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Theoretical description of the high-field susceptibility of magnetically ordered transition metal systems with applications to Fe, Co, Ni, and $\mathrm{Fe}_{1-x}\mathrm{Co}_x$

S. Mankovsky and H. Ebert
Department Chemie und Biochemie, Universität München, Butenandtstr. 5-13, D-81377 München, Germany

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A theoretical description of the high-field susceptibility of magnetically ordered transition metal systems is presented that is based on linear response theory formulated using the Green's function technique. The approach allows to treat all spin and orbital contributions in a consistent way on the same footing. In contrast to previous work, the Landau susceptibility is included and spin-orbit induced contributions to the susceptibility are accessible by using a fully relativistic formulation. As an application of our approach, results obtained for the ferromagnetic transition metals Fe, Co, and Ni, as well as for the alloy system $\mathrm{Fe}_{1-x}\mathrm{Co}_x$, are presented. These are discussed and compared to corresponding experimental data.

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I. INTRODUCTION

The magnetic susceptibility of nonmagnetic transition-metal systems has been studied in great detail in the past.[1–11] Usually the influence of spin-orbit coupling is ignored, and as a consequence, the spin and the orbital contributions to the total susceptibility can be investigated separately. The mechanism giving rise to the Stoner enhancement of the spin susceptibility was successfully investigated within the framework of local spin density approximation (LSDA) to density functional theory.[12–14] Although expressions for the orbital susceptibility were worked out decades ago,[8–10] corresponding numerical studies could be presented only many years later.[4,15,16] This seems to be caused by the fact that these expressions gave the wave-vector-dependent orbital susceptibility $\chi(\vec{q})$ that had to be extrapolated properly to $\vec{q}=0$ to get the static susceptibility for a homogeneous external field. Using linear response theory on the basis of the Green's function technique, a proper decomposition of the orbital susceptibility into its Van Vleck, Landau, and diamagnetic parts, could be made.[4] The influence of relativistic effects on the magnetic susceptibility has been studied by various authors.[3,5,17] As was demonstrated by Yasui and Shimizu,[18] spin-orbit coupling gives rise to a cross-term contribution χ_{so} to the susceptibility. Staunton[3] worked out a relativistic expression for the Stoner-enhanced spin susceptibility of disordered alloys that was first applied to pure transition metals.[19] This approach was extended by Ebert *et al.*[5,20] to include the orbital Van Vleck term as well, and applied to a number of transition-metal alloy systems.

In contrast to the situation for nonmagnetic transition-metal systems, only relatively few theoretical investigations on the high-field susceptibility of magnetically ordered systems can be found in the literature. The influence of the Stoner enhancement on the high field spin susceptibility of ferromagnets was studied already in 1962.[21] Corresponding numerical investigations were done only around 20 years later within the framework of LSDA on the pure ferromagnets Fe, Co, and Ni.[22] To allow for a direct comparison with experiment, additional calculations were performed on the orbital susceptibility as well.[23] Apart from these promising studies, not much further theoretical work was done, obviously, in the field—presumably, because of the complexity and large numerical effort required by the theoretical formulation used so far (see, for example, Refs. 24 and 11).

As is demonstrated below, the various problems encountered when dealing with the high-field susceptibility of magnetically ordered systems can be overcome by using linear response theory on the basis of the Green's function technique. This very powerful approach allows one to deal with spin and orbital susceptibility on the same footing, to include the Landau susceptibility not considered so far, and to account for spin-orbit induced contributions using a relativistic formulation. The use of the Green's function technique, in addition, allows us to deal in principle with any type of system. This is demonstrated by presenting corresponding results for the transition metals Fe, Co, and Ni, as well as for the alloy system $\mathrm{Fe}_{1-x}\mathrm{Co}_x$, with the latter treated by means of the coherent potential approximation (CPA) alloy theory.

II. EXPRESSION FOR THE HIGH-FIELD SUSCEPTIBILITY

A. Electronic structure

The formal basis for our description of the high-field susceptibility is the presentation of the electronic structure in terms of the retarded single-particle Green's function $G(\vec{r},\vec{r}\,',E)$. A very accurate and flexible expression for $G(\vec{r},\vec{r}\,',E)$ is obtained by using its formulation within the Korringa-Kohn-Rostoker (KKR) method of band structure calculations or, in other words, by applying multiple scattering theory. This leads to the expression[25,26]

$$
\begin{aligned}
G(\vec{r},\vec{r}\,',E) = {} & \sum_{\Lambda\Lambda'} Z^{n}_{\Lambda}(\vec{r},E)\,\tau^{nn'}_{\Lambda\Lambda'}(E)\,Z^{n'\times}_{\Lambda'}(\vec{r}\,',E) \\
& - \sum_{\Lambda} [Z^{n}_{\Lambda}(\vec{r},E)J^{n\times}_{\Lambda}(\vec{r}\,',E)\Theta(r'-r) \\
& + J^{n}_{\Lambda}(\vec{r},E)Z^{n\times}_{\Lambda}(\vec{r}\,',E)\Theta(r-r')]\delta_{nn'}, \qquad (1)
\end{aligned}
$$

where the spatial vectors $\vec{r}$ and $\vec{r}\,'$ are assumed to be within the atomic cell centered at sites n and n', respectively. Within the fully relativistic formulation used here, the combined quantum number $\Lambda=(\kappa,\mu)$ stands for the relativistic spin-orbit and magnetic quantum numbers κ and μ, respectively.[27] Accordingly, Z^n_Λ and J^n_Λ are four-component wave functions obtained as regular and irregular, respectively, solutions to the single-site Dirac equation for the isolated potential well V^n centered at site n. The symbol "×" as a superscript of Z^n_Λ and J^n_Λ indicates the left-hand side solution to the Dirac equation. Dealing with a ferromagnetically ordered system within the framework of LSDA, the potential V^n is spin-dependent. As a consequence, $Z^n_\Lambda = \Sigma_{\Lambda'} Z^n_{\Lambda'\Lambda}$ (and also J^n_Λ) stands for a superposition of various partial waves with spin-angular character Λ'.[28,29] Finally, the quantity $\tau^{nn'}_{\Lambda\Lambda'}$ is the so-called scattering-path operator that represents the transfer of a wave coming in at site n' with character Λ' to a wave outgoing from site n with character Λ and all possible scattering events taking place in-between accounted for in a self-consistent way.[25]

B. Dyson equation and perturbation Hamiltonian

As for paramagnetic systems,[3,5] the influence of an external magnetic field B_{ext} can be accounted for by a linear approximation to Dyson's equation. Accordingly, the Green's function representing the system in the presence of B_{ext} is given by

$$G^B(\vec{r},\vec{r}\,',E) = G(\vec{r},\vec{r}\,',E) + \int d^3r'' G(\vec{r},\vec{r}\,'',E)\Delta\mathcal{H}(\vec{r}\,'')G(\vec{r}\,'',\vec{r}\,',E). \quad (2)$$

In this expression $\Delta\mathcal{H}$, which is proportional to B_{ext}, represents all modifications in the electronic potential that are caused in a direct or indirect way by the external field. Obviously, the perturbation $\Delta\mathcal{H}(\vec{r})$ has to represent the direct Zeeman interaction due to the coupling of the external field to the spin and orbital degrees of freedom. In addition, it represents the change in the single-particle potential due to induced changes in the spin and orbital magnetization and the particle density. Dealing with $\Delta\mathcal{H}(\vec{r})$ within the framework of spin-density functional theory (SDFT) one has

$$\Delta\mathcal{H}(\vec{r}) = \beta\sigma_z\mu_B B_{ext} + \beta\hat{l}_z\mu_B B_{ext} + \Delta v^{xc}(\vec{r}) + \Delta v_H(\vec{r}). \quad (3)$$

Here $\Delta v^{xc}(\vec{r})$ depends on changes in the spin magnetization and particle density, $\Delta m^s(\vec{r})$ and $\Delta n(\vec{r})$, respectively;

$$\Delta v^{xc}(\vec{r}) = (K[n,m^s] + \sigma_z L[n,m^s])\Delta n(\vec{r}) + (L[n,m^s] + \sigma_z J[n,m^s])\Delta m^s(\vec{r}), \quad (4)$$

with

$$K[n,m^s] = \left(\frac{\delta E_{xc}[n,m^s]}{\delta n(r)\,\delta n(r)}\right)_{n=n_0,m^s=m^s_0},$$

$$L[n,m^s] = \left(\frac{\delta E_{xc}[n,m^s]}{\delta n(r)\,\delta m^s(r)}\right)_{n=n_0,m_s=m^s_0},$$

$$J[n,m^s] = \left(\frac{\delta E_{xc}[n,m^s]}{\delta m^s(r)\,\delta m^s(r)}\right)_{n=n_0,m^s=m^s_0}, \quad (5)$$

where the index 0 indicates the various densities of the undistorted system without field. The field-induced change in the particle density $\Delta n(\vec{r})$ not only gives rise to a change in the exchange-correlation potential according to Eq. (4), but also to a direct Hartree term

$$\Delta v_H(\vec{r}) = \int d^3r' \frac{e^2\Delta n(\vec{r}\,')}{|\vec{r}-\vec{r}\,'|}. \quad (6)$$

In principle, $\Delta\mathcal{H}$ given by Eq. (3) should also contain terms that represent changes in the exchange-correlation potential due to the orbital magnetization. A formulation of such terms is possible within the framework of current-density functional theory (CDFT).[30] As the corresponding parametrization for the exchange-correlation potential available so far are not completely satisfying,[31,32] we ignore an orbital exchange-correlation contribution to $\Delta\mathcal{H}$ in the following.

In dealing with Eqs. (3)–(6), it is advantageous to represent the induced changes in the densities by a product of the corresponding spatial-resolved local susceptibilities $\chi^d_n(\vec{r})$ for atomic sites n and the external field B_{ext} with $d=s$, o, or c for the spin and orbital magnetization and particle density, respectively. For the changes in the magnetization densities $\Delta m^{s(o)}(\vec{r})$, one may split off in addition, the normalized density response function $\gamma^{s(o)}_n(\vec{r})$ leading to

$$\Delta m^{s(o)}(\vec{r}) = \gamma^{s(o)}_n(\vec{r})\chi^{s(o)}_n B_{ext}. \quad (7)$$

A rather good approximation for $\gamma^{s(o)}_n(\vec{r})$ is obtained by assuming that the spatial variation of the induced changes in the densities is primarily caused by a repopulation of the states around the Fermi level.[12,14] Using Eqs. (3)–(7), the reduced perturbation $\Delta\bar{\mathcal{H}} = \Delta\mathcal{H}/B_{ext}$ can be written in the compact form

$$\Delta\bar{\mathcal{H}}(\vec{r}) = \beta\sigma_z\mu_B + \beta\hat{l}_z\mu_B + \Delta v^s_n(\vec{r})\gamma^s_n(\vec{r})\chi^s_n + \Delta v^c_n(\vec{r})\chi^c_n(\vec{r}) + \int d^3r' \frac{e^2\chi^c_n(\vec{r}\,')}{|\vec{r}-\vec{r}\,'|}, \quad (8)$$

where $\Delta v^d_n(\vec{r})$ are given by the exchange-correlation kernels in Eqs. (4) and (5). Finally, it should be noted that the coupling of the external field to the orbital degree of freedom represented by Eq. (8) gives rise only to the Van Vleck susceptibility. Making use of the decomposition of the orbital susceptibility worked out by Benkowitsch and Winter,[4] the additional Landau and diamagnetic contributions are dealt with separately (see below).

C. Expressions for the high-field susceptibility

Obviously, any change $\Delta\langle\mathcal{A}\rangle$ of an observable $\mathcal{A}$ with respect to the field-free case can be derived from the second term in Eq. (2). However, in contrast to a nonmagnetic system, a shift of the Fermi energy ΔE_F caused by B_{ext} cannot be excluded in general. Accordingly, one has for the corresponding spatial density

$$\Delta\langle\mathcal{A}(\vec{r})\rangle = -\frac{1}{\pi}\Delta E_F \,\mathrm{Tr\,Im}\, \mathcal{A}G(\vec{r},\vec{r},E_F) - \frac{1}{\pi}\mathrm{Tr\,Im}\int^{E_F} dE \times \int_{all\ space} d^3r' \,\mathcal{A}G(\vec{r},\vec{r}',E)\Delta\mathcal{H}(\vec{r}')G(\vec{r}',\vec{r},E). \tag{9}$$

To account for all contributions to $\Delta\mathcal{H}$ [see Eqs. (3)–(6)] we have to consider in the following the field-induced spin and orbital magnetization as well as the particle density connected with the operators $\mathcal{A}=\beta\sigma_z\mu_B$, $\beta\hat{l}_z\mu_B$, and $\hat{1}$, respectively. The corresponding changes $\Delta\langle\mathcal{A}\rangle_n$ in the spin and orbital magnetic moments and particle number within atomic cell n are obtained from Eq. (9) by integrating over the cell volume Ω_n. For the induced spin magnetic moment m_n^s on the site n one gets this way, for example,

$$\Delta m_n^s = -\frac{\mu_B}{\pi}\Delta E_F \,\mathrm{Tr\,Im}\,\beta\sigma_z G(E_F) - \frac{\mu_B}{\pi}\mathrm{Tr\,Im}\int^{E_F} dE\beta\sigma_z G\Delta\mathcal{H}G, \tag{10}$$

where the spatial arguments and integrations on the right-hand side have been omitted [see Eq. (9)] and the Trace operation implicitly includes the spatial integration over the volume of cell n.

Restricting for the moment to a one-component system with one atom per unit cell, the charge conservation imposes the restriction $\Delta n_n=0$ on the induced charge Δn_n for all sites n. Evaluating Δn_n via an expression corresponding to Eq. (10) allows one to determine the shift in the Fermi energy by making use of the charge conservation

$$\Delta E_F = -\frac{\mathrm{Tr\,Im}\int^{E_F} dEG\Delta\mathcal{H}G}{\mathrm{Tr\,Im}\,G(E_F)}. \tag{11}$$

This way ΔE_F can be eliminated in Eq. (10) and all other corresponding equations.

Dividing the induced spin and orbital magnetic moments and change in the particle number by the external magnetic field, the corresponding site projected susceptibilities $\chi_n^{\mathcal{A}} = \Delta\langle\mathcal{A}\rangle_n/B_{ext}$ are obtained. Making use of Eq. (11) one is led, for example, for the high-field spin susceptibility to

$$\chi_n^s = \frac{\mu_B}{\pi}\frac{\mathrm{Tr\,Im}\int^{E_F} dEG\Delta\bar{\mathcal{H}}G}{\mathrm{Tr\,Im}\,G(E_F)}\mathrm{Tr\,Im}\,\beta\sigma_z G_n(E_F) - \frac{\mu_B}{\pi}\mathrm{Tr\,Im}\int^{E_F} dE\beta\sigma_z G_n\Delta\bar{\mathcal{H}}G_n \tag{12}$$

$$= -\frac{\mu_B}{\pi}\frac{P_n^s(E_F)}{n(E_F)}\mathrm{Tr\,Im}\int^{E_F} dEG\Delta\bar{\mathcal{H}}G - \frac{\mu_B}{\pi}\mathrm{Tr\,Im}\int^{E_F} dE\beta\sigma_z G_n\Delta\bar{\mathcal{H}}G_n. \tag{13}$$

Here Tr Im $G(E_F)$ and Tr Im $\beta\sigma_z G_n(E_F)$ have been replaced by means of the total density of states per unit cell and partial spin polarization on site n, $n(E_F)$, and $P_n^s(E_F)$ at the Fermi energy, respectively. From this expression it is obvious that the first term does not appear for nonmagnetic solids as in this case $P_n^s(E_F)$ is zero. Equation (13) gives the spin susceptibility in a very general form that accounts for all enhancement effects as well as a possible influence of other susceptibilities. In addition, it can be applied in principle to any type of system. Nevertheless, using some simplifications it can be given a very transparent form and interpretation. In particular, it contains the well-known expression for the Stoner-enhanced high-field spin susceptibility[21] as a special case. This is shown in detail in Appendix A.

Inserting the expression for the induced perturbation $\Delta\bar{\mathcal{H}}$ given in Eq. (8) into Eq. (13), χ_n^s is expressed in terms of the partial susceptibilities χ_m^d $(d=s,o,c)$ for all other sites m. This is shown in some detail in Appendix B. For a periodic system the partial susceptibilities with one atom per unit cell do not depend on the site index m. If all contributions of the sites m are summed up, Eq. (13) leads to an inhomogeneous linear equation for χ^s in terms of all other susceptibilities χ^d $(d=s,o,c)$. Corresponding equations can be set up for χ^o and χ^s leading to a set of inhomogeneous linear equations for the unknown susceptibilities χ^d that can be solved by matrix inversion.

Having a periodic system with more than one site q per unit cell, or a disordered alloy with several components α with concentration x_α, the charge conservation ensures having $\Sigma_q\Delta n_q=0$ or $\Sigma_\alpha x_\alpha\Delta n_\alpha=0$, respectively. While this restriction still allows one to fix the shift in the Fermi energy ΔE_F via Eq. (11), it also implies that one might have a charge redistribution within the unit cell or among the components of an alloy induced by an external field. Also, the considerations following Eq. (13) have to be extended only slightly. For compounds or alloys, the local susceptibilities χ_n^d for different sites n are only identical if they are occupied by the same atom type. Accordingly, one has to set up an equation like Eq. (13) for each type d of susceptibility χ_t^d and each inequivalent atom type t, separately. This leads finally to a set of $3\times N_t$ equations for the various partial susceptibilities with N_t the number of inequivalent atom types, which again can be solved by matrix inversion.

The possible shift of the Fermi energy represented by Eq. (12) obviously leads to the interconnection of the various types of susceptibility. An additional interconnection is caused by spin-orbit coupling, which leads to spin and orbital cross terms in the susceptibility. This means that inclusion of the coupling of the external field to the orbital degree of freedom in the Hamiltonian $\Delta\mathcal{H}$ [see Eq. (3)] leads to a contribution to the spin susceptibility of the second term in Eq. (13). As this spin-orbit susceptibility is relatively small for the systems considered here, it will be combined in the following with the standard spin-spin or Pauli susceptibility to the spin susceptibility χ^s. Analogously, the orbital susceptibility χ^o will combine the standard orbital-orbital Van Vleck susceptibility with the orbital-spin susceptibility that occurs due to the inclusion of spin-orbit coupling within our relativistic formulation.

D. Landau and diamagnetic susceptibility

The orbital susceptibility of nonmagnetic solids has been investigated in detail already by Benkowitsch and Winter[4] adopting a nonrelativistic approach. Accordingly, we present only the main steps necessary for an extension to a relativistic formulation applicable, in particular, also to ferromagnetic solids. For the sake of transparency we restrict to a pure system with one atom per unit cell.

The starting point is to express the total current density $\vec{j}(\vec{r})$ induced by an external magnetic field represented by a vector potential $\vec{A}(\vec{r})$,

$$\vec{j}(\vec{r}) = \int d^3r' \mathbf{D}(\vec{r},\vec{r}')\vec{A}(\vec{r}'), \tag{14}$$

where $\mathbf{D}(\vec{r},\vec{r}')$ is the current-current correlation function. Within a relativistic approach, the total current density is represented by the operator[27]

$$\vec{j} = -ec\vec{\boldsymbol{\alpha}} \tag{15}$$

with e the modulus of the electronic charge, c the speed of light, and $\vec{\boldsymbol{\alpha}}$ the vector of Dirac matrices. To separate $\vec{j}$ into its spin and orbital parts, a Gordon decomposition can be made that leads to an electronic state i represented by a four-component wave function $\phi_i(\vec{r})$ to[18,27]

$$\begin{aligned}\phi_i^\dagger(\vec{r})\vec{j}\phi_i(\vec{r}) = {}& \frac{ie\hbar}{2m}[\phi_i^\dagger(\vec{r})\beta(\vec{\nabla}\phi_i(\vec{r})) - (\vec{\nabla}\phi_i^\dagger(\vec{r}))\beta\phi_i(\vec{r})] \\ & - \frac{e^2}{mc}\phi_i^\dagger(\vec{r})\beta\phi_i(\vec{r})\vec{A}(\vec{r}) \\ & - \frac{e\hbar}{2m}\vec{\nabla}\times(\phi_i^\dagger(\vec{r})\beta\vec{\boldsymbol{\sigma}}\phi_i(\vec{r}))\end{aligned} \tag{16}$$

with β one of the standard Dirac matrices. The first two terms can be identified with the counterparts to the nonrelativistic form of the paramagnetic and diamagnetic current density contributions, while the third term represents the spin-current-density contribution. Accordingly, the expression worked out by Benkowitsch and Winter for the correlation function can be used with minor corrections to deal with the orbital part of the induced current density leading to

$$\begin{aligned}\mathbf{D}_{ij}(\vec{r},\vec{r}') = {}& +\frac{e^2}{mc}\frac{1}{\pi}\,\mathrm{Im\,Tr}\int dE\beta G(\vec{r},\vec{r},E)\delta(\vec{r},\vec{r}')\delta_{ij} \\ & - \frac{e^2\hbar^2}{m^2c}\frac{1}{\pi}\,\mathrm{Im\,Tr}\int dE\beta\nabla_i G(\vec{r},\vec{r}',E)\beta\nabla_j G(\vec{r}',\vec{r},E)\end{aligned} \tag{17}$$

with i and j indicating Cartesian coordinates (x,y,z). Here we have to interpret $G(\vec{r},\vec{r}',E)$ as a 4×4-matrix function [see Eq. (1)] and the Dirac matrix β occurs in addition. Due to these minor modifications, the subsequent derivation of the wave-vector-dependent orbital susceptibility can be generalized straightforwardly from the nonrelativistic to the fully relativistic version. This also applies to the analytical extrapolation to the case $\vec{q}=0$. This way one gets for the zz component of the full susceptibility tensor representing the induced magnetization along z due to a magnetic field along z, the expression

$$\begin{aligned}\chi_{zz} = {}& \frac{e^2\hbar^2}{m^2c^2}\frac{1}{\pi}\frac{1}{\Omega}\,\mathrm{Im\,Tr}\int dE\int_\Omega d^3r\int d^3r'\beta\nabla_x G(\vec{r},\vec{r}',E) \\ & \times\beta\nabla_{x'}G(\vec{r}',\vec{r},E)\frac{1}{2}(y-y')^2,\end{aligned} \tag{18}$$

where Ω is the volume of an atomic cell and we are restricted to a system of cubic symmetry.

Splitting off from χ_{zz}, the diamagnetic susceptibility

$$\chi^{dia} = \frac{e^2}{4mc^2}\frac{1}{\pi}\frac{1}{\Omega}\,\mathrm{Im\,Tr}\int dE\int_\Omega d^3r(x^2+y^2)\beta G(\vec{r},\vec{r},E) \tag{19}$$

and the Van Vleck paramagnetic susceptibility

$$\begin{aligned}\chi^{VV} = {}& \frac{e^2\hbar^2}{4m^2c^2}\frac{1}{\pi}\frac{1}{\Omega}\,\mathrm{Im\,Tr}\int dE\int_\Omega d^3r\int d^3r' \\ & \times\beta\hat{l}_z G(\vec{r},\vec{r}',E)\beta\hat{l}_{z'}G(\vec{r}',\vec{r},E),\end{aligned} \tag{20}$$

using the relativistic extensions to their standard expressions[33] one ends up with the following expression for the Landau susceptibility:

$$\begin{aligned}\chi^{Lan} = {}& \frac{e^2\hbar^2}{m^2c^2}\frac{1}{\pi}\frac{1}{\Omega}\,\mathrm{Im\,Tr}\int dE\int_\Omega d^3r \\ & \times\sum_m\int d^3r'_m\Big[\beta\nabla_x G(\vec{r},\vec{r}'_m,E)\beta y'_m Y'_m\nabla_{x'}G(\vec{r}'_m,\vec{r},E) \\ & + \frac{1}{2}\beta\nabla_x G(\vec{r},\vec{r}'_m,E)\beta Y'^2_m\nabla_{x'}G(\vec{r}'_m,\vec{r},E)\Big],\end{aligned} \tag{21}$$

where Y_m is the y component of the lattice vector $\vec{R}_m$ of cell m measured from the central cell. From this expression one can see that χ^{Lan} represents a contribution to the orbital magnetization with an atomic cell, which stems from induced currents outside this cell. Accordingly, it has only a negligible counterpart when dealing with hyperfine interaction[34] or neutron scattering.

To evaluate Eq. (21), the expression for the Green's function given by Eq. (1) has to be inserted leading to

$$\chi^{Lan} = \frac{e^2\hbar^2}{m^2c^2}\frac{1}{\pi}\frac{1}{\Omega}\,\mathrm{Im}\int dE \sum_{m}\sum_{\Lambda\Lambda'}\sum_{\Lambda''\Lambda'''}\tau^{0m}_{\Lambda\Lambda'}(E)\,\tau^{m0}_{\Lambda''\Lambda'''}(E)$$
$$\times\Bigg[Y'_m\int_\Omega d^3r Z^{\times}_{\Lambda'''}(\vec{r},E)\beta\nabla_x Z_\Lambda(\vec{r},E)$$
$$\times\int_\Omega d^3r'_m Z^{\times}_{\Lambda'}(\vec{r}'_m,E)\beta y'_m\nabla_{x'}Z_{\Lambda''}(\vec{r}'_m,E)$$
$$+\frac{1}{2}Y'^2_m\int_\Omega d^3r Z^{\times}_{\Lambda'''}(\vec{r},E)\beta\nabla_x Z_\Lambda(\vec{r},E)$$
$$\times\int_\Omega d^3r'_m Z^{\times}_{\Lambda'}(\vec{r}'_m,E)\beta\nabla_{x'}Z_{\Lambda''}(\vec{r}'_m,E)\Bigg]. \qquad (22)$$

The sum over the lattice sites m involving the product of the scattering path operators τ^{0m} and τ^{m0} can in principle be summed up exactly in a similar way as indicated by Eq. (B2) in Appendix B. However, because of the occurrence of the lattice vector component Y_m, this leads to expressions that involve integrands of the form $\tau(\vec{k})\frac{\partial}{\partial k_y}\tau(\vec{k})$ and $\tau(\vec{k})\frac{\partial^2}{\partial k_y^2}\tau(\vec{k})$ that are numerically difficult to be handled. For this reason we kept the real-space expression given by Eq. (22) and made sure that the result is properly converged with respect to the summation over lattice sites. This is in general achieved if about 12 atomic shells around the central site are included.

III. RESULTS AND DISCUSSION

A. The pure ferromagnets Fe, Co, and Ni

The formalism presented above has been implemented using the spin-polarized relativistic version of the KKR band-structure method[26] making use of the ASA (atomic sphere approximation). Exchange and correlation have been dealt within the framework of LSDA using the parametrization for the exchange-correlation potential given by Vosko *et al.*[35]

Dealing with the high-field susceptibility of ferromagnets it is—in contrast to nonmagnetic solids—necessary to consider the impact of the external field on the particle density [see Eqs. (3)–(6) and (11)]. For elemental systems there is no field-induced charge transfer among the atomic cells but a charge rearrangement within an atomic cell may nevertheless occur. Corresponding results for the spatially resolved particle-density susceptibility $\chi^c(r)$ of Fe is shown in Fig. 1 together with the integral function $I^c(r)=\int_0^r r'^2 dr'\,\chi^c(r')$. As one notes, the integral goes to zero for the Wigner-Seitz radius r_{WS}, reflecting the charge conservation. The variation of $\chi^c(r)$ with r obviously corresponds to a shift of the charge distribution closer to the nucleus. This charge rearrangement is primarily caused by changes in the population of states close to the Fermi energy. In this context it is important to note that—in contrast to a nonmagnetic solid—states at the Fermi energy with different spin character may have very different angular momentum character. In addition, the radial wave functions depend on the spin character.

As reflected by Eq. (3), a field-induced change in the particle density modifies the effective electronic potential and that way influences the ordinary spin and orbital susceptibilities. The corresponding modification of the exchange-correlation potential is expressed in Eq. (4) in terms of the exchange-correlation interaction kernels K, L, and J defined in Eq. (5) as variation of the exchange-correlation energy E_{xc} with respect to the particle density n and spin magnetization m_s. Figure 2 presents for bcc-Fe the result for K, L, and J as a function of the spatial variable r. As one notes, the cross term $L(r)$ that is connected with the induced change in the particle density Δn and spin magnetization is rather small and negligible. The other two terms, $J(r)$ and $K(r)$, are much larger and of the same order of magnitude. Here it should be noted that $J(r)$ is the only term to be considered when dealing with the Stoner enhancement of the spin susceptibility of nonmagnetic solids. It should also be noted that the results for L and J, similar to those given in Fig. 1, have been obtained by Yamada *et al.*[22] [these authors did not present results for $K(r)$].

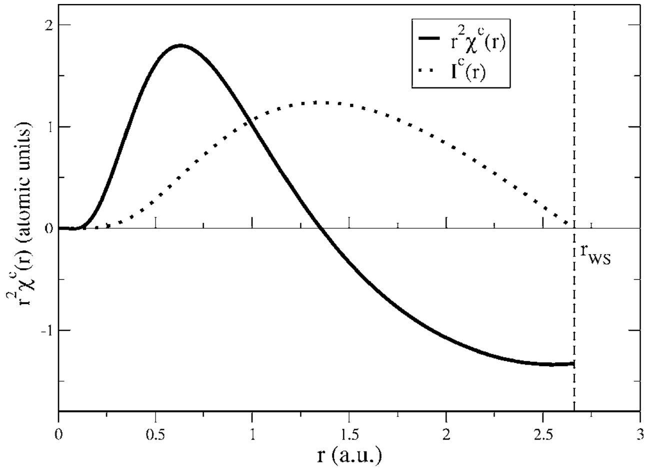

FIG. 1. Spatially resolved particle density susceptibility $\chi^c(r)$ (multiplied by r^2) of bcc-Fe together with the integral function $I^c(r)=\int_0^r r'^2 dr'\,\chi^c(r')$. The vertical dashed line indicates the Wigner-Seitz radius r_{WS}.

Table I summarizes the results for the various contributions to the high-field susceptibility of bcc-Fe, hcp-Co, and

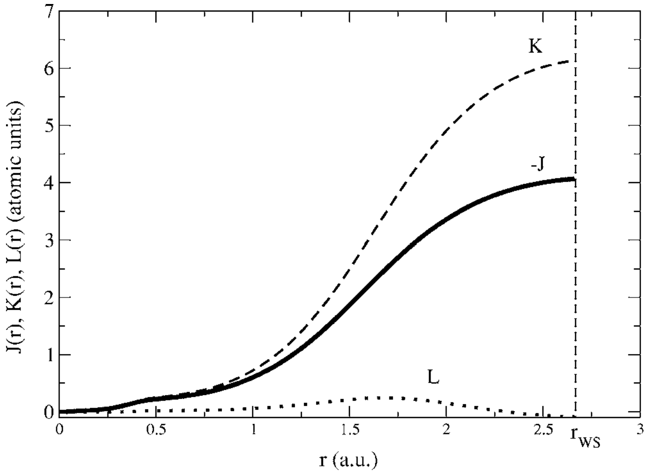

FIG. 2. Spatially resolved exchange-correlation interaction kernels $J(r)$, $K(r)$, and $L(r)$ [defined in Eq. (5)] of bcc Fe.

TABLE I. Theoretical (first line) spin, orbital, and Landau susceptibilities χ^s, χ^o, and χ^{Lan}, respectively, for the elemental ferromagnets bcc Fe, hcp Co, and fcc Ni. (χ^{Lan} for hcp Co has been evaluated for fcc-Co.) Adding the diamagnetic susceptibility (−20.4, −19.7, −19.4) the given total susceptibility is obtained. These results are compared with theoretical results of other authors (second line) and with experiment (all data are given in units of 10^{-6} emu/mol).

	bcc-Fe				hcp-Co				fcc-Ni			
	Spin	Orbital	Landau	Total	Spin	Orbital	Landau	Total	Spin	Orbital	Landau	Total
Theoretical	37.9	168.5	29.8	215.7	17.9	144	6.4	148.5	35.5	75.3	2.5	93.9
Theoretical	37	101		138								
Experimental				231[a]				210.3[b]				93.3[b]
				306[c]								111.3[c]
				201[b]								
				266[d]								

[a] Reference 37.
[b] Reference 38.
[c] Reference 39.
[d] Reference 40.

fcc-Ni. Obviously, the spin susceptibility χ^s is clearly dominated by the other terms and contributes at most around 1/3 to the total susceptibility in the case of Ni. The orbital contribution χ^o is by far largest in all cases. Because the spin-orbit coupling strength is relatively small for the $3d$ elements, χ^o stems primarily from the Van Vleck term, i.e., the orbital-spin cross contribution is much smaller. A rather large Van Vleck contribution is not unexpected as an estimate for it can be made using the expression $n_o n_u/\Delta E$, where $n_{o(u)}$ is the number of occupied (unoccupied) d states and ΔE is the width of the d band.[1] In line with this simplified consideration one finds χ^o to decrease upon filling of the d band when going from Fe to Co and Ni (for a more stringent test, of course, one should use data obtained for the same crystal structure).[36] Table I shows that there is also an appreciable contribution coming from the Landau susceptibility. As found before for several nonmagnetic transition metals,[4] it turned out to have positive sign and should therefore not be called diamagnetic. Including the diamagnetic susceptibility term χ^{dia}, which goes monotonously with the number of electrons, one obtains the total susceptibilities given in Table I that compare very well with the available experimental data. For the spin susceptibility of Fe, fairly good agreement with the previous work of Yamada *et al.* is found.[22] However, in the case of the orbital susceptibility agreement is not as good. To some extent this is due to the fact that χ^o given by Yamada *et al.* includes all nonspin contributions. More important seems to be the influence of numerical problems connected with the formalism used by these authors to evaluate χ^o.

B. Application to $\mathrm{Fe}_{1-x}\mathrm{Co}_x$

The great advantage of the formalism presented in Sec. II is that it can be applied without problems to complex systems including, in particular, disordered alloys. This feature has been exploited here for a detailed study of the high-field susceptibility of bcc-$\mathrm{Fe}_{1-x}\mathrm{Co}_x$ alloys that included an investigation on the influence of structural ordering in the concentration regime around $x \approx 0.5$.

As demonstrated already for pure Fe (see Fig. 1), the application of an external field leads also for disordered $\mathrm{Fe}_{1-x}\mathrm{Co}_x$ as well as ordered FeCo, to a charge rearrangement. This is demonstrated in Fig. 3 by the results for the partial spatially resolved particle density susceptibility $\chi^c_\alpha(r)$ for α = Fe and Co in ordered FeCo having CsCl structure. While $\chi^c_{\mathrm{Fe}}(r)$ of Fe is similar to that of pure Fe (Fig. 1), one notes that it has a positive sign throughout. On the other hand, $\chi^c_{\mathrm{Co}}(r)$ of Co has a negative sign for $r \leqslant 2$ a.u. This implies that the field leads to a charge transfer from Co to Fe. The corresponding integral function $I^c(r)$, also shown in Fig. 3, goes to 0 at the Wigner-Seitz radius reflecting the charge conservation.

Similar results have also been obtained for the disordered alloy $\mathrm{Fe}_{1-x}\mathrm{Co}_x$. Corresponding results for the element-resolved integral function $I^c_\alpha(r_{WS}) = \chi^c_\alpha$ for the particle number are given in Fig. 4. As the integration is done up to the Wigner-Seitz radius r_{WS}, $I^c_\alpha(r_{WS})$ reflects here the charge transfer from Co to Fe as a function of the concentration x.

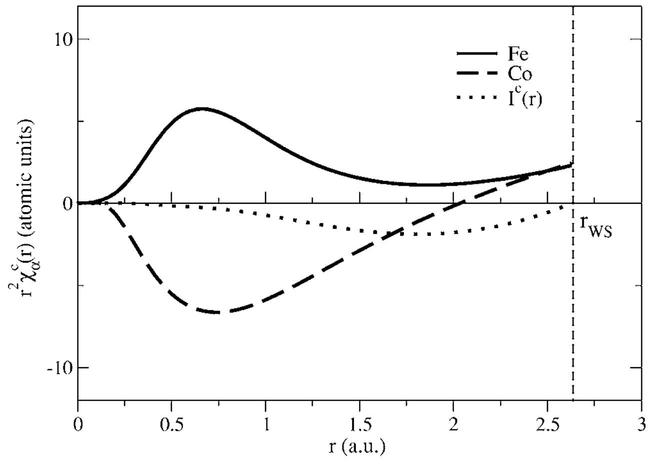

FIG. 3. Partial spatially resolved particle density susceptibility $\chi^c_\alpha(r)$ (multiplied by r^2) for α=Fe and Co in the compound FeCo. The dotted line gives the integral function $I^c(r) = \int_0^r r'^2 dr' [\chi^c_{\mathrm{Fe}}(r') + \chi^c_{\mathrm{Co}}(r')]$.

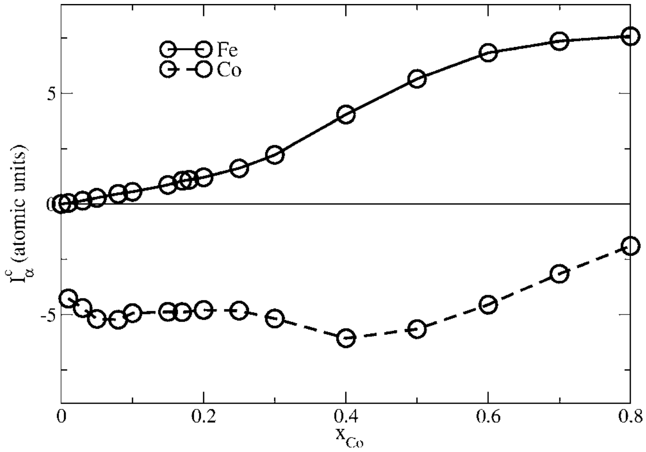

FIG. 4. Element-resolved integral function $I^c_\alpha(r_{WS})=\chi^c_\alpha$ for the particle number of Fe and Co in disordered $Fe_{1-x}Co_x$ reflecting the charge transfer from Co to Fe.

Again, adding the two functions $I^c_\alpha(r_{WS})$ weighted by the concentration x_α, one gets zero due to charge conservation.

The spin susceptibility of disordered bcc $Fe_{1-x}Co_x$ is shown in Fig. 5 in a component resolved way. Obviously, the pronounced maximum in $\chi^s(x)$ at around $x\approx 0.07$ is primarily caused by the variation of $\chi^s_{Fe}(x)$ with composition. The contribution of Co, $\chi^s_{Co}(x)$, on the other hand, shows a much weaker concentration dependence. Nevertheless, for $x>0.3$ it compensates the decrease of $\chi^s_{Fe}(x)$ leading to a second broad maximum of $\chi^s(x)$ around $x=0.6$. Ignoring the Stoner enhancement, the partial spin susceptibilities $\chi^{s0}_\alpha(x)$ would directly reflect the corresponding component and spin-resolved density of states (DOS) at the Fermi level $n^{\uparrow(\downarrow)}_\alpha(E_F,x)$ (see Appendix A). The corresponding unenhanced spin susceptibility $\chi^{s0}(x)=(1-x)\chi^{s0}_{Fe}(x)+x\chi^{s0}_{Co}(x)$ is shown in Fig. 5 together with its enhanced counterpart $\chi^s(x)$. Obviously, the concentration dependence of $\chi^s(x)$ follows closely that of the unenhanced $\chi^{s0}(x)$ implying that its variation with x is essentially determined by that of $n^{\uparrow(\downarrow)}_\alpha(E_F,x)$. On the other hand, one notes that the Stoner enhancement represented by the ratio $\chi^s(x)/\chi^{s0}(x)$ varies in a nonmonotonous way with composition, being lowest around $x=0.3$.

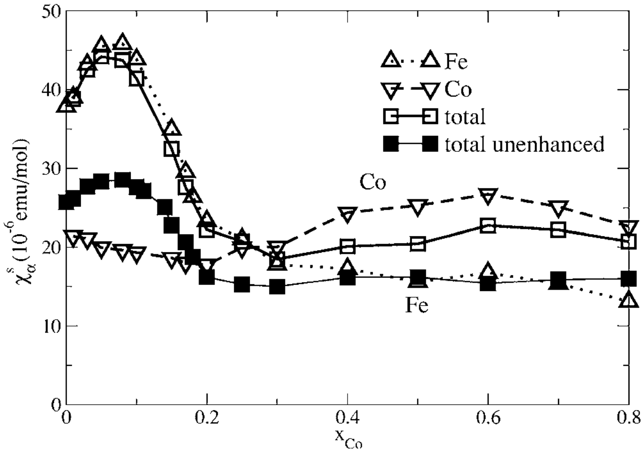

FIG. 5. Element-resolved contributions to the spin-magnetic susceptibility χ^s_α of Fe and Co in disordered bcc $Fe_{1-x}Co_x$.

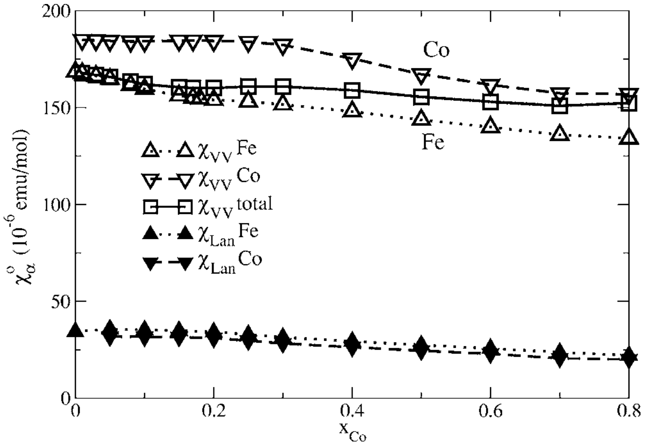

FIG. 6. Element-resolved contributions to the orbital-magnetic susceptibility χ^o_α of Fe and Co in disordered bcc $Fe_{1-x}Co_x$.

As found already for the pure elements, the high-field susceptibility of disordered bcc-$Fe_{1-x}Co_x$ is dominated by its orbital contribution χ^o that combines the Van Vleck and orbital-spin contributions. As found before for paramagnetic transition metal alloys,[5,20,41] a monotonous and rather weak concentration dependence is found for the partial susceptibilities $\chi^o_{Fe}(x)$ and $\chi^o_{Co}(x)$ (Fig. 6). As these are by far dominated by their Van Vleck part, this can again be explained on the basis of the expression $n_o n_e/\Delta E$ already mentioned when discussing χ^o for the pure elements. As these parameters should not change too much with concentration, the same holds true for χ^o.

Figure 6 also shows results for the Landau susceptibility $\chi^{Lan}_\alpha(x)$ in a component-resolved way. As expected from the results for the pure elements given in Table I, $\chi^{Lan}_\alpha(x)$ is positive and comparable to $\chi^s_\alpha(x)$ throughout the whole concentration regime. The finding that $\chi^{Lan}_{Fe}(x)$ and $\chi^{Lan}_{Co}(x)$ are nearly identical can be explained by the fact that χ^{Lan} is primarily determined by the distribution of the field-induced current density around a selected component (see above, as well as Ref. 4). As the CPA is a single-site theory, the effective CPA medium is used for Fe as well as for Co to represent their environment in the disordered alloys. Accordingly, there are not many differences between $\chi^{Lan}_{Fe}(x)$ and $\chi^{Lan}_{Co}(x)$.

The remaining contribution to the total susceptibility is the diamagnetic susceptibility $\chi^{dia}_\alpha(x)$, which is determined by the expectation value $\langle r^2\rangle$ for all core and valence electrons within an atomic cell.[4] According to this, only a weak concentration dependency for the partial susceptibilities $\chi^{dia}_\alpha(x)$ is found that is primarily caused by the variation of the lattice parameter with x. This leads to a very weak and nearly linear variation of $\chi^{dia}(x)$ between the values for pure Fe $(-20.4\times 10^{-6}$ emu/mol) and that of $Fe_{0.2}Co_{0.8}$ $(-20.1\times 10^{-6}$ emu/mol).

Combining all contributions to the high-field susceptibility $\chi(x)$ of disordered bcc-$Fe_{1-x}Co_x$ we find $\chi(x)$ to be nearly constant between $x=0$ and 0.08 because the variation of $\chi^s(x)$ and $\chi^o(x)$ nearly cancel each other (see Fig. 7). For higher Co concentrations x up to 0.18, both contributions

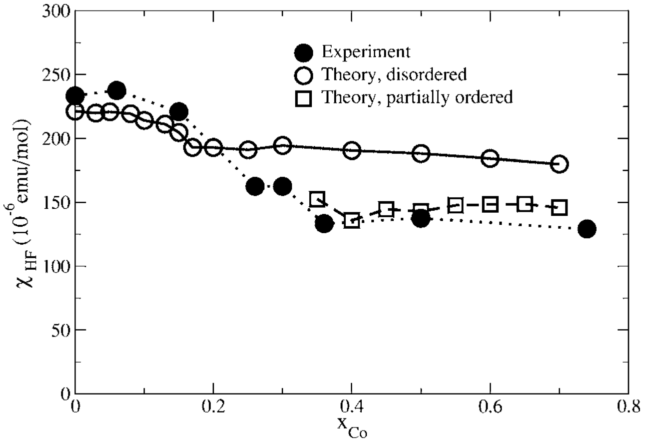

FIG. 7. Total high-field magnetic susceptibility of bcc $Fe_{1-x}Co_x$ alloys. Empty circles correspond to randomly disordered alloys, while empty squares correspond to partially ordered alloys (see text). Experimental data are given by filled circles (Ref. 37).

decrease leading to a substantial drop in the total susceptibility. With x increasing further, the ongoing decrease of $\chi^o(x)$ is more or less outweighed by an increase of the spin susceptibility $\chi^s(x)$ leading to a total susceptibility $\chi(x)$ that is only very weakly concentration-dependent. As Fig. 7 shows, these results are in rather good agreement with available experimental data for x in the regime 0–0.18 while for a higher Co concentration the theoretical susceptibility lies above the experimental one. On the basis of previous work on $Fe_{1-x}Co_x$ (Ref. 42) one may expect that this behavior is due to ordering in the samples. Assuming for $x=0.5$ a perfectly ordered system with CsCl structure, our calculations lead to a reduction by 33.8×10^{-6} emu/mol compared to the disordered alloy. As can be seen in Fig. 7, this brings the theoretical result, indeed, in very good agreement with experiment. Apart from the diamagnetic susceptibility, all other contributions to the total one are affected by the structural change. The reduction of χ^s by 5.0×10^{-6} emu/mol upon ordering can be traced back easily by a corresponding change in the DOS at the Fermi energy. The disorder in $Fe_{0.5}Co_{0.5}$ causes a smearing out of the DOS that results in an increase of the DOS at the Fermi level compared to ordered FeCo. As a consequence, the spin susceptibility χ^s is higher for the disordered state. The change of the orbital susceptibility χ^o upon ordering can also be explained by the structural dependency of the DOS. A reduction of the DOS around the Fermi level with ordering should lead, according to the standard expression for the Van Vleck susceptibility,[1] to a reduction in χ^o. In fact our calculations for the ordered FeCo give a value 14.4×10^{-6} emu/mol lower than for disordered $Fe_{0.5}Co_{0.5}$. The largest change upon ordering is found for the Landau susceptibility, that is, 18.1×10^{-6} emu/mol lower for the ordered than for the disordered state.

As the theoretical results obtained for ordered FeCo agree very well with available experimental data for $x=0.5$, it seems plausible that the measurements were performed on ordered or partly ordered samples (see also Ref. 42). Therefore additional calculations were done for the concentration regime around $x=0.5$ for which a partially ordered CsCl structure was assumed. For this purpose it was assumed that for $x<0.5$ ($x>0.5$) one sublattice is completely occupied by Fe (Co) while the other is occupied randomly by Fe and Co atoms according to the global concentration x.[42] Using this prescription to model partly ordered alloys leads to a very satisfying agreement of the calculated susceptibility with experiment as can be seen in Fig. 7. These model calculations, in particular, reproduce the increase of the experimental susceptibility when the Co concentration is decreased below $x=0.35$.

IV. SUMMARY

We have presented a very flexible formalism to calculate the high-field susceptibility of transition-metal systems that is based on the Green's-function technique implemented within the framework of the KKR formalism. This approach gives access to all contributions to the susceptibility and allows for their evaluation. Applications to the pure ferromagnets Fe, Co, and Ni led to a very good agreement with experiment and demonstrated the importance of the Van Vleck and Landau contributions. The extreme flexibility of our approach was demonstrated by applications to disordered $Fe_{1-x}Co_x$ alloys for which, in particular, the impact of ordering was investigated. The achieved results were also found in very satisfying agreement with experiment and indicated the influence of partial ordering for the available experimental susceptibility data.

ACKNOWLEDGMENTS

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APPENDIX A

To reach a transparent interpretation of the general expression for the spin susceptibility χ_n^s given in Eq. (12) we ignore in the following the influence of relativistic effects. This implies the Green's function to be a diagonal 2×2 matrix with respect to the spin index σ ($\sigma=\downarrow,\uparrow$). In addition, we restrict to a periodic solid with one atom per unit cell (i.e., χ_n^s does not depend on the site index n). Allowing the perturbation $\Delta\bar{\mathcal{H}}$ to couple only to the spin [see Eq. (8)] one has

$$\chi^s = \frac{\mu_B}{\pi}\frac{\mathrm{Tr}\,\sigma_z G(E_F)}{\mathrm{Tr}\,G(E_F)}\mathrm{Tr}\int^{E_F} dE G[\mu_B\sigma_z + \Delta V^s(r)\sigma_z\chi^s]G - \frac{\mu_B}{\pi}\mathrm{Tr}\int^{E_F} dE\sigma_z G[\mu_B\sigma_z + \Delta V^s(r)\sigma_z\chi^s]G. \quad \text{(A1)}$$

This expression can be simplified by making use of the relations

$$-\frac{1}{\pi}\mathrm{Tr}\,G(E_F) = n^{\uparrow}(E_F) + n^{\downarrow}(E_F), \quad \text{(A2)}$$

$$-\frac{1}{\pi}\mathrm{Tr}\,\sigma_z G(E_F) = n^{\uparrow}(E_F) - n^{\downarrow}(E_F), \quad \text{(A3)}$$

$$-\frac{1}{\pi}\mathrm{Tr}\int^{E_F} dEG\sigma_z G = n^{\uparrow}(E_F) - n^{\downarrow}(E_F), \tag{A4}$$

$$-\frac{1}{\pi}\mathrm{Tr}\int^{E_F} dE\sigma_z G\sigma_z G = n^{\uparrow}(E_F) + n^{\downarrow}(E_F), \tag{A5}$$

where use has been made of the so-called sum rule

$$G_{\sigma\sigma}(E) = -\int^{E} dE' G_{\sigma\sigma}(E')G_{\sigma\sigma}(E'). \tag{A6}$$

This leads to the expression

$$\begin{aligned}\chi^s = -\mu_B^2\Bigg[&\frac{n^{\uparrow}(E_F) - n^{\downarrow}(E_F)}{n^{\uparrow}(E_F) + n^{\downarrow}(E_F)}[n^{\uparrow}(E_F) - n^{\downarrow}(E_F)](1 + I\chi^s)\\ &+ [n^{\uparrow}(E_F) + n^{\downarrow}(E_F)](1 + I\chi^s)\Bigg],\end{aligned} \tag{A7}$$

where we used the abbreviation

$$I = \frac{1}{\mu_B}\ \mathrm{Trace}\int^{E_F} dEG\Delta V^s\sigma_z G. \tag{A8}$$

Finally, one obtains in this way the expression for the high-field spin susceptibility as derived within the Stoner-Wohlfarth picture[21]

$$(\chi^s)^{-1} = \frac{1}{4\mu_B^2}\left(\frac{1}{n^{\uparrow}(E_F)} - \frac{1}{n^{\downarrow}(E_F)} - I\right), \tag{A9}$$

where I plays role of the Stoner parameter.

APPENDIX B

To deal with the spin susceptibility as given by Eq. (13), the expressions for the Green's function $G(\vec{r},\vec{r}\,',E)$ [Eq. (1)] and the reduced perturbation Hamiltonian $\Delta\bar{\mathcal{H}}(\vec{r})$ [Eqs. (3) and (8)] have to be inserted. Restricting for the sake of clarity here $\Delta\bar{\mathcal{H}}(\vec{r})$ to the coupling to the spin of an electron and one atom site per unit cell, one gets

$$\begin{aligned}\chi_n^s =\ &\frac{\mu_B}{\pi}\frac{P_n^s(E_F)}{n(E_F)}\mathrm{Tr\,Im}\int^{E_F} dE\int_{\Omega_n} d^3rG(\vec{r},\vec{r},E)\Delta\bar{\mathcal{H}}(\vec{r})G(\vec{r},\vec{r},E) - \frac{\mu_B}{\pi}\mathrm{Tr\,Im}\int^{E_F} dE\int_{\Omega_n} d^3r\beta\sigma_z G(\vec{r},\vec{r},E)\Delta\bar{\mathcal{H}}(\vec{r})G(\vec{r},\vec{r},E)\\
=\ &\frac{\mu_B}{\pi}\frac{P_n^s(E_F)}{n(E_F)}\ \mathrm{Im}\int^{E_F} dE\sum_m\sum_{\Lambda_1\Lambda_2\Lambda_3\Lambda_4}\tau^{nm}_{\Lambda_1\Lambda_2}\tau^{mn}_{\Lambda_3\Lambda_4}\int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zm}_{\Lambda_2\Lambda_3}(r_{WS}) + \bar{\mathcal{H}}^{A,xcm}_{\Lambda_2\Lambda_3}(r_{WS})\chi^s_m]\\
&-\sum_{\Lambda_1\Lambda_3\Lambda_4}\tau^{nn}_{\Lambda_3\Lambda_4}\Bigg[\int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})J^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_1\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})Z^n_{\Lambda_1}(\vec{r})[\tilde{\bar{\mathcal{H}}}^{B,Zn}_{\Lambda_1\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{B,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n]\Bigg]\\
&-\sum_{\Lambda_1\Lambda_2\Lambda_3}\tau^{nn}_{\Lambda_1\Lambda_2}\Bigg[\int_{\Omega_n} d^3rJ^{n\times}_{\Lambda_3}(\vec{r})Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_2\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_2\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_3}(\vec{r})Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{C,Zn}_{\Lambda_2\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{C,xcn}_{\Lambda_2\Lambda_3}(r)\chi^s_n]\Bigg]\\
&+\sum_{\Lambda_1\Lambda_3}\Bigg[\int_{\Omega_n} d^3rJ^{n\times}_{\Lambda_3}(\vec{r})J^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_1\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_3}(\vec{r})Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{D,Zn}_{\Lambda_1\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{D,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n]\Bigg]\\
&-\frac{\mu_B}{\pi}\ \mathrm{Im}\int^{E_F} dE\sum_m\sum_{\Lambda_1\Lambda_2\Lambda_3\Lambda_4}\tau^{nm}_{\Lambda_1\Lambda_2}\tau^{mn}_{\Lambda_3\Lambda_4}\int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})\beta\sigma_z Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zm}_{\Lambda_2\Lambda_3}(r_{WS}) + \bar{\mathcal{H}}^{A,xcm}_{\Lambda_2\Lambda_3}(r_{WS})\chi^s_m]\\
&-\sum_{\Lambda_1\Lambda_3\Lambda_4}\tau^{nn}_{\Lambda_3\Lambda_4}\Bigg[\int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})\beta\sigma_z J^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_1\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_4}(\vec{r})\beta\sigma_z Z^n_{\Lambda_1}(\vec{r})[\tilde{\bar{\mathcal{H}}}^{B,Zn}_{\Lambda_1\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{B,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n]\Bigg]\\
&-\sum_{\Lambda_1\Lambda_2\Lambda_3}\tau^{nn}_{\Lambda_1\Lambda_2}\Bigg[\int_{\Omega_n} d^3rJ^{n\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_2\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_2\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{C,Zn}_{\Lambda_2\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{C,xcn}_{\Lambda_2\Lambda_3}(r)\chi^s_n]\Bigg]\\
&+\sum_{\Lambda_1\Lambda_3}\Bigg[\int_{\Omega_n} d^3rJ^{n\times}_{\Lambda_3}(\vec{r})\beta\sigma_z J^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{A,Zn}_{\Lambda_1\Lambda_3}(r) + \bar{\mathcal{H}}^{A,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n] + \int_{\Omega_n} d^3rZ^{n\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z^n_{\Lambda_1}(\vec{r})[\bar{\mathcal{H}}^{D,Zn}_{\Lambda_1\Lambda_3}(r) + \tilde{\bar{\mathcal{H}}}^{D,xcn}_{\Lambda_1\Lambda_3}(r)\chi^s_n]\Bigg].\end{aligned} \tag{B1}$$

Here we have used the auxiliary integrals

$$\bar{\mathcal{H}}^{A,\zeta n}_{\Lambda_1\Lambda_2}(r) = \int_0^r d^3r' Z^{n\times}_{\Lambda_1}(\vec{r}\,')\Delta\bar{\mathcal{H}}^{\zeta}(\vec{r}\,')Z^{n}_{\Lambda_2}(\vec{r}\,'),$$

$$\bar{\mathcal{H}}^{B,\zeta n}_{\Lambda_1\Lambda_2}(r) = \int_0^r d^3r' J^{n\times}_{\Lambda_1}(\vec{r}\,')\Delta\bar{\mathcal{H}}^{\zeta}(\vec{r}\,')Z^{n}_{\Lambda_2}(\vec{r}\,'),$$

$$\bar{\mathcal{H}}^{C,\zeta n}_{\Lambda_1\Lambda_2}(r) = \int_0^r d^3r' Z^{n\times}_{\Lambda_1}(\vec{r}\,')\Delta\bar{\mathcal{H}}^{\zeta}(\vec{r}\,')J^{n}_{\Lambda_2}(\vec{r}\,'),$$

$$\bar{\mathcal{H}}^{D,\zeta n}_{\Lambda_1\Lambda_2}(r) = \int_0^r d^3r' J^{n\times}_{\Lambda_1}(\vec{r}\,')\Delta\bar{\mathcal{H}}^{\zeta}(\vec{r}\,')J^{n}_{\Lambda_2}(\vec{r}\,'),$$

$$\tilde{\bar{\mathcal{H}}}^{X,\zeta n}_{\Lambda_1\Lambda_2}(r) = \bar{\mathcal{H}}^{X,\zeta n}_{\Lambda_1\Lambda_2}(r_{WS}) - \bar{\mathcal{H}}^{X,\zeta n}_{\Lambda_1\Lambda_2}(r) \quad \text{for } X = A,B,C,D$$

with r_{WS} the Wigner-Seitz radius, and have split $\Delta\bar{\mathcal{H}}(\vec{r})$ into its Zeeman ($\zeta = Z$) and exchange-correlation ($\zeta = \mathrm{xc}$) part. These correspond to the first and third terms, respectively, on the right-hand side of Eq. (8) with the spin-susceptibility χ^s_m split off in case of the xc term.

Dealing with a periodic system the susceptibility χ^s_m as well as all radial integrals occurring in Eq. (B1) depend only on the position q within the unit cell. Accordingly, one can split the sum over the atomic sites Σ_m into a sum over the atomic positions q within the unit cell and a sum over the Bravais lattice. The latter one involves only the product of scattering path operators, which can be evaluated exactly by use of Fourier transformation.[43] For one atom per unit cell, for example, one has

$$\sum_m \tau^{nm}_{\Lambda_1\Lambda_2}\tau^{mn}_{\Lambda_3\Lambda_4} = \frac{1}{\Omega_{BZ}}\int_{\Omega_{BZ}} d^3k\, \tau_{\Lambda_1\Lambda_2}(\vec{k})\tau_{\Lambda_3\Lambda_4}(\vec{k}). \tag{B2}$$

Here Ω_{BZ} is the volume of the Brillouin zone and $\tau_{\Lambda_1\Lambda_2}(\vec{k})$ is the inverse of the standard KKR matrix[25]

$$\tau_{\Lambda_1\Lambda_2}(\vec{k}) = [t^{-1} - G(\vec{k})]^{-1}_{\Lambda_1\Lambda_2}, \tag{B3}$$

with t the single-site t matrix and $G(\vec{k})$ the KKR structure constant matrix. With this rearrangement of the terms we end up with an equation for χ^s of the form

$$\chi^s = \frac{P^s(E_F)}{n(E_F)}(T^Z_{cs} + T^{xc}_{cs}\chi^s) + T^Z_{ss} + T^{xc}_{ss}\chi^s, \tag{B4}$$

where the superscripts Z and xc denote terms connected with the Zeeman and exchange-correlation terms defined in Eq. (B1). The subscripts c and s indicate the operators for the particle density and spin magnetization to be involved. To be more specific, T^Z_{ss} is given by

$$\begin{aligned} T^Z_{ss} = -\frac{\mu_B}{\pi}\mathrm{Im}\int^{E_F} dE \sum_{\Lambda_1\Lambda_2\Lambda_3\Lambda_4} \int_\Omega d^3r Z^{\times}_{\Lambda_4}(\vec{r})\beta\sigma_z Z_{\Lambda_1}(\vec{r})\bar{\mathcal{H}}^{A,Z}_{\Lambda_2\Lambda_3}(r_{WS})\frac{1}{\Omega_{BZ}}\int_{\Omega_{BZ}} d^3k\,\tau_{\Lambda_1\Lambda_2}(\vec{k})\tau_{\Lambda_3\Lambda_4}(\vec{k}) \\ - \sum_{\Lambda_1\Lambda_3\Lambda_4} \tau_{\Lambda_3\Lambda_4}\left[\int_\Omega d^3r Z^{\times}_{\Lambda_4}(\vec{r})\beta\sigma_z J_{\Lambda_1}(\vec{r})\bar{\mathcal{H}}^{A,Z}_{\Lambda_1\Lambda_3}(r) + \int_\Omega d^3r Z^{\times}_{\Lambda_4}(\vec{r})\beta\sigma_z Z_{\Lambda_1}(\vec{r})\tilde{\bar{\mathcal{H}}}^{B,Z}_{\Lambda_1\Lambda_3}(r)\right] \\ - \sum_{\Lambda_1\Lambda_2\Lambda_3} \tau_{\Lambda_1\Lambda_2}\left[\int_\Omega d^3r J^{\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z_{\Lambda_1}(\vec{r})\bar{\mathcal{H}}^{A,Z}_{\Lambda_2\Lambda_3}(r) + \int_\Omega d^3r Z^{\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z_{\Lambda_1}(\vec{r})\tilde{\bar{\mathcal{H}}}^{C,Z}_{\Lambda_2\Lambda_3}(r)\right] \\ + \sum_{\Lambda_1\Lambda_3}\left[\int_\Omega d^3r J^{\times}_{\Lambda_3}(\vec{r})\beta\sigma_z J_{\Lambda_1}(\vec{r})\bar{\mathcal{H}}^{A,Z}_{\Lambda_1\Lambda_3}(r) + \int_\Omega d^3r Z^{\times}_{\Lambda_3}(\vec{r})\beta\sigma_z Z_{\Lambda_1}(\vec{r})\tilde{\bar{\mathcal{H}}}^{D,Z}_{\Lambda_1\Lambda_3}(r)\right]. \end{aligned} \tag{B5}$$

Equation (B4) can be straightforwardly solved for the spin susceptibility χ^s. If the full perturbation Hamiltonian in Eq. (8) is used instead of its restriction to its spin part one gets additional terms on the right-hand side of Eq. (B4) connected with the orbital- and particle-density susceptibilities χ^o and χ^c, respectively. Setting up corresponding equations for these, the resulting inhomogeneous linear system of equations can be solved for χ^s, χ^o, and χ^c.

Dealing with a system with many atoms per unit cell, Eq. (B4) has to be set up for each nonequivalent site q in the unit cell. The rearrangement of the lattice summation Σ_m in Eq. (B1) then leads the right-hand side to a sum over the inequivalent lattice sites q' involving the local susceptibilities $\chi^d_{q'}$ ($d = s,o,c$). Again one ends up with an inhomogeneous system of linear equations for the local susceptibilities χ^d_{q}.

When dealing with a disordered alloy, the above scheme has to be modified accordingly.[3] Assuming a binary alloy $A_{x_A}B_{x_B}$ ($x_A + x_B = 1$) with one site per unit cell, one has for the global average susceptibility in terms of the partial susceptibilities χ^s_α,

$$\chi^s = \sum_{\alpha=A,B} x_\alpha \chi^s_\alpha \tag{B6}$$

with χ^s_α calculated by an extension of Eq. (B1). For this purpose a configurational average has to be considered where the site m may be occupied by A or B atoms according to the concentration. This problem has been considered already by Staunton[3] within the CPA for the spin susceptibilities of nonmagnetic alloys. Similar work has been done by Butler[43] that, in particular, include the so-called vertex corrections when dealing with the dc conductivity of disordered alloys. When applying these procedures, one again ends up with a system of linear equations for the partial susceptibilities χ^s_α $(\alpha{=}A,B)$, that can easily be solved. Also, an extension to disordered systems with many sites per unit cell can be done straightforwardly.

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