theory, which we shall report in a future paper, but we hope that this method will be useful to solve problems of Bloch electrons in high magnetic fields.

In Sec. II we make a general formulation of the magnetic-pseudopotential theory. In Sec. III we obtain an expression for this pseudopotential in a form such that it can be calculated to any order in the magnetic field. We also show that the magnetic pseudopotentials of Misra and Roth,[1] and Misra,[2] and the zero-field pseudopotentials[11-14] are obtained in the appropriate limits. In Sec. IV we derive a much simpler expression for metals. In Sec. V we summarize and discuss our results.

II. GENERAL MAGNETIC-PSEUDOPOTENTIAL FORMULATION

We begin with the Hamiltonian for an electron in a periodic potential $V(\vec{r})$ and a constant magnetic field $\vec{H}$ described by the vector potential $\vec{A}(\vec{r})$:

$$\mathcal{H}=\frac{1}{2m}\left(\vec{p}+\frac{e\vec{A}}{c}+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V(\vec{r})\right)^2+\frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H}+V(\vec{r}) \,, \tag{1}$$

where $\vec{p}$ is the momentum operator, μ_B is the Bohr magneton, $\vec{\sigma}$ is the Pauli spin operator, $g_0\approx 2$ is the free-electron g factor, and e, m, and c have their usual meanings ($\hbar=1$ throughout the paper). This gives the spin-orbit interaction correctly to order ϵ/mc^2 and is gauge invariant.

It is well known that in the case of Bloch electrons in a magnetic field, the Bloch functions do not form a useful representation as there are singular interband matrix elements. Recently, Misra[15] has obtained a class of representation for Bloch electrons in a magnetic field (without considering spin) by using only the translational properties of the Hamiltonian. When the condition that these basis functions reduce to Bloch functions is included, a set of magnetic Bloch functions, first used by Roth,[4] is obtained. It has also been shown by Misra[16] that these functions are complete with respect to the wave function of an electron in a periodic potential and a uniform magnetic field. We shall use this representation with suitable modifications to include spin. This modified Bloch representation is defined as follows. Let $U_{n\vec{k}s}(\vec{r})$ be spinors,

$$U_{n\vec{k}s}(\vec{r}) = (U_{n\vec{k}\uparrow}(\vec{r}),\ U_{n\vec{k}\downarrow}(\vec{r}))\ , \tag{2}$$

where n is a band index, $\vec{k}$ is the wave vector, s is the spin index, and $\mathfrak{U}_{n\vec{k}}(\vec{r})$ are periodic functions of $\vec{r}$ of the Bloch function for zero magnetic field (without spin). We assume that $U_{n\vec{k}s}(\vec{r})$ are linearly related, in spin space, to the functions $\mathfrak{U}_{n\vec{k}}(\vec{r})$:

$$U_{n\vec{k}s} = \mathfrak{U}_{n\vec{k}}\chi_s\ . \tag{3}$$

Let $\underline{\kappa}$ be the Fourier transform of the free-particle kinetic-momentum operator,

$$\underline{\kappa} = \vec{k} + (e/c)\vec{A}(i\nabla_k)\ . \tag{4}$$

Then the basis functions in the magnetic field are[15,16]

$$\phi_{n\vec{k}s} = U_{n\underline{\kappa}^* s}\, e^{i\vec{k}\cdot\vec{r}}\ , \tag{5}$$

where the wave vector $\vec{k}$ has been replaced by the operator $\underline{\kappa}^*$ symmetrically in $U_{n\vec{k}s}$. Thus the wave function of the system has the form

$$\Psi(\vec{r}, t) = \sum_{n\vec{k}s} U_{n\underline{\kappa}^* s}\, e^{i\vec{k}\cdot\vec{r}}\, \psi_{ns}(\vec{k}, t)$$
$$= \sum_{n\vec{k}s} e^{i\vec{k}\cdot\vec{r}}\, U_{n\underline{\kappa}s}(\vec{r})\, \psi_{ns}(\vec{k}, t)\ , \tag{6}$$

the latter being obtained by an integration by parts.

In order to use the above representation for the crystal in the magnetic field, we shall first obtain the Bloch functions in the zero-magnetic-field case. Let Φ_{cs} be spinors pertaining to a state c where c stands for quantum numbers n, l, and m. We have

$$\Phi_{cs} = (\phi_{c\uparrow},\ \phi_{c\downarrow})\ . \tag{7}$$

The spinors Φ_{cs} are not necessarily the core eigenfunctions of the full Hamiltonian, but they are the eigenfunctions of the spinless Hamiltonian with eigenvalues $\epsilon_{cs} = \epsilon_c$. We assume, as is done in the theory of atomic states, that the actual core functions are linearly related in spin space to the no-spin core functions

$$\Phi_{cs} = \phi_c\,\chi_s\ . \tag{8}$$

We now construct the Bloch functions in a tight-binding approximation

$$\Phi_{c\vec{k}s}(\vec{r}) = \frac{1}{\sqrt{N}}\sum_j e^{i\vec{k}\cdot\vec{r}_j}\,\Phi_{cs}(\vec{r} - \vec{r}_j)\ , \tag{9}$$

where $\vec{r}_j$ is the position of the ions which can be of different types. So the periodic part is

$$U_{c\vec{k}s}(\vec{r}) = \frac{1}{\sqrt{N}}\sum_j e^{i\vec{k}\cdot(\vec{r}_j - \vec{r})}\,\phi_c(\vec{r} - \vec{r}_j)\,\chi_s\ . \tag{10}$$

We construct an orthogonalized plane wave

$$\Phi_{\vec{q},\vec{k},s}(\vec{r}) = e^{i(\vec{k}+\vec{q})\cdot\vec{r}}\chi_s - \sum_{cs'} |\,\Phi_{c\vec{k}s'}\rangle\langle\Phi_{c\vec{k}s'}|\,e^{i(\vec{k}+\vec{q})\cdot\vec{r}}\rangle\chi_s\ , \tag{11}$$

where $\vec{q}$ is a wave number. Thus the periodic part is

$$U_{\vec{q},\vec{k},s}(\vec{r}) = e^{i\vec{q}\cdot\vec{r}}\chi_s - \textstyle\sum_c |\mathfrak{U}_{c\vec{k}}\rangle\langle\mathfrak{U}_{c\vec{k}}|\,e^{i\vec{q}\cdot\vec{r}}\rangle\chi_s\ . \tag{12}$$

We now construct $U_{c\underline{\kappa}s}(\vec{r})$ and $U_{\vec{q},\underline{\kappa},s}(\vec{r})$ from these spinors by replacing $\vec{k}$ by $\underline{\kappa}$ symmetrically, i.e., in the exponential. We can show

$$\langle U_{c\underline{\kappa}s}(\vec{r})\,|\,U_{c'\underline{\kappa}s}(\vec{r})\rangle = \delta_{cc'}\ , \tag{13}$$

$$\langle U_{c\underline{\kappa}s}(\vec{r})\,|\,U_{\vec{q},\underline{\kappa}s}(\vec{r})\rangle = 0\ , \tag{14}$$

which is due to the neglect of the overlap function. Therefore, for the crystal in a magnetic field, the basis functions are the complete set

$$U_{c\underline{\kappa}^* s}(\vec{r})\,e^{i\vec{k}\cdot\vec{r}},\quad U_{\vec{q},\underline{\kappa}^*,s}(\vec{r})\,e^{i\vec{k}\cdot\vec{r}}\ ,$$

and these have the symmetry of the Bloch functions. Thus the wave function of an eigenstate of the problem is

$$\Psi(\vec{r}, t) = \sum_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} \times\left(\sum_{cs} U_{c\underline{\kappa}s}(\vec{r})\,\psi_{cs}(\vec{k}, t) + \sum_{\vec{q}s} U_{\vec{q}\underline{\kappa}s}(\vec{r})\psi_{\vec{q}s}(\vec{k}, t)\right)\ , \tag{15}$$

where $\psi_{cs}(\vec{k}, t)$ and $\psi_{\vec{q}s}(\vec{k}, t)$ are the time-dependent coefficients. We make the requirement $\psi_{\vec{q}s}(\vec{k}+\vec{q}', t) = \psi_{\vec{q}+\vec{q}',s}(\vec{k}, t)$, so that the summand is periodic in $\vec{k}$. Substituting Eqs. (1) and (15) in

$$\mathcal{H}\Psi(\vec{r}) = E\Psi(\vec{r})\ , \tag{16}$$

we obtain

$$\left[\frac{1}{2m}\left(\vec{p} + \frac{e\vec{A}}{c} + \frac{\mu_B}{2c}\,\vec{\sigma}\times\nabla V(\vec{r})\right)^2 + \frac{g_0\mu_B}{2}\,\vec{\sigma}\cdot\vec{H} + V(\vec{r})\right]\sum_{\vec{k}} e^{i\vec{k}\cdot\vec{r}}\left(\sum_{cs} U_{c\underline{\kappa}s}(\vec{r})\psi_{cs}(\vec{k}) + \sum_{\vec{q}s} U_{\vec{q},\underline{\kappa},s}(\vec{r})\psi_{\vec{q}s}(\vec{k})\right)$$
$$= E\sum_{\vec{k}} e^{i\vec{k}\cdot\vec{r}}\left(\sum_{cs} U_{c\underline{\kappa}s}(\vec{r})\,\psi_{cs}(\vec{k}) + \sum_{\vec{q}s} U_{\vec{q}\underline{\kappa}s}(\vec{r})\,\psi_{\vec{q}s}(\vec{k})\right)\ . \tag{17}$$

It can be shown by integrating parts that Eq. (17) can be written as

$$\sum_{\vec{k}} e^{i\vec{k}\cdot\vec{r}}\left[\frac{1}{2m}\left(\vec{p} + \underline{\kappa} + \frac{\mu_B}{2c}\,\vec{\sigma}\times\nabla V(\vec{r})\right)^2 + \frac{g_0\mu_B}{2}\,\vec{\sigma}\cdot\vec{H} + V(\vec{r})\right]\left(\sum_{cs} U_{c\underline{\kappa}s}(\vec{r})\,\psi_{cs}(\vec{k}) + \sum_{\vec{q}s} U_{\vec{q}\underline{\kappa}s}(\vec{r})\,\psi_{\vec{q}s}(\vec{k})\right)$$
$$= E\sum_{\vec{k}} e^{i\vec{k}\cdot\vec{r}}\left(\sum_{cs} U_{c\underline{\kappa}s}(\vec{r})\,\psi_{cs}(\vec{k}) + \sum_{\vec{q}s} U_{\vec{q},\underline{\kappa},s}(\vec{r})\,\psi_{\vec{q}s}(\vec{k})\right)\ . \tag{18}$$

Let

$$\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}, \vec{\sigma}) = \frac{1}{2m}\left(\vec{p}+\underline{\kappa}+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V(\vec{r})\right)^2 + \frac{g_0\mu_B}{2}\,\vec{\sigma}\cdot\vec{H} + V(\vec{r}) \;. \tag{19}$$

We now define the operator $V_{\underline{\kappa}s}$ such that

$$V_{\underline{\kappa}s}\, e^{i\vec{q}\cdot\vec{r}}\chi_s = -\sum_c \left[\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}, \vec{\sigma}) - E\right] |\mathfrak{u}_{c\underline{\kappa}}\rangle\langle\mathfrak{u}_{c\underline{\kappa}}|\, e^{i\vec{q}\cdot\vec{r}}\rangle\,\chi_s \;. \tag{20}$$

From Eqs. (18) and (20), we have

$$\sum_{c',\vec{k}',s'} e^{i\vec{k}'\cdot\vec{r}}\left[\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}', \vec{\sigma}) + V_{\underline{\kappa}'s'} - E\right] U_{c'\underline{\kappa}'s'}(\vec{r})\,\psi_{c's'}(\vec{k})' + \sum_{\vec{q}',\vec{k}',s'} e^{i\vec{k}'\cdot\vec{r}}\left[\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}', \vec{\sigma}) + V_{\underline{\kappa}'s'} - E\right] e^{i\vec{q}'\cdot\vec{r}}\chi_{s'}\,\psi_{\vec{q}'s'}(\vec{k}') = 0 \;. \tag{21}$$

Multiplying by $e^{-i(\vec{k}+\vec{q})\cdot\vec{r}}\chi^\dagger$ on the left, we have

$$\sum_{c'\vec{k}'s'}\int d\vec{r}\, e^{-i(\vec{q}+\vec{k}-\vec{k}')\cdot\vec{r}}\chi_s^\dagger\left[\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}', \vec{\sigma}) - E\right] U_{c'\underline{\kappa}'s'}\,\psi_{c's'}(\vec{k}') + \sum_{\vec{q}'\vec{k}'s'}\int d\vec{r}\, e^{-i(\vec{q}+\vec{k}-\vec{k}')\cdot\vec{r}}\chi_s^\dagger\left[\mathcal{H}(\vec{r}, \vec{p}+\underline{\kappa}', \vec{\sigma}) + V_{\underline{\kappa}'s'} - E\right]\chi_{s'}\psi_{\vec{q}'s'}(\vec{k}') = 0 \;. \tag{22}$$

We can also multiply Eq. (21) on the left by $U^\dagger_{c\underline{\kappa}s}e^{-i\vec{k}\cdot\vec{r}}$ and integrate over the crystal. Then we would obtain another equation relating $\psi_{cs}(\vec{k})$ and $\psi_{\vec{q}s}(\vec{k})$. We are at present concerned with the conduction-electron case, so we can eliminate ψ_{cs} from these equations. Then we shall obtain

$$\sum_{\vec{q}'s'}\left\{\int d\vec{r}\, e^{-i\vec{q}\cdot\vec{r}}\chi_s^\dagger\left[\frac{1}{2m}\left(\vec{p}+\underline{\kappa}+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V\right)^2 + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} + V(\vec{r}) + V_{\underline{\kappa}s} - E\right] e^{i\vec{q}'\cdot\vec{r}}\chi_{s'}\,\psi_{\vec{q}'s'}(\vec{k}) + I_{\vec{q}\vec{q}'ss'}\psi_{\vec{q}'s'}(\vec{k})\right\} = 0 \;, \tag{23}$$

where $I_{\vec{q}\vec{q}'ss'}$ is a complicated interaction part between core terms and conduction-electron terms. It can be shown that

$$I_{\vec{q}\vec{q}'ss'} = -N\sum_{c's''}\frac{1}{(\epsilon_{c'}-E)}\int d\vec{r}\,\Phi^\dagger_{\vec{q}\underline{\kappa}s}\frac{\vec{h}}{m}\cdot\vec{r}\times\left(\vec{p}+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V\right)\Phi_{c's''}(\vec{r})\int d\vec{r}'\,\Phi^\dagger_{c's''}(\vec{r}')\,\frac{\vec{h}}{m}\cdot\vec{r}'\times\left(\vec{p}'+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V\right)\Phi_{\vec{q}'\underline{\kappa}s'}$$
$$-N\sum_{c's''}\int d\vec{r}\, e^{-i(\underline{\kappa}+\vec{q})\cdot\vec{r}}\chi_s^\dagger\int d\vec{r}'\,\Phi^\dagger_{c's''}(\vec{r}')\left[\frac{\vec{h}}{m}\cdot\vec{r}'\left(\vec{p}'+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V\right) + \frac{h_{\alpha\beta}h_{\gamma\beta}}{2m}r'_\alpha r'_\gamma\right]\Phi_{\vec{q}'\underline{\kappa}s'}(\vec{r}')\,\Phi_{c's''}(\vec{r}) \;. \tag{24}$$

This part contributes to the susceptibility of the conduction electrons a term which is similar to the Van Vleck paramagnetism except that it is a contribution to the susceptibility of Bloch electrons due to the presence of the core. It also consists of matrix elements between OPW's and core terms. These terms are important for evaluation of the total magnetic susceptibility, but we shall not discuss them here since they are not included in the magnetic-pseudopotential formulation. We can now write Eq. (23) in the alternate form (excluding the interaction term)

$$\mathcal{H}(\underline{\kappa})\psi(\vec{k}) = E\psi(\vec{k}) \;, \tag{25}$$

where

$$\mathcal{H}(\underline{\kappa}) = \frac{1}{2m}\left(\vec{p}+\underline{\kappa}+\frac{\mu_B}{2c}\vec{\sigma}\times\nabla V\right)^2 + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} + W_{\underline{\kappa}s}(\vec{r}) \tag{26}$$

and $W_{\underline{\kappa}s}(\vec{r})$ is the magnetic pseudopotential

$$W_{\underline{\kappa}s}(\vec{r}) = V(\vec{r}) + V_{\underline{\kappa}s}(\vec{r}) \;. \tag{27}$$

Thus we have now obtained an effective Hamiltonian $\mathcal{H}(\underline{\kappa})$. However, it differs from the effective Hamiltonian of Roth[4] because we now have a magnetic pseudopotential in the Hamiltonian. As shown in Refs. 1 and 6, the calculation of the diamagnetic susceptibility is considerably simplified in the magnetic-pseudopotential formalism. In a future paper we shall show that our formulation of the magnetic pseudopotential considerably simplifies the calculation of the total magnetic susceptibility of metals which would include the spin-orbit interaction. We note that there are alternate ways of constructing the magnetic pseudopotential. For example, we can start with the empty lattice magnetic Bloch functions[17,18] and orthogonalize them to the "tight-binding" magnetic Bloch functions in the spirit of the OPW and construct a pseudopotential.

III. CALCULATION OF GENERAL MAGNETIC PSEUDOPOTENTIAL

In order to obtain an expression for the magnetic pseudopotential in a form such that it can be calculated to different orders of the magnetic field, we shall use the extended multiplication theorem of Roth.[2] This theorem states that if $A(\vec{k})$, $B(\vec{k})$, and $C(\vec{k})$ are symmetrized functions of $\vec{k}$ from which the operators $A(\underline{\kappa})$, $B(\underline{\kappa})$, and $C(\underline{\kappa})$ are formed by replacing $\vec{k}$ by $\underline{\kappa}$, and if

$$A(\underline{\kappa})B(\underline{\kappa})C(\underline{\kappa}) = D(\underline{\kappa}) \ , \tag{28}$$

then $D(\underline{\kappa})$ is the operator formed from

$$D(\vec{k}) = \exp[-i\vec{h}\cdot(\nabla_k\times\nabla_{k'} + \nabla_k\times\nabla_{k''} + \nabla_{k'}\times\nabla_{k''}]$$
$$\times A(\vec{k})B(\vec{k}')C(\vec{k}'')|_{k''=k'=k} \ , \tag{29}$$

where

$$\vec{h} = e\vec{H}/2c \tag{30}$$

and $\vec{H}$ is the magnetic field. Using this in Eq. (20) it can be shown that the matrix elements of the symmetrized function $V_{\vec{k}s}$ are given by

$$\langle\vec{q}'s'|V_{\vec{k}s}|\vec{q}s\rangle = -\frac{1}{N}\sum_{clm}\int d\vec{r}\,d\vec{r}'\,e^{-i\vec{q}'\cdot\vec{r}_l + i\vec{q}\cdot\vec{r}_m}\exp[-i(\vec{k}+\vec{q}')\cdot(\vec{r}-\vec{r}_l) + i(\vec{k}+\vec{q})\cdot\vec{r}']\chi_{s'}^{\dagger}\Big[\Big(1 - i\vec{h}\cdot(\vec{r}-\vec{r}_l)\times\vec{r}'$$
$$-\frac{h_{\alpha\beta}h_{\gamma\delta}}{2}(\vec{r}-\vec{r}_l)_\alpha(\vec{r}-\vec{r}_l)_\gamma r'_\beta r'_\delta\Big)\Big(\frac{p^2}{2m} + V(\vec{r}) + \frac{e}{4m^2c^2}\vec{p}\cdot(\vec{\sigma}\times\nabla V) + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} + \frac{\mu_B^2}{8mc^2}(\vec{\sigma}\times\nabla V)^2 - E\Big)$$
$$+\frac{\vec{h}}{m}\cdot(\vec{r}-\vec{r}_l)\times\vec{p} + \frac{h_{\alpha\beta}h_{\gamma\delta}}{2m}[(\vec{r}-\vec{r}_l)_\beta(\vec{r}-\vec{r}_l)_\delta\delta_{\alpha\gamma} + 2i(\vec{r}-\vec{r}_l)_\beta(\vec{r}-\vec{r}_l)_\gamma p_\alpha r'_\delta]$$
$$+\frac{e}{4m^2c^2}[1 - ih_{\gamma\delta}(\vec{r}-\vec{r}_l)_\gamma r'_\delta][(\vec{r}-\vec{r}_l)\cdot\nabla V(\vec{h}\cdot\vec{\sigma}) - (\vec{h}\cdot\nabla V)(\vec{r}-\vec{r}_l)\cdot\vec{\sigma}]\Big]\times\phi_c(\vec{r}-\vec{r}_l)\,\phi_c^*(\vec{r}')\chi_s \ , \tag{31}$$

where we have used the notation

$$h_{\alpha\beta} = \epsilon_{\alpha\beta\gamma}h_\gamma \ , \tag{32}$$

where $\epsilon_{\alpha\beta\gamma}$ is the antisymmetric tensor of third rank, and we use the Einstein summation convention. The matrix elements of $V_{\kappa s}$, the repulsive part of the magnetic pseudopotential, are obtained by replacing $\vec{k}$ by $\underline{\kappa}$ in the exponential (i.e., symmetrically). We note that for the calculation of the magnetic susceptibility it is sufficient to calculate the matrix elements of $V_{\vec{k}s}$. It can be shown that Eq. (31) can be written in the alternate form

$$\langle\vec{q}'s'|V_{\vec{k}s}|\vec{q}s\rangle = \langle\vec{q}'s'|V_{R\vec{k}}|\vec{q}s\rangle + \frac{1}{N}\sum_{clm}\int d\vec{r}\,d\vec{r}'\,e^{-i\vec{q}'\cdot\vec{r}_l + i\vec{q}\cdot\vec{r}_m}\exp[-i(\vec{k}+\vec{q}')\cdot(\vec{r}-\vec{r}_l) + i(\vec{k}+\vec{q})\cdot\vec{r}']\chi_{s'}^{\dagger}$$
$$\times\Big[i\vec{h}\cdot(\vec{r}-\vec{r}_l)\times\vec{r}'\Big(\frac{p^2}{2m} + V(\vec{r}) + \frac{e}{4m^2c^2}\vec{p}\cdot(\vec{\sigma}\times\nabla V) + \frac{\mu_B^2}{8mc^2}(\vec{\sigma}\times\nabla V)^2 + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} - E\Big)$$
$$-\frac{\vec{h}}{m}\cdot(\vec{r}-\vec{r}_l)\times\vec{p} - \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} + E_2 - \frac{e}{4m^2c^2}[(\vec{r}-\vec{r}_l)\cdot\nabla V(\vec{h}\cdot\vec{\sigma}) - (\vec{h}\cdot\nabla V)(\vec{r}-\vec{r}_l)\cdot\vec{\sigma}] + \frac{h_{\alpha\beta}h_{\gamma\delta}}{2}(\vec{r}-\vec{r}_l)_\alpha(\vec{r}-\vec{r}_l)_\gamma r'_\beta r'_\delta$$
$$\times\Big(\frac{p^2}{2m} + V(\vec{r}) + \frac{e}{4m^2c^2}\vec{p}\cdot(\vec{\sigma}\times\nabla V) + \frac{\mu_B^2}{8mc^2}(\vec{\sigma}\times\nabla V)^2 + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} - E\Big) - \frac{h_{\alpha\beta}h_{\gamma\delta}}{2m}[(\vec{r}-\vec{r}_l)_\beta(\vec{r}-\vec{r}_l)_\delta\delta_{\alpha\gamma}$$
$$+2i(\vec{r}-\vec{r}_l)_\beta(\vec{r}-\vec{r}_l)_\gamma p_\alpha r'_\delta] + \frac{ieh_{\gamma\delta}}{4m^2c^2}(\vec{r}-\vec{r}_l)_\gamma r'_\delta[(\vec{r}-\vec{r}_l)\cdot\nabla V(\vec{h}\cdot\vec{\sigma}) - (\vec{h}\cdot\nabla V)(\vec{r}-\vec{r}_l)\cdot\vec{\sigma}]\Big]\phi_c(\vec{r}-\vec{r}_l)\,\phi_c^*(\vec{r}')\chi_s \ , \tag{33}$$

where $\langle\vec{q}'s'|V_{R\vec{k}}|\vec{q}s\rangle$ are the matrix elements of the zero-field pseudopotential and E_2 is the contribution to the energy due to the spin part only. It can be easily shown that for zero spin, Eq. (33) reduces to the expression for the magnetic pseudopotential obtained by Misra.[2] It is also evident from Eq. (33) that in the limit of zero magnetic field the magnetic pseudopotential reduces to the zero-field pseudopotential,[13,14] as it should.

IV. MAGNETIC PSEUDOPOTENTIAL FOR METALS

In the case of metals, Eq. (33) can be further simplified. We have

$$V(\vec{r}) = \textstyle\sum_j v(\vec{r}-\vec{r}_j) \ . \tag{34}$$

Further, the zero-field pseudopotential can be separated into a sum of individual pseudopotentials centered upon the ions:

$$V_{R\vec{k}}=\sum_j \mathcal{V}_{R\vec{k}}(\vec{r}-\vec{r}_j) \quad . \tag{35}$$

From Eqs. (33)–(35) we have

$$\langle \vec{q}'s' | V_{\vec{k}s} | \vec{q}s \rangle = S(\vec{q},\vec{q}') \times \langle \vec{q}'s' | \mathcal{V}^{(0)}_{\vec{k}s} + \mathcal{V}^{(1)}_{\vec{k}s} + \mathcal{V}^{(2)}_{\vec{k}s} + \cdots | \vec{q}s \rangle , \tag{36}$$

where

$$S(\vec{q},\vec{q}')=\frac{1}{N^2}\sum_{l,m} e^{-i\vec{q}'\cdot\vec{r}_l + i\vec{q}\cdot\vec{r}_m} \tag{37}$$

and $\mathcal{V}^{(0)}_{\vec{k}s}$, $\mathcal{V}^{(1)}_{\vec{k}s}$, $\mathcal{V}^{(2)}_{\vec{k}s}$, ..., are the symmetrized functions of $\vec{k}$ to different orders in the magnetic field,

$$\langle \vec{q}'s' | \mathcal{V}^{(0)}_{\vec{k}s} | \vec{q}s \rangle = \langle \vec{q}'s' | \mathcal{V}_{R\vec{k}} | \vec{q}s \rangle = \int d\vec{r}\, d\vec{r}'\, e^{-i(\vec{q}'+\vec{k})\cdot\vec{r}}\, \chi^\dagger_{s'} \mathcal{V}_R\, e^{i(\vec{q}+\vec{k})\cdot\vec{r}'} \chi_s , \tag{38}$$

$$\langle \vec{q}'s' | \mathcal{V}^{(1)}_{\vec{k}s} | \vec{q}s \rangle = \frac{1}{\Omega_0}\sum_c \int d\vec{r}\, d\vec{r}'\, \exp[-i(\vec{k}+\vec{q}')\cdot\vec{r} + i(\vec{k}+\vec{q})\cdot\vec{r}']\chi^\dagger_{s'} \times\Big[i\vec{h}\cdot\vec{r}\times\vec{r}'\Big(\epsilon_c + \frac{e}{4m^2c^2}\vec{p}\cdot(\vec{\sigma}\times\nabla V) + \frac{\mu_B^2}{8mc^2}(\vec{\sigma}\times\nabla V)^2 - E\Big) - \frac{\vec{h}}{m}\cdot(\vec{L}+2\vec{S}) + E_2 - \frac{e}{4m^2c^2} \times[(\vec{r}\cdot\nabla V)(\vec{h}\cdot\vec{\sigma}) - (\vec{h}\cdot\nabla V)(\vec{r}\cdot\vec{\sigma})]\Big]\phi_c(\vec{r})\phi^*_c(\vec{r}')\chi_s , \tag{39}$$

$$\langle \vec{q}'s' | \mathcal{V}^{(2)}_{\vec{k}s} | \vec{q}s \rangle = \frac{1}{\Omega_0}\sum_c \int d\vec{r}\, d\vec{r}'\, \exp[-i(\vec{k}+\vec{q}')\cdot\vec{r} + i(\vec{k}+\vec{q})\cdot\vec{r}']\chi^\dagger_{s'} \Big[\frac{h_{\alpha\beta}h_{\gamma\delta}}{2} r_\alpha r_\gamma r'_\beta r'_\delta\Big(\epsilon_c + \frac{e}{4m^2c^2}\vec{p}\cdot(\vec{\sigma}\times\nabla V) + \frac{\mu_B^2}{8mc^2}(\vec{\sigma}\times\nabla V)^2 + \frac{g_0\mu_B}{2}\vec{\sigma}\cdot\vec{H} - E\Big) - \frac{h_{\alpha\beta}h_{\gamma\delta}}{2m} r_\beta r_\delta \delta_{\alpha\gamma} + \frac{ih_{\alpha\beta}}{m} r_\alpha r'_\beta \vec{h}\cdot(\vec{L}+2\vec{S}) + \frac{ieh_{\gamma\delta}}{4m^2c^2} r_\gamma r'_\delta[(\vec{r}\cdot\nabla V)(\vec{h}\cdot\vec{\sigma}) - (\vec{h}\cdot\nabla V)(\vec{r}\cdot\vec{\sigma})]\Big]\phi_c(\vec{r})\phi^*_c(\vec{r}')\chi_s , \tag{40}$$

where $\Omega_0=\Omega/N=$ volume per ion. The matrix elements of $\mathcal{V}^{(0)}_{\kappa s}$, $\mathcal{V}^{(1)}_{\kappa s}$, $\mathcal{V}^{(2)}_{\kappa s}$, ..., are obtained from the matrix elements of $\mathcal{V}^{(0)}_{\vec{k}s}$, $\mathcal{V}^{(1)}_{\vec{k}s}$, $\mathcal{V}^{(2)}_{\vec{k}s}$, ..., by replacing the wave vector $\vec{k}$ by the operator $\underline{\kappa}$ in the exponential, i.e., symmetrically.

It can be easily shown from Eq. (37) that all $S(\vec{q},\vec{q}')$ terms vanish except those for which both $\vec{q}$ and $\vec{q}'$ lie on a reciprocal lattice. So the only nonvanishing terms are the $S(\vec{G},\vec{G}')$ terms, where $\vec{G}$ and $\vec{G}'$ are the reciprocal-lattice vectors.

V. SUMMARY AND CONCLUSION

In this paper a general magnetic-pseudopotential theory has been formulated for Bloch electrons in a magnetic field. In this method, tight-binding and OPW functions have been constructed which have the symmetry of the magnetic Bloch functions and which form a complete set for the wave function of the Hamiltonian of the crystal in a magnetic field. These have been used as basis states for the wave function of an eigenstate of the problem, and an effective Hamiltonian is obtained which is different from the effective Hamiltonians of Kohn,[3] Roth,[4] Blount,[5] and Misra, Mohanty, and Roth[6] in the sense that there is a general magnetic pseudopotential in the Hamiltonian. The expression for the magnetic pseudopotential has been obtained in a form such that it can be calculated to any order in the magnetic field. It is shown that this expression becomes particularly simple for metals.

The immediate purpose of this formulation is to use it to calculate the total magnetic susceptibility of metals, which shall be reported in a future paper. This approach would be different from traditional band calculations in the sense that perturbation theory can be used to calculate the total magnetic susceptibility of metals. However, it is hoped that this method can be applied to other problems of Bloch electrons in a magnetic field. It is also noted that there can be alternate ways of constructing the magnetic pseudopotential just as there are alternate ways of constructing the zero-field pseudopotential.

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