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Multiple-scattering theory of itinerant electron magnetism in random muffin-tin alloys

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We discuss the equilibrium electronic structure of a random binary alloy within the framework of a spin-dependent muffin-tin Hamiltonian. The disorder is treated on the basis of the single-site approximations (SSA), especially the average t -matrix and the coherent potential approximations. The local-spin-density (LSD) functional approach is employed to relate the electron and the spin densities with the atomic potentials, thus providing a fully self-consistent description of the ground-state properties of the random alloy. By using the atomic magnetic moments as expansion parameters, a Stoner-type linearized form of the full SSA-LSD formalism is developed. This analysis yields insights into the nature of the semiphenomenological isotropic Stoner parameter as it enters a proper multiple-scattering treatment of the electron states in perfect crystals and disordered metals. In particular, the exchange splitting of the atomic scattering matrices, rather than that of the energy bands, is seen to play a more fundamental role in the theory. The linearized scheme would permit a significantly simplified self-consistent determination of the possible ferromagnetic instabilities in the ground state in terms of the solutions of the corresponding nonmagnetic problem. A variety of alternative expressions for the electron and spin densities in the alloy, which are useful from the viewpoint of making contact with relevant experiments, are also presented.

I. INTRODUCTION

The magnetic properties of disordered metals have to date been discussed largely on the basis of the one- or two-band—tight-binding model Hamiltonian extended to include an intra-atomic Coulomb interaction of the Hubbard form.^{1,2} Despite the progress that has resulted from these studies, it is generally agreed that the tight-binding model Hamiltonian represents a rather simple parametrization of the electronic structure of close-packed systems in general and the transition and noble metals and their alloys in particular and that, in order to describe the electron states in these systems realistically, the muffin-tin Hamiltonian should be employed. With this motivation, a considerable effort has been devoted in recent years to delineating the properties of the muffin-tin Hamiltonian.^{3–5}

During the last decade, the understanding of the electronic spectra of the nonmagnetic muffin-tin alloys has reached a good level of maturity on the basis of the single site approximations (SSA), especially the average T matrix (ATA) and the coherent-potential (CPA) approximations.^{4,5} The treat-

ment of the ground-state properties of the nonmagnetic as well as the magnetic perfect metals has also proven quite successful within the local-spin-density (LSD) functional approach.⁶ For these reasons, the use of single-site—local-spin-density (SSA-LSD) approximations in the present attempt to develop equilibrium one-electron structure of random magnetic alloys possesses a natural basis.⁷

We emphasize that the precise form of the exchange-correlation energy functional does not enter our consideration in any significant manner. Therefore continuing progress⁸ in understanding the nature of the density functional (e.g., the inclusion of relativistic⁹ or non-local effects¹⁰) can be brought to bear on the alloy problem in a rather straightforward manner. It should be noted in this connection that the LSD approach has been extended to include finite-temperature effects.¹¹ Attempts have also been made very recently to describe the finite-temperature properties of the Hubbard Hamiltonian.^{12,13} Despite these important developments, the subject of finite-temperature magnetism in metals and alloys would appear to be an unclear¹⁴ one and we will, therefore, implicitly confine ourselves to the discussion of the zero-

temperature ground-state properties in this article.

Our assumption of complete randomness of disorder is dictated by the current state of the theory of nonmagnetic muffin-tin alloys.¹⁵ Although the chemical short-range-ordering effects are obviously important in describing some properties,¹⁶ it is unlikely that the average electronic structure of the alloy (nonmagnetic as well as magnetic) is sensitive to these effects.

An outline of this article and its principal conclusions is as follows. The introductory remarks in Sec. I are followed in Sec. II by a summary of the general SSA-LSD formalism for the random binary alloy A_xB_{1-x} . The underlying magnetic Hamiltonian is described in Sec. II A. The relevant expressions for the spin-dependent electron density $\rho_\sigma^{A(B)}(\vec{r})$ on A (B) site [or equivalently the total electron density $\rho^{A(B)}(\vec{r})$, and the net spin density $m^{A(B)}(\vec{r})$], presented in Sec. II B, are applicable to either the ATA or the CPA depending upon the choice of the effective medium. To close the full self-consistency loop for treating the ground-state properties, Sec. II C then takes up the question of constructing a new set of A (B) atom potentials $v_\sigma^{A(B)}(\vec{r})$ in terms of the quantities $\rho^{A(B)}(\vec{r})$ and $m^{A(B)}(\vec{r})$. We also derive a particularly useful equation which displays explicitly the exchange splitting of the A and B atomic potentials and the reaction matrices $k_\sigma^{A(B)}$ (related directly to the inverse of the usual $t_\sigma^{A(B)}$ matrices). We note that the formal developments of Sec. II closely parallel the corresponding treatment of the nonmagnetic alloys, since the consideration of the magnetic Hamiltonian requires the solution of two essentially independent alloy problems, one for each spin direction. The formulas of Sec. II are also presented without making the usual assumption of spherical symmetry of atomic potentials within the muffin-tin spheres.

In the context of itinerant electron magnetism, the Stoner model has played a historically important role.¹⁷ To capture the essence of this model, Sec. III presents and discusses a linearized version of the full SSA-LSD framework (outlined in Sec. II), with the atomic magnetic moment as parameter in a first-order perturbation expansion. Using the example of a perfect crystal, Sec. III A delineates the relationship between the exchange splitting of the Bloch bands and the spin-dependent potential or the scattering matrices and thus provides an insight into the nature of the Stoner parameter as it enters the multiple-scattering theory of perfect metals. Section III B extends these considerations

to random alloys. To expose the simplifications in the structure of the formalism resulting from symmetry, we specialize to cubic systems and to the use of only $l \leq 2$ phase shifts for the constituent atoms. The linearized form of the theory for the electron and spin densities and for the exchange splitting of the atomic potentials and scattering matrices is described in Secs. III B 1 and III B 2, respectively. We also discuss the expressions for the spin density $m^{A(B)}(\vec{r})$ in the A (B) cell and its anisotropy from the standpoint of analyzing the neutron diffraction measurements on metals and alloys.

Our considerations show that the exchange splitting of the scattering matrices $k_\sigma^{A(B)}$, rather than that of the energy bands, plays a fundamental role in the theory and that, this is the form in which the theory generalizes most readily to the case of a disordered alloy. The quantities which connect the exchange splitting of the matrices $k_\sigma^{A(B)}$ to the atomic magnetic moments $\mu^{A(B)}$ (and are, therefore, akin in physical significance to the Stoner parameter) are found to be energy-dependent matrices in angular momentum space. We discuss appropriate reductions of the formalism in order to clarify the nature of assumptions underlying the use of a *single* isotropic scalar Stoner parameter in the theory of metals and alloys.

The question of application of the present formalism to realistic systems is discussed in Sec. IV. Note that the full SSA-LSD theory, summarized in Sec. II, involves self-consistency in the treatment of disorder (in the sense of the CPA) as well as of the spin-dependent atomic potentials (i.e., the electron and spin densities on the A and B sites should reproduce themselves). These fully self-consistent solutions can be expected to describe possible ferromagnetic instabilities¹⁸ of the system with respect to parameters such as the magnetization of the A or B unit cells or the changes in the alloy composition. We emphasize that the question of existence of magnetic instabilities can be investigated on the basis of the linearized theory of Sec. III. This scheme permits the inclusion of magnetic effects in terms of the solution to the corresponding nonmagnetic problem and may, therefore, be viewed as an approximate procedure for extending the nonmagnetic solutions into the magnetic domain. In any event, such a solution can be expected, on physical grounds, to serve as a good starting guess to the fully self-consistent SSA-LSD machinery of Sec. II.

In conclusion, it is our contention that, given the

overall success of the ATA- and CPA-type theories in describing the one-electron equilibrium electronic structure of nonmagnetic alloys (e.g., the complex bands,¹⁹ the average densities of states^{4,5} and the momentum densities²⁰⁻²²) and the success of the LSD approach in describing the (zero-temperature) ground-state properties of perfect metals²³ (e.g., cohesive energies, lattice constants, and bulk moduli), the present SSA-LSD formalism should yield a comparably accurate description of the electronic states in random magnetic alloys.

II. GENERAL FORMALISM

A. The Hamiltonian

We discuss the substitutionally disordered binary alloy $A_x B_{1-x}$ within the framework of the spin-dependent one-electron Hamiltonian (in atomic units)

$$H = -\nabla^2 + \sum_n v_\sigma^{A(B)}(\vec{r} - \vec{R}_n). \quad (2.1)$$

Here, the crystal potential is given as a superposition of the atomic potentials v_σ^A and v_σ^B , centered on the set of N ionic sites $\{\vec{R}_n\}$. The probability that a given site is occupied by an A (or a B) atom is proportional to its concentration x (or $y = 1 - x$). The atomic potential $v_\sigma^{A(B)}$ is assumed to be of the muffin-tin form, i.e.,

$$v_\sigma^{A(B)}(\vec{r}) = \begin{cases} v_\sigma^{A(B)}(\vec{r}) & \text{for } r < R_m \\ v_\sigma^I & \text{for } r > R_m, \end{cases} \quad (2.2a)$$

$$(2.2b)$$

where the radius R_m is chosen so that the various muffin-tin spheres do not overlap. The value of the constant potential in the interstitial region v_σ^I is not a statistical variable, although it depends on the spin index σ . (The two possible values of σ will be denoted by $+$ and $-$.) Finally, we note that the formal developments in this section are carried out without the usual assumption of spherical symmetry of the atomic potentials for $r < R_m$.

B. Spin-dependent densities within the single-site approximation

As noted in the Introduction, in view of its form, properties of Hamiltonian (2.1) can be ob-

tained by considering, in effect, two *separate* disordered alloy problems (one for each spin direction), which are coupled to each other via a common Fermi energy (E_F). For this reason, the SSA formalism for the present Hamiltonian closely parallels the corresponding nonmagnetic case. The following summary of the relevant expressions for the electron densities is provided, nonetheless, for the sake of convenient reference and to stress the main ideas underlying the present formalism.

The first step in the multiscattering approach requires the one-scatterer solutions, i.e., the wave functions (regular at the origin) that satisfy the equation

$$[-\nabla^2 + v_\sigma^{A(B)}(\vec{r}) - E]Z_{\sigma,L}^{A(B)}(\vec{r}, E) = 0 \quad (2.3a)$$

within the A (B) sphere and possess the normalization

$$Z_{\sigma,L}^{A(B)}(\vec{r}, E) = \sum_{L'} Y_{L'}(\hat{r}) j_{l'}(\kappa_\sigma r) [t_\sigma^{A(B)}]_{L'L}^{-1} - i\kappa_\sigma Y_L(\hat{r}) h_l(\kappa_\sigma r) \text{ for } r > R_m. \quad (2.3b)$$

In Eq. (2.3b),

$$\kappa_\sigma^2 = E - v_\sigma^I \quad (2.3c)$$

is the kinetic energy of the electron in the interstitial region. $Y_L(\hat{r})$ [with $L \equiv (l, m)$] is the real spherical harmonic associated with the unit vector \hat{r} ; $j_l(x)$ and $h_l(x)$ are, respectively, the spherical Bessel and Hankel functions and $t_\sigma^{A(B)}(E)$ is the matrix of on-the-energy shell elements of the A (B) t matrices.²⁴ The multiscattering problem now amounts to solving the set of operator equations

$$T_\sigma^{nn'} = t_\sigma^n \delta_{nn'} + t_\sigma^n G_0 \sum_{m \neq n} T_\sigma^{mn'} \quad (2.4)$$

for the path operators $T_\sigma^{nn'}$ (Ref. 25) in terms of the one-scatterer operators t_σ^n .

As emphasized frequently in connection with the CPA and more recently the ATA, these approximation schemes physically involve the consideration of a single A or B atom embedded in an appropriately chosen effective medium (characterized by the scattering matrix t_σ^{eff}). In particular, the spin-dependent electron density associated with an A (B) atom is given by^{4,21}

$$\rho_\sigma^{A(B)}(\vec{r}) = \int_{-\infty}^{E_F} dE \rho_\sigma^{A(B)}(\vec{r}, E), \quad (2.5)$$

where

$$\rho_{\sigma}^{A(B)}(\vec{r}, E) = -\frac{1}{\pi} \text{Im} \sum_{L, L'} Z_{\sigma, L}^{A(B)}(\vec{r}, E) [T_{\sigma}^{A(B)}(E)]_{LL'} Z_{\sigma, L'}^{A(B)}(\vec{r}, E). \quad (2.6)$$

Here, $T_{\sigma}^{A(B)}(E)$ represents the matrix²⁶ (in L space) of the energy-shell elements of the scattering path operators for a single A (B) impurity placed in the effective medium, and is given by

$$T_{\sigma}^{A(B)} = \{1 - T_{\sigma}^{\text{eff}} [(t_{\sigma}^{\text{eff}})^{-1} - (t_{\sigma}^{A(B)})^{-1}]\}^{-1} T_{\sigma}^{\text{eff}} \quad (2.7)$$

in terms of the medium path operator

$$T_{\sigma}^{\text{eff}} = \frac{1}{N} \sum_{\vec{k}} [(t_{\sigma}^{\text{eff}})^{-1} - B(\vec{k}, E)]^{-1}, \quad (2.8)$$

where the summation extends over the first Brillouin zone and $B(\vec{k}, E)$ is the matrix of the usual Korringa-Kohn-Rostoker (KKR) structure functions.²⁷

In order to evaluate the Fermi energy E_F occurring on the right-hand side of (2.5), it is useful to consider the energy dependent A (B) component density of states

$$\rho_{\sigma}^{A(B)}(E) = \int_{\Omega} d\vec{r} \rho_{\sigma}^{A(B)}(\vec{r}, E) \quad (2.9)$$

obtained by integrating (2.6) over the Wigner-Seitz cell volume Ω . A further integration over E yields the corresponding integrated density:

$$N_{\sigma}^{A(B)}(E) = \int_{-\infty}^E dE' \rho_{\sigma}^{A(B)}(E'). \quad (2.10)$$

The total (i.e., including both spins) integrated density of states function can now be written as

$$N(E) = N_+(E) + N_-(E), \quad (2.11)$$

where N_{σ} denotes the average of N_{σ}^A and N_{σ}^B . E_F is then determined by the obvious requirement

$$N(E_F) - N(E_0) = xz_A + yz_B, \quad (2.12)$$

where E_0 is the energy of the valence-band edge and $z_{A(B)}$ is the number of A (B) valence electrons.

Finally, the magnetic moment associated with an A (B) site in the alloy is given by

$$\mu^{A(B)} = N_+^{A(B)}(E_F) - N_-^{A(B)}(E_F), \quad (2.13)$$

and the average magnetic moment per site is

$$\mu = x\mu^A + y\mu^B \quad (2.14a)$$

$$= N_+(E_F) - N_-(E_F). \quad (2.14b)$$

Equations (2.5)–(2.14) are applicable for any choice of the effective medium. In particular, the

ATA (Ref. 28) is specified by the choice $t_{\sigma}^{\text{eff}} = t_{\sigma}^{\text{ATA}}$, with

$$t_{\sigma}^{\text{ATA}} = xt_{\sigma}^A + yt_{\sigma}^B, \quad (2.15)$$

whereas in the CPA $t_{\sigma}^{\text{eff}} = t_{\sigma}^{\text{CPA}}$ is given by the self-consistency requirement

$$(t_{\sigma}^{\text{CPA}})^{-1} = x(t_{\sigma}^A)^{-1} + y(t_{\sigma}^B)^{-1} + [(t_{\sigma}^{\text{CPA}})^{-1} - (t_{\sigma}^A)^{-1}] \times T_{\sigma}^{\text{CPA}} [(t_{\sigma}^{\text{CPA}})^{-1} - (t_{\sigma}^B)^{-1}], \quad (2.16)$$

where T_{σ}^{CPA} is defined by Eq. (2.8) with the replacement $t_{\sigma}^{\text{eff}} \rightarrow t_{\sigma}^{\text{CPA}}$.

Starting with a set of A and B atom potentials v_{σ}^A and v_{σ}^B , the equations presented in this section would allow the computation, within either the ATA or the CPA, of the various one-electron properties of the random alloy, including the electron densities which are of particular interest to us.²⁹

C. Fully self-consistent atomic potentials within the local spin density functional approximation

Given the total electron density

$$\rho^{A(B)}(\vec{r}) = \rho_+^{A(B)}(\vec{r}) + \rho_-^{A(B)}(\vec{r}) \quad (2.17)$$

and the net spin density

$$m^{A(B)}(\vec{r}) = \rho_+^{A(B)}(\vec{r}) - \rho_-^{A(B)}(\vec{r}) \quad (2.18)$$

in the A (B) Wigner-Seitz cell (obtained for a set of starting A and B atom potentials in Sec. II B), we now discuss how a new spin-dependent potential in the A (B) cell can be constructed using the LSD framework. To this end, we decompose the atomic potentials into the electrostatic (Coulomb) and the exchange-correlation parts, i.e.,

$$v_{\sigma}^{A(B)}(\vec{r}) = v_c^{A(B)}(\vec{r}) + v_{\sigma, xc}^{A(B)}(\vec{r}). \quad (2.19)$$

In the spirit of the SSA, the Coulomb contribution $v_c^{A(B)}(\vec{r})$ is obtained by solving the electrostatic problem for an A (B) atom placed in the average medium, i.e.,

$$\begin{aligned}
v_c^{A(B)}(\vec{r}) = & -\frac{2Z^{A(B)}}{r} + \int_{\Omega} d\vec{r}' \frac{\rho^{A(B)}(\vec{r}')}{|\vec{r}-\vec{r}'|} \\
& + \sum_{m \neq 0} \int_{\Omega} \frac{d\vec{r}'}{|\vec{r}-\vec{r}'-\vec{R}_m|} \{x[\rho^A(\vec{r}'-\vec{R}_m)-Z^A\delta(\vec{r}'-\vec{R}_m)] + y[\rho^B(\vec{r}'-\vec{R}_m)-Z^B\delta(\vec{r}'-\vec{R}_m)]\}
\end{aligned} \quad (2.20)$$

where the integrals extend over the volume Ω of the central unit cell and the intercellular contributions [i.e., the second integral on the right-hand side of (2.20)] have been randomly averaged. Note that the overall charge neutrality of the system implies that, on the average, any Wigner-Seitz cell will be electrically neutral.

The exchange-correlation part $v_{\sigma,xc}^{A(B)}(\vec{r})$ is given in the LSD formalism by^{30,31}

$$v_{\sigma,xc}^{A(B)}(\vec{r}) = \frac{\partial}{\partial \rho_{\sigma}^{A(B)}} [\rho^{A(B)}(\vec{r}) \epsilon_{xc}(\rho_+^{A(B)}(\vec{r}), \rho_-^{A(B)}(\vec{r}))], \quad (2.21)$$

in terms of the exchange-correlation energy per electron ϵ_{xc} . As emphasized in the Introduction, the precise form of the functional ϵ_{xc} is not important for the present considerations and will therefore not be specified.

It is useful to display the spin dependence of the atomic potentials explicitly. This can be achieved simply by replacing the variable ρ_+ and ρ_- in (2.21) by the total electron and spin densities defined by Eqs. (2.17) and (2.18). Thus, we write

$$v_{\sigma,xc}^{A(B)}(\vec{r}) = v_{xc}^{A(B)}(\vec{r}) + \sigma v_{sxc}^{A(B)}(\vec{r}), \quad (2.22)$$

where

$$v_{sxc}^{A(B)}(\vec{r}) = \frac{1}{2} [v_{+,xc}^{A(B)}(\vec{r}) - v_{-,xc}^{A(B)}(\vec{r})] \quad (2.23)$$

and v_{xc} is defined by a similar equation involving a sum (rather than a difference) of $v_{+,xc}$ and $v_{-,xc}$.

The potential in the interstitial flat may also be written in analogy with (2.22) as³²

$$v_{\sigma}^I = v^I + \sigma v_{sxc}^I. \quad (2.24)$$

Note that the quantities that enter the multiple scattering theory naturally are not the atomic potentials discussed so far, but the t matrices defined previously in Eqs. (2.3a)–(2.3c), or equivalently the so-called reaction matrices $k_{\sigma}^{A(B)}$:

$$(k_{\sigma}^{A(B)})^{-1} = (t_{\sigma}^{A(B)})^{-1} - i\kappa_{\sigma} I, \quad (2.25)$$

where I denotes the unit matrix and κ_{σ} was defined

above in (2.3c). We conclude this section by deriving an expression for $k_{\sigma}^{A(B)}$, which displays its σ dependence in a particularly pronounced form.

For this purpose, we define the functions $R_{\sigma,L}^{A(B)}(\vec{r}, E)$, related to the solutions $Z_{\sigma,L}^{A(B)}(\vec{r}, E)$ of the wave equation [see Eqs. (2.3)] by

$$R_{\sigma,L}^{A(B)}(\vec{r}, E) = \sum_{L'} Z_{\sigma,L'}^{A(B)}(\vec{r}, E) [k_{\sigma}^{A(B)}]_{L'L}. \quad (2.26)$$

In view of Eqs. (2.3b) and (2.26), $R_{\sigma,L}^{A(B)}(\vec{r}, E)$ possess the asymptotic form

$$\begin{aligned}
R_{\sigma,L}^{A(B)}(\vec{r}, E) = & Y_L(\hat{r}) j_l(\kappa_{\sigma} r) \\
& + \kappa_{\sigma} \sum_{L'} Y_{L'}(\hat{r}) n_{l'}(\kappa_{\sigma} r) [k_{\sigma}^{A(B)}]_{L'L}
\end{aligned} \quad \text{for } r > R_m. \quad (2.27)$$

The corresponding σ -independent solutions $R_L^{A(B)}(\vec{r}, E)$ are obtained by setting $\sigma=0$ in the various relevant equations and will possess the normalization

$$\begin{aligned}
R_L^{A(B)}(\vec{r}, E) = & Y_L(\hat{r}) j_l(\kappa r) \\
& + \kappa \sum_{L'} Y_{L'}(\hat{r}) n_{l'}(\kappa r) [k^{A(B)}]_{L'L}
\end{aligned} \quad \text{for } r > R_m, \quad (2.28)$$

where $k^{A(B)}$ is the k matrix for the σ independent case and in view of Eqs. (2.3c) and (2.27)

$$\kappa^2 = E - v^I. \quad (2.29)$$

Comparing (2.27) and (2.28), it is clear that $R_L^{A(B)}(\vec{r}, E - \sigma v_{sxc}^I)$ satisfies the differential equation

$$\begin{aligned}
[-\nabla^2 + v_c^{A(B)}(\vec{r}) + v_{xc}^{A(B)}(\vec{r}) + \sigma v_{sxc}^I - E] \\
\times R_L^{A(B)}(\vec{r}, E - \sigma v_{sxc}^I) = 0 \quad \text{for } r < R_m. \quad (2.30)
\end{aligned}$$

We now subtract (2.30) multiplied by $R_{\sigma,L}^{A(B)}$ from (2.3a) multiplied by $R_L^{A(B)}(\vec{r}, E - \sigma v_{sxc}^I)$. The integration of the resulting expression over the muffin-tin sphere volume Ω_{MT} with the use of Green's theorem and a few obvious manipulations

yields the desired relationship between the spin-polarized and the nonpolarized k matrices

$$k_{\sigma}^{A(B)}(E) = k^{A(B)}(E) + \sigma S^{A(B)}(E), \quad (2.31a)$$

where

$$\begin{aligned} S_{L'L}^{A(B)}(E) = & \int_{\Omega_{MT}} d\vec{r} R_L^{A(B)}(\vec{r}, E - \sigma \epsilon_{sxc}^I) \\ & \times [\sigma \epsilon_{sxc}^{A(B)}(\vec{r}) - \sigma \epsilon_{sxc}^I] \\ & \times R_{\sigma, L'}^{A(B)}(\vec{r}, E). \end{aligned} \quad (2.31b)$$

Equations (2.31) will be seen in the following section to play an important role in the formulation of a Stoner-type model of itinerant magnetism.

III. STONER-TYPE MODEL OF ITINERANT MAGNETISM IN THE MULTIPLE SCATTERING FORMULATION

A. Interpretation of Stoner parameter in a perfect crystal

The exact equations (2.31) can be used to derive an explicit expression for the exchange splitting of

$$G_{\sigma, \vec{k}}(\vec{r}, \vec{r}'; E) = \sum_{L, L'} R_{\sigma, L}(\vec{r}, E) [A^{-1}(\vec{k}, E) - k_{\sigma}]^{-1} R_{\sigma, L'}(\vec{r}', E) + \kappa_{\sigma} \sum_L R_{\sigma, L}(\vec{r}, E) N_{\sigma, L}(\vec{r}', E), \quad (3.2)$$

where the regular solutions $R_{\sigma, L}$ were introduced above in (2.26) and $N_{\sigma, L}$ are the related irregular solutions possessing the asymptotic form

$$N_{\sigma, L}(\vec{r}, E) = n_l(\kappa_{\sigma} r) Y_L(\hat{r}) \quad \text{for } r > R_m, \quad (3.3)$$

and the real matrix $A(\vec{k}, E)$ is related to the KKR structure functions $B(\vec{k}, E)$ by

$$A(\vec{k}, E) = B(\vec{k}, E) - i\kappa_{\sigma} I. \quad (3.4)$$

In view of Eqs. (3.1), (3.2), and (2.31), the magnetic band structure is given by the determinantal equation:

$$||A^{-1}(\vec{k}, E) - k - \sigma S|| = 0. \quad (3.5)$$

In the vicinity of the Bloch level $\epsilon_n(\vec{k})$, the matrix in Eq. (3.5) can be approximated as (setting $\sigma=0$)

$$A^{-1}(\vec{k}, E) - k \cong \sum_n [E - \epsilon_n(\vec{k})] \lambda_n(\vec{k}) P_n(\vec{k}), \quad (3.6)$$

where $P_n(\vec{k})$ is the projector (in angular momentum space) for the (n, \vec{k}) state and the weight factor $\lambda_n(\vec{k})$ can be found from the condition

the energy bands in a perfect crystal. Such an analysis yields insights into the nature of the Stoner parameter as it enters the multiple scattering theory.

The one-electron Green's function $G_{\sigma, \vec{k}}(\vec{r}, \vec{r}'; E)$ in a perfect metal can be expressed in terms of the Bloch functions $\psi_{\sigma, \vec{k}}^{(n)}(\vec{r})$ and the corresponding energies $\epsilon_{\sigma, n}(\vec{k})$ as

$$\begin{aligned} G_{\sigma, \vec{k}}(\vec{r}, \vec{r}'; E) = & \sum_n \psi_{\sigma, \vec{k}}^{(n)}(\vec{r}) [E - \epsilon_{\sigma, n}(\vec{k})]^{-1} \\ & \times \psi_{\sigma, \vec{k}}^{(n)}(\vec{r}'), \end{aligned} \quad (3.1)$$

where \vec{k} and n denote the reduced wave vector and the band index, respectively. The functions $\psi_{\sigma, \vec{k}}^{(n)}$ will be assumed to be normalized to unity within the Wigner-Seitz cell.

Alternatively, $G_{\sigma, \vec{k}}$ can be expanded in the unit cell (for $r < r'$) using solutions of the Schrödinger equation in the muffin-tin sphere as

$$\text{Lt}_{E \rightarrow \epsilon_n(\vec{k})} \int_{\Omega} d\vec{r} [E - \epsilon_n(\vec{k})] G_{\vec{k}}(\vec{r}, \vec{r}) = 1, \quad (3.7)$$

which represents the normalization of $\psi_{\vec{k}}^{(n)}$ over the cell volume [see Eq. (3.1)]. (The index σ has been dropped in this discussion, since we are considering the nonmagnetic case.) The use of (3.2), and (3.6) in (3.7) yields in a straightforward manner

$$\lambda_n(\vec{k}) = \text{Tr}[P_n(\vec{k}) U(\epsilon_n(\vec{k}))], \quad (3.8)$$

where the matrix $U(E)$ is defined by

$$U_{LL'}(E) = \int_{\Omega} d\vec{r} R_L(\vec{r}, E) R_{L'}(\vec{r}, E). \quad (3.9)$$

By treating the σS term in (3.5) as a perturbation (in first order), the spin-polarized band structure is easily shown to be obtainable from the equation

$$\epsilon_{\sigma, n}(\vec{k}) \cong \epsilon_n(\vec{k}) + \sigma \frac{\text{Tr}[S(\epsilon_n(\vec{k})) P_n(\vec{k})]}{\text{Tr}[U(\epsilon_n(\vec{k})) P_n(\vec{k})]}. \quad (3.10)$$

We now define the generalized Stoner parameter via

$$\epsilon_{+, n}(\vec{k}) - \epsilon_{-, n}(\vec{k}) = -\mu I_n(\vec{k}), \quad (3.11)$$

where μ denotes the magnetic moment of the unit cell. A comparison of (3.10) and (3.11) allows the identification

$$I_n(\vec{k}) = -\frac{2}{\mu} \frac{\text{Tr}[S(\epsilon_n(\vec{k}))P_n(\vec{k})]}{\text{Tr}[U(\epsilon_n(\vec{k}))P_n(\vec{k})]}, \quad (3.12)$$

which shows that this quantity depends on \vec{k} and also the band index n . A scalar Stoner parameter can be retrieved if we assume the matrices S and U to take the special form

$$S(E) = \mu \bar{S}(E) I, \quad (3.13a)$$

and

$$U(E) = \bar{U}(E) I. \quad (3.13b)$$

Using (3.13a) and (3.13b) in (3.12) gives

$$\bar{I}(E) = -2 \frac{\bar{S}(E)}{\bar{U}(E)}. \quad (3.14)$$

Equations (3.14) and (3.12) provide the connection between the Stoner parameter (and its proper generalization) and the nonmagnetic atomic potentials. In particular, $\bar{I}(E)$ in Eq. (3.14) plays the role of the energy-dependent Stoner parameter discussed in Refs. 33 and 17.

B. Random binary alloy

The linearization procedure (using the magnetic moment as an expansion parameter), which forms

$$\rho_{\sigma}^{A(B)}(\vec{r}, E) = -\frac{1}{\pi} \text{Im} \sum_{l,t} [Z_{\sigma,l}^{A(B)}(r, E)]^2 [T_{\sigma}^{A(B)}(E)]_{l,t} F_l(\hat{r}) N_t, \quad (3.16)$$

where

$$F_l(\hat{r}) = \frac{1}{N_t} \sum_{\mu} K_{l,t\mu}(\hat{r}) K_{l,t\mu}(\hat{r}). \quad (3.17)$$

The physical density $\rho_{\sigma}^{A(B)}(\hat{r})$ [or $\rho_{\sigma}^{A(B)}(E)$] is then obtained by integrating (3.16) as in Eq. (2.5).

We emphasize that due to the nonspherical shape of the Wigner-Seitz cell, l decomposition of the various densities is well defined only within the muffin-tin sphere rather than the entire unit cell. For this reason, it is convenient to introduce several auxiliary quantities. The contribution of (l, t) symmetry to the density of states from the A (B) muffin-tin sphere is

$$\rho_{\sigma,l}^{A(B)}(E) = -\frac{1}{\pi} \text{Im} \int_{\Omega_{\text{MT}}} d\vec{r} [Z_{\sigma,l}^{A(B)}(r, E)]^2 [T_{\sigma}^{A(B)}(E)]_{l,t} F_l(\hat{r}) N_t, \quad (3.18)$$

and the corresponding integrated A (B) density of states is

$$N_{\sigma,l}^{A(B)}(E) = \int_{-\infty}^E dE \rho_{\sigma,l}^{A(B)}(E). \quad (3.19)$$

the basis of a Stoner-type model, is developed in this section. The maximum use of symmetry is made in considering the various physical quantities. Many of the formulas presented below are particularly useful from the practical standpoint of application of the theory to the analysis of relevant experiments.

1. Charge and spin densities

To expose the characteristic simplification resulting from the use of symmetry, we rewrite first the previously presented formulas for charge and spin densities (see Sec. II B) for the special case of a cubic system in which the pure A and B phase shifts for only $l \leq 2$ are employed. In this case, the scattering matrices (i.e., t_{σ}^{eff} or k_{σ}^{eff}) can be shown to be diagonal. The extension to other crystal structures or the inclusion of higher l values is relatively straightforward.

For cubic symmetry, the natural basis functions for expanding the angular dependence of various quantities are the cubic rather than the spherical harmonics. Thus, we write the solutions of the wave equation (2.3a) in the form

$$Z_{\sigma,l\mu}^{A(B)}(\vec{r}, E) = Z_{\sigma,l}^{A(B)}(r, E) K_{l,t\mu}(\hat{r}), \quad (3.15)$$

where $K_{l,t\mu}(\hat{r})$ is the real cubic harmonic of angular momentum l belonging to the t th irreducible representation (dimension N_t) of the cubic group and μ denotes a row index. In view of (3.15), expression (2.6) takes the form

We also define radial wave functions $P_{\sigma,l}^{A(B)}(r, E)$ related to $Z_{\sigma,l}^{A(B)}(r, E)$, but normalized to unity in the muffin-tin sphere volume, i.e.,

$$P_{\sigma,lt}^{A(B)}(r,E) = \frac{Z_{\sigma,lt}^{A(B)}(r,E)}{\left[\int_0^{R_m} dr r^2 [Z_{\sigma,lt}^{A(B)}(r,E)]^2 \right]^{1/2}}. \quad (3.20)$$

To obtain the desired expression for the total electron density $\rho^{A(B)}(\vec{r})$ [given by summing (3.16) over σ , as in (2.17)], we introduce the radial weight functions

$$W_{c,lt}^{A(B)}(r) = \sum_{\sigma} \int_{-\infty}^{E_F} dE [P_{\sigma,lt}^{A(B)}(r,E)]^2 \times \frac{\rho_{\sigma,lt}^{A(B)}(E)}{N_{lt}^{A(B)}(E_F)}, \quad (3.21a)$$

where

$$N_{lt}^{A(B)}(E_F) = \sum_{\sigma} N_{\sigma,lt}^{A(B)}(E_F) \quad (3.21b)$$

is the total number of electrons of (l,t) type in the $A(B)$ muffin-tin sphere. The electron density $\rho^{A(B)}(\vec{r})$ is now given in terms of $W_c^{A(B)}$ and $N_{lt}^{A(B)}(E_F)$ by

$$\rho^{A(B)}(\vec{r}) = \sum_{l,t} W_{c,lt}^{A(B)}(r) N_{lt}^{A(B)}(E_F) F_{lt}(\hat{r}). \quad (3.22)$$

The validity of (3.22) is easily checked by substituting (3.21a) and (3.20) in (3.16) and summing over σ . Equation (3.22) gives $\rho^{A(B)}(\vec{r})$ in a particularly transparent form in which the angular and radial dependences are completely separated out.

The manipulations which lead to form (3.22) for the electron density are easily repeated to derive a similar formula for the net spin density $m^{A(B)}(\vec{r})$ defined by (2.18). The final result is

$$m^{A(B)}(\vec{r}) = \sum_{l,t} W_{m,lt}^{A(B)}(r) \mu_{lt}^{A(B)} F_{lt}(\hat{r}), \quad (3.23)$$

where the weights $W_{m,lt}^{A(B)}(r)$ are defined in analogy with (3.21a) by

$$W_{m,lt}^{A(B)}(r) = \sum_{\sigma} \sigma \int_{-\infty}^{E_F} dE [P_{\sigma,lt}^{A(B)}(r,E)]^2 \times \frac{\rho_{\sigma,lt}^{A(B)}(E)}{\mu_{lt}^{A(B)}}, \quad (3.24a)$$

and $\mu_{lt}^{A(B)}$ is the partial contribution of (l,t) symmetry to the magnetic moment on the $A(B)$ site from the muffin-tin sphere, i.e.,

$$\mu_{lt}^{A(B)} = \sum_{\sigma} \sigma N_{\sigma,lt}^{A(B)}(E_F). \quad (3.24b)$$

Note that, since the spin density $m^{A(B)}(\vec{r})$ involves the difference of up and down electron densities [cf. Eq. (2.18)], the contributions to $m^{A(B)}(\vec{r})$ arise primarily from electrons in the vicinity of the Fermi energy. Therefore, the weight factor $W_{m,lt}^{A(B)}$ [Eq. (3.24a)] may reasonably be approximated by

$$W_{m,lt}^{A(B)}(r) \cong \sum_{\sigma} \sigma [P_{\sigma,lt}^{A(B)}(r,E_F)]^2 \times \frac{N_{\sigma,lt}^{A(B)}(E_F)}{\mu_{lt}^{A(B)}}. \quad (3.25)$$

If the spin-polarization of the functions $P_{\sigma,lt}^{A(B)}(r,E_F)$ is further neglected, we obtain

$$W_{m,lt}^{A(B)}(r) \cong [P_{lt}^{A(B)}(r,E_F)]^2. \quad (3.26)$$

The use of (3.26) in (3.23) yields

$$m^{A(B)}(\vec{r}) = \mu^{A(B)} \bar{m}^{A(B)}(\vec{r}) \quad (3.27a)$$

$$= \mu^{A(B)} \sum_{l,t} [P_{lt}^{A(B)}(r,E_F)]^2 \times \gamma_{lt}^{A(B)} F_{lt}(\hat{r}), \quad (3.27b)$$

where $\mu^{A(B)}$ is the total magnetic moment of the $A(B)$ muffin-tin sphere. The quantity $\bar{m}^{A(B)}(\vec{r})$ is defined by (3.27a) and $\gamma_{lt}^{A(B)}$ by the equation

$$\gamma_{lt}^{A(B)} = \mu_{lt}^{A(B)} / \mu^{A(B)}. \quad (3.27c)$$

The usefulness of form (3.27b) results from the fact that this equation permits the computation of the spin-density $m^{A(B)}(\vec{r})$ in terms of the magnetic moments $\mu_{lt}^{A(B)}$.

Finally, we consider the special case where only the d electrons are assumed to contribute to magnetism. It is interesting to note that in this case, the coefficient $\gamma_{l,(12)}^{A(B)}$ [(12) denotes e_g symmetry] can be identified as the so-called "asphericity parameter",³⁴

$$\gamma_{2,(12)}^{A(B)} = \gamma_{2,(12)}^{A(B)}, \quad (3.28)$$

which is used extensively in the analysis of the magnetic form factors determined from neutron diffraction measurements. Equations (3.23)–(3.27) thus provide a proper theoretical basis for defining the "asphericity parameters" more generally.

If only d electrons are included, an explicit form for the normalized spin density $\bar{m}^{A(B)}(\vec{r})$ [Eq. (3.27a)] can be obtained by expressing the function $F_{lt}(\hat{r})$ [see Eq. (3.17)] in terms of the invariant cubic harmonics. Assuming spherically symmetric charge density³⁵ (in the muffin-tin sphere) it is straightforward to show that

$$\bar{m}^{A(B)}(\vec{r}) = [P_2^{A(B)}(r, E_F)]^2 \frac{1}{\sqrt{4\pi}} \left[K_{0,1}(\hat{r}) + \frac{2}{\sqrt{21}} \left(\frac{5}{2} \gamma^{A(B)} - 1 \right) K_{4,1}(\hat{r}) \right]. \quad (3.29)$$

An expression similar to (3.29) has been used previously on a semiphenomenological basis in Refs. 36–38 in connection with the analysis of neutron diffraction experiments on metals and alloys.

2. Atomic potentials and scattering matrices

In order to develop the necessary linearization scheme, we express the spin-dependent part of the potential (2.22) to the lowest order in $m^{A(B)}(\vec{r})$ as

$$v_{\text{sxc}}^{A(B)}(\vec{r}) \cong v_s^{A(B)}(\rho^{A(B)}(\vec{r})) m^{A(B)}(\vec{r}), \quad (3.30)$$

where, in view of (2.23) and (2.21),

$$v_s^{A(B)}(\rho^{A(B)}(\vec{r})) = \frac{\partial^2}{(\partial m^{A(B)})^2} [\rho^{A(B)}(\vec{r}) \epsilon_{\text{xc}}(\rho^{A(B)}, m^{A(B)})]_{m^{A(B)}(\vec{r})=0}. \quad (3.31)$$

The spin dependence of the scattering matrix $k_\sigma^{A(B)}$ [i.e., the matrix $S^{A(B)}$ in Eq. (2.31a)] is now obtained by substituting (3.30) into (2.31b), which yields

$$S^{A(B)} = S_m^{A(B)} + S_I^{A(B)}, \quad (3.32)$$

where the contributions $S_m^{A(B)}$ and $S_I^{A(B)}$ from the muffin-tin sphere and the interstitial region are given, respectively, by the equations

$$[S_m^{A(B)}(E)]_{LL'} = \int_{\Omega_{\text{MT}}} d\vec{r} R_L^{A(B)}(\vec{r}, E) v_s^{A(B)}(\rho(\vec{r})) m^{A(B)}(\vec{r}) R_{L'}^{A(B)}(\vec{r}, E) \quad (3.33a)$$

and

$$[S_I^{A(B)}(E)]_{LL'} = -v_{\text{sxc}}^I \int_{\Omega_{\text{MT}}} d\vec{r} R_L^{A(B)}(\vec{r}, E) R_{L'}^{A(B)}(\vec{r}, E). \quad (3.33b)$$

Since we are keeping terms to only the lowest order in magnetization, the spin dependence of the wave functions [occurring in (2.31b)] has been neglected in writing Eq. (3.33).

To clarify the simplifications resulting from the use of symmetry, we specialize, once again, to the case of cubic systems with spherically symmetric electron densities, and write the wave functions $R_L^{A(B)}$ in terms of the cubic harmonics as in Eq. (3.15), i.e.,

$$R_{l,t\mu}^{A(B)}(\vec{r}, E) = R_l^{A(B)}(r, E) K_{l,t\mu}(\hat{r}). \quad (3.34)$$

Using (3.34) and (3.23) in (3.33a) together with the approximation (3.26) for $W_{m,lt}^{A(B)}(r)$, the part $S_m^{A(B)}$ takes the (diagonal) form

$$[S_m^{A(B)}(E)]_{ll'} = \mu^{A(B)} \sum_{l'''} C_{ll'''}^{l'''} [I^{A(B)}(E)]_{ll'''}^{l'''} \gamma_{l'''}^{A(B)}, \quad (3.35)$$

where

$$[I^{A(B)}(E)]_{ll'''}^{l'''} = \int_0^{R_m} dr r^2 [R_l^{A(B)}(r, E)]^2 v_s^{A(B)}(\rho(r)) [P_{l'''}^{A(B)}(r, E_F)]^2 \quad (3.36)$$

and

$$C_{ll'''}^{l'''} = \int_{(4\pi)} d\Omega F_{ll'''}(\hat{r}) F_{l'''}(\hat{r}). \quad (3.37)$$

The quantities $F_{ll'''}(\hat{r})$, $\gamma_{l'''}^{A(B)}$, and $\mu^{A(B)}$ were defined in the preceding section. The coefficients $C_{ll'''}^{l'''}$ are easily calculated³⁹ and are listed in Table I for $l, l' \leq 2$.

If the s and p contributions to the total magnetic

moment are neglected [i.e., $\gamma_0 = \gamma_1 = 0$ in (3.35)],⁴⁰ expression (3.35) reduces to the form

$$[S_m^{A(B)}]_{ll} \cong \mu^{A(B)} \bar{S}_l^{A(B)}(E) C_{ll}^{A(B)}, \quad (3.38)$$

where

$$\bar{S}_l^{A(B)}(E) = \frac{1}{4\pi} \int_0^{R_m} dr r^2 [R_l^{A(B)}(r, E)]^2 v_s^{A(B)}(\rho(r)) [P_2^{A(B)}(r, E_F)]^2, \quad (3.39)$$

and the coefficients $C_{lt}^{A(B)}$ are defined by

$$C_{lt}^{A(B)} = \begin{cases} 1 & \text{for } l=0, 1 \\ 1 - \frac{2}{21}(5\gamma^{A(B)} - 2) & \text{for } t_{2g} (\Gamma_{25'}) \\ 1 + \frac{1}{7}(5\gamma^{A(B)} - 2) & \text{for } e_g (\Gamma_{12}) \end{cases} \quad (3.40a)$$

$$(3.40b)$$

$$(3.40c)$$

Note that in the special case of spherically symmetric spin density (i.e., $\gamma^A = \gamma^B = 0.4$)

$$C_{lt}^{A(B)} = 1, \quad (3.41)$$

and, therefore,

$$[S_m^{A(B)}]_{lt} = \mu^{A(B)} \bar{S}_l^{A(B)}(E). \quad (3.42)$$

The comparison of (3.42) and the corresponding perfect-crystal equation (3.14) makes it clear that, excepting normalization, the quantity $\bar{S}_l^{A(B)}$ in (3.42) possesses the physical significance of the Stoner parameter. In fact, expression (3.39) for $l=2$ is essentially equivalent to that presented in Ref. 33 (see also Ref. 17) for the Stoner parameter.⁴¹ Our derivation of (3.42) therefore exposes the nature of the assumptions inherent in the use of a single isotropic Stoner parameter in the discussion of itinerant magnetism.

Equation (3.35) shows that the exchange splitting of the atomic scattering matrices $k_{\sigma}^{A(B)}$ (via the matrix $[I^{A(B)}(E)]_{lt}^{l't'}$) rather than that of the energy bands plays a more fundamental role in the present formalism. This then is the form in which the spirit of the Stoner-type phenomenological model is captured most naturally in the multiple scattering theory of metals and alloys.

IV. APPLICATION TO TRANSITION- AND NOBLE-METAL ALLOYS

The application of the fully self-consistent single-site—local-spin-density formalism of Sec. II to even a *nonmagnetic* *d*-band alloy is quite difficult. The possible existence of magnetism involves an additional level of complexity in the present case. For these reasons, it is important to explore schemes of implementing this framework from the viewpoint of practical applications. Aspects of this question are considered below.

One interesting question concerns the existence

of stable magnetic solutions for a given composition of the alloy. The linearization procedure of Sec. III is well suited to study such a magnetic instability. In this case, the iterations can be started with an initial guess for the partial magnetic moments $[\mu_{lt}^A]^{in}$, $[\mu_{lt}^B]^{in}$, and the wave functions (and their appropriate integrals) for the nonmagnetic problem (i.e., for the case where the exchange splitting of the k matrices is set equal to zero). Equation (3.35) then yields the spin-dependent input scattering matrices $[k_{\sigma}^A]^{in}$ and $[k_{\sigma}^B]^{in}$ [the parameters $I_{lt}^{l't'}(E)$, in view of (3.36), are obtainable in terms of the nonmagnetic wave functions]. The random-alloy problem (for each of the two spin directions) is now solved using the single-site framework (ATA or CPA) outlined in Sec. II B. More specifically, we require the quantities k_{σ}^{eff} and $T_{\sigma}^{A(B)}(E)$ together with the Fermi energy, E_F ,²⁹ in terms of which the partial densities $\rho_{\sigma,lt}^{A(B)}(E)$ [see Eq. (3.18); note that, by neglecting the spin dependence of the wave functions, the \bar{r} -space integral in this equation can be calculated purely in terms of the nonmagnetic computations and need not be repeated] and the integrated densities $N_{\sigma,lt}^{A(B)}(E)$ can be obtained. Finally, the use of $N_{\sigma,lt}^{A(B)}(E_F)$ in expression (3.24b) yields a new set of values $[\mu_{lt}^A]^{out}$ and $[\mu_{lt}^B]^{out}$ for the partial magnetic

TABLE I. Values of coefficients $C_{lt}^{l't'}$ defined in (3.37) (all entries should be divided by 4π).

l	t	0 Γ_1	1 Γ_{15}	2 $\Gamma_{25'}$	2 Γ_{12}
0	Γ_1	1	1	1	1
1	Γ_{15}	1	1	1	1
2	$\Gamma_{25'}$	1	1	$\frac{25}{21}$	$\frac{5}{7}$
2	Γ_{12}	1	1	$\frac{5}{7}$	$\frac{10}{7}$

moments. The entire process is to be repeated until self-consistency is achieved. The nonvanishing of the converged values of $[\mu_h^A]$ or $[\mu_h^B]$ implies the existence of magnetic solutions.

Note that the procedure outlined in the preceding paragraph does not involve an *explicit* knowledge of the \bar{r} dependence of the electron and spin densities during the iterations. In this sense, this procedure amounts to carrying through a self-consistent treatment of only the magnetic part (and possibly also the disorder, if CPA is employed) of the problem and represents a considerable computational simplification. We emphasize, however, that the various restrictions discussed in Sec. III above can be relaxed selectively and the extent to which a given solution is fully self-consistent within the SSA-LSD framework can be ascertained. In any

event, the electron, the spin, and the other physical densities, together with the related one-electron properties (e.g., the spin-polarized band structures and the neutron form factors) can be obtained for comparison with experiments at any stage of the calculations.

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$$[t_{\sigma}^{A(B)}]_l = -\kappa_{\sigma}^{-1} \sin[\delta_{\sigma,l}^{A(B)}(E)] \\ \times \exp[i\delta_{\sigma,l}^{A(B)}(E)].$$

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²⁹The \vec{r} and E integrations [see Eqs. (2.9) and (2.10)] can in many cases be carried through formally, at least in an approximate manner, by using Lloyd-type formulas for the density of states. For example, the spin-dependent density of state $\rho_{\sigma}(E)$ can be written rigorously for the CPA in the form of a perfect derivative and, as a result, permits a considerable simplification in the computation of the integrated density of states $N_{\sigma}(E)$ and the Fermi energy. (See Refs. 3–5 for details.)

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⁴⁰We emphasize that the use of (3.38) as input will, however, lead in general to a nonzero s and p contribution to the spatial spin-density distribution.

⁴¹See, in particular, Eq. (2.15) of Ref. 33. The integration in Eq. (2.15) of this reference extends to the Wigner-Seitz radius R_{WS} , and not to the muffin-tin radius R_m as in our Eq. (3.39). This difference is related to the fact that Ref. 33 invokes the atomic-sphere approximation to treat the interstitial region whereas we do not. We, of course, obtain an additional contribution to the exchange splitting of the energy bands via $S_f^{A(B)}(E)$ [Eq. (3.33b)].