Relativistic corrections in magnetic systems

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We present a weak-relativistic limit comparison between the Kohn-Sham-Dirac equation and its approximate form containing the exchange coupling, which is used in almost all relativistic codes of density-functional theory. For these two descriptions, an exact expression of the Dirac Green's function in terms of the nonrelativistic Green's function is first derived and then used to calculate the effective Hamiltonian, i.e., Pauli Hamiltonian, and effective velocity operator in the weak-relativistic limit. We point out that, besides neglecting orbital magnetism effects, the approximate Kohn-Sham-Dirac equation also gives relativistic corrections which differ from those of the exact Kohn-Sham-Dirac equation. These differences have quite serious consequences: in particular, the magnetocrystalline anisotropy of an uniaxial ferromagnet and the anisotropic magnetoresistance of a cubic ferromagnet are found from the approximate Kohn-Sham-Dirac equation to be of order $1/c^2$, whereas the correct results obtained from the exact Kohn-Sham-Dirac equation are of order $1/c^4$. We give a qualitative estimate of the order of magnitude of these spurious terms.

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I. INTRODUCTION

The relativistic effects play a fundamental role in magnetic systems. They are responsible for multiple physical properties of great fundamental interest and technological relevance:[1,2] magnetocrystalline anisotropy; magnetostriction; magneto-optical phenomena such as Faraday effect, Kerr effect, or magnetic dichroism; anomalous Hall effect and anisotropic magnetoresistance (AMR) in metallic materials; etc. To describe these properties, one has to use either a full relativistic Hamiltonian, i.e., Dirac equation, or an effective Hamiltonian, i.e., Pauli equation, which includes magnetic interactions and spin-orbit coupling. However, it is not obvious to find all the different terms which have to be taken into account in the effective Hamiltonian. For this reason, it appears necessary to study the weak-relativistic limit of the Dirac equation for magnetic system in order to extract the complete expression of the effective Hamiltonian.

For many-body systems, the relativistic density-functional theory allows us to replace the many-body Dirac equation by the Kohn-Sham-Dirac equation, which has the form[3–5]

$$H^A = c\boldsymbol{\alpha}\cdot(\mathbf{p}-e\mathbf{A}_{eff}) + \beta mc^2 + V_{eff}, \tag{1}$$

where $\boldsymbol{\alpha}$ and β are the (4×4) Dirac matrices respectively related to the (2×2) Pauli matrix $\boldsymbol{\sigma}$ and unit matrix 1,

$$\boldsymbol{\alpha}\equiv\begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix}, \qquad \beta\equiv\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \tag{2}$$

The effective potential V_{eff} and the effective vector potential $\mathbf{A}_{eff}$ are functions of the external potential V_{ext}, of the external vector potential $\mathbf{A}_{ext}$, of the exchange-correlation energy functional E_{xc}, as well as the electron density $n(\mathbf{r}) \equiv \Sigma_{\varepsilon_i\leq\varepsilon_F}\psi_i^\dagger(\mathbf{r})\psi_i(\mathbf{r})$ and the current density $\mathbf{J}(\mathbf{r}) \equiv \Sigma_{\varepsilon_i\leq\varepsilon_F}\psi_i^\dagger(\mathbf{r})c\boldsymbol{\alpha}\psi_i(\mathbf{r})$ where $\psi_i(\mathbf{r})$ is the four-component Dirac spinor,

$$V_{eff}(\mathbf{r}) \equiv V_{ext}(\mathbf{r}) + \frac{e^2}{4\pi\epsilon_0}\int\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}' + \frac{\delta E_{xc}[n(\mathbf{r}),\mathbf{J}(\mathbf{r})]}{\delta n(\mathbf{r})}, \tag{3}$$

$$\mathbf{A}_{eff}(\mathbf{r}) \equiv \mathbf{A}_{ext}(\mathbf{r}) + \frac{e}{4\pi\epsilon_0 c^2}\int\frac{\mathbf{J}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}' + \frac{1}{e}\frac{\delta E_{xc}[n(\mathbf{r}),\mathbf{J}(\mathbf{r})]}{\delta\mathbf{J}(\mathbf{r})}, \tag{4}$$

where ϵ_0 is the vacuum permeability.

MacDonald and Vosko[6] have shown that by neglecting orbital magnetism, a Gordon decomposition[7,8] allows us to approximate Eq. (1) as

$$H^B = c(\boldsymbol{\alpha}\cdot\mathbf{p}) + \beta mc^2 + V_{eff} - \mu_B\beta(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff}). \tag{5}$$

The last term corresponds to the exchange coupling involving an effective magnetic field $\mathbf{B}_{eff} \equiv \boldsymbol{\nabla}\times\mathbf{A}_{eff} = \mathbf{B}_{ext} + \mathbf{B}_{xc}$ where $\mathbf{B}_{ext}$ is the external magnetic field and $\mathbf{B}_{xc}$ the exchange-correlation field given by

$$\mathbf{B}_{xc}(\mathbf{r}) \equiv \frac{1}{\mu_B}\frac{\delta E_{xc}[n(\mathbf{r}),\mathbf{m}(\mathbf{r})]}{\delta\mathbf{m}(\mathbf{r})}, \tag{6}$$

where $\mathbf{m}(\mathbf{r}) \equiv \Sigma_{\varepsilon_i\leq\varepsilon_F}\psi_i^\dagger(\mathbf{r})\beta\boldsymbol{\sigma}\psi_i(\mathbf{r})$ is the spin density and $\mu_B \equiv e\hbar/2m$. The presence of the β matrix in the last term of Eq. (5) and in the definition of $\mathbf{m}(\mathbf{r})$ results directly from the Gordon decomposition[7] where the expression of the four-component Dirac spinor is extracted by multiplying the Dirac equation by a β matrix either at the left side or at the right side.[9]

To describe relativistic effects in magnetic systems, one can either start from H^A or from H^B. Even if the second Hamiltonian is an approximate description in comparison to the former one, it has two advantages: on the first hand, it is simpler because the vector potential is not more present in the kinetic energy and, on the other hand, it has a more convenient form for magnetic systems because one can in-

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clude the magnetic interactions directly in the scalar potential which is thus a sum of spin-independent and spin-dependent parts. Therefore H^B is generally used.[10–17] However, there is no check on the consistency of Eqs. (1) and (5) in the weak-relativistic limit. Apart from the orbital magnetism which is clearly neglected in H^B, we should obtain the same relativistic corrections. We point out in the present paper that the weak relativistic limits of H^A and H^B differ. These differences have quite serious consequences: in particular, the magnetocrystalline anisotropy of a uniaxial ferromagnet and the AMR of a cubic ferromagnet are found from the approximate form H^B to be of order $1/c^2$, whereas the correct results obtained from the exact form H^A are of order $1/c^4$. We give a qualitative estimate of the order of magnitude of these spurious terms.

The paper is organized in the following way. In Sec. II, we present the derivation of a useful expression of the Dirac Green's function in term of the nonrelativistic Green's function. Then, using this Dirac Green's function as an alternative method to decouple particles from antiparticles, we extract in Sec. III the effective Hamiltonian and effective velocity operator. These operations are done starting both from H^A and H^B in order to compare the relativistic corrections of order $1/c^2$. The calculations starting from H^A are given in detail in the text whereas the calculations from H^B, which are similar in their principles but lead to different expressions, are summarized in the appendixes. Finally in Sec. IV, we discuss the results and present a qualitative evaluation of the difference between the two descriptions.

II. DIRAC GREEN'S FUNCTION

The derivation of the Dirac Green's function has been done both for H^A and H^B. Below, we present in detail the calculation for H^A while the calculation for H^B is summarized in Appendix A. In order to simplify the notations, we introduce $\boldsymbol{\pi}\equiv\mathbf{p}-e\mathbf{A}_{eff}$, and write simply V instead of V_{eff} and $\mathbf{A}$ instead of $\mathbf{A}_{eff}$. Since we are interested mainly in the electrons states in the weak relativistic limit, we shift the zero of energy by $-mc^2$; for positrons, we would have to shift the zero of energy by $+mc^2$. We express the Hamiltonian H^A in terms of (2×2) matrices as

$$H^A=\begin{pmatrix} V & c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) \\ c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) & V-2mc^2 \end{pmatrix}. \tag{7}$$

A series of algebraic manipulations allows to express the Dirac Green's function $G(z)\equiv(z-H^A)^{-1}$ in terms of the (2×2) nonrelativistic Green's function $\tilde{G}(z)\equiv(z-\tilde{H}^A)^{-1}$ associated with the (2×2) nonrelativistic Hamiltonian $\tilde{H}^A\equiv(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2/2m+V$. This kind of transformation was initially proposed by Gesztesy *et al.*[18] Following their work, we write first G as a product of matrices

$$\begin{aligned} G(z)&=\left[\begin{pmatrix} z & -c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) \\ -c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) & 2mc^2 \end{pmatrix}+\begin{pmatrix} -V & 0 \\ 0 & z-V \end{pmatrix}\right]^{-1} \\ &=\left[1+\mathcal{A}^{-1}(z)\begin{pmatrix} -V & 0 \\ 0 & z-V \end{pmatrix}\right]^{-1}\mathcal{A}^{-1}(z), \end{aligned} \tag{8}$$

where we have introduced

$$\mathcal{A}(z)=\begin{pmatrix} z & -c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) \\ -c(\boldsymbol{\sigma}\cdot\boldsymbol{\pi}) & 2mc^2 \end{pmatrix}. \tag{9}$$

The explicit inversion of $\mathcal{A}$ yields

$$\mathcal{A}^{-1}(z)=\begin{pmatrix} \tilde{G}_0(z) & \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z) \\ \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z) & \dfrac{z}{2mc^2}\tilde{G}_0(z) \end{pmatrix}, \tag{10}$$

where $\tilde{G}_0(z)\equiv[z-(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2/2m]^{-1}$. Using Eq. (10) in Eq. (8), we get

$$\begin{aligned} G(z)=&\begin{pmatrix} 1-\tilde{G}_0(z)V & \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z)(z-V) \\ -\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z)V & 1+\dfrac{z}{2mc^2}\tilde{G}_0(z)(z-V) \end{pmatrix}^{-1} \\ &\times\begin{pmatrix} \tilde{G}_0(z) & \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z) \\ \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}_0(z) & \dfrac{z}{2mc^2}\tilde{G}_0(z) \end{pmatrix}. \end{aligned} \tag{11}$$

The first factor in the right-hand side of the above equation is calculated by direct matrix inversion, and is equal to

$$\begin{pmatrix} \tilde{G}(z)\tilde{G}_0^{-1}(z)-\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)D(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}[\tilde{G}(z)\tilde{G}_0^{-1}(z)-1] & -\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)D(z) \\ D(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}[\tilde{G}(z)\tilde{G}_0^{-1}(z)-1] & D(z) \end{pmatrix}, \tag{12}$$

where we have used the relation $\tilde{G}_0V\tilde{G}=\tilde{G}-\tilde{G}_0$ and introduced the (2×2) matrix

$$D(z)=\left(1+Q(z)\frac{(z-V)}{2mc^2}\right)^{-1}, \tag{13}$$

with $Q(z)=1+(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})\tilde{G}(z)(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})/2m$. We insert Eq. (12) in Eq. (11) and finally obtain

$$G(z)=\begin{pmatrix} \tilde{G}(z)-\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)D(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}Q^{-1}(z)D(z)Q(z) \\ D(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & \dfrac{1}{2mc^2}D(z)Q(z) \end{pmatrix}. \tag{14}$$

The corresponding expression associated with H^B is given in Appendix A by Eq. (A3). The originality of our result is that we have succeeded in expressing the (4×4) Dirac Green's function entirely in terms of (2×2) matrices, in particular the nonrelativistic Green's function $\tilde{G}$, whereas in the result presented by Gesztesy *et al.*[18] or more recently by Brouder *et al.*,[19] a product of (4×4) matrices is still present in the final expression. Such formulation of the Dirac Green's function provides an alternative method to separate particles form antiparticles in the weak-relativistic limit by a simple block diagonalization. More comments and illustration of this method are presented in Sec. III.

In the limit of low electron energies, it is useful to have a semirelativistic expression. In order to get it, we perform an expansion of the operator D in powers of $1/c$ which allows us to write the Dirac Green's function as a series,

$$G(z)=\sum_{n=0}^{\infty} G^{(n)}(z), \tag{15}$$

where $G^{(n)}(z)$ is the term of order $1/c^n$. The successive terms are

$$G^{(0)}(z)=\begin{pmatrix} \tilde{G}(z) & 0 \\ 0 & 0 \end{pmatrix}, \tag{16}$$

$$G^{(2k+1)}(z)=\begin{pmatrix} 0 & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\left[-\dfrac{(z-V)}{2mc^2}Q(z)\right]^k \\ \left[-Q(z)\dfrac{(z-V)}{2mc^2}\right]^k\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & 0 \end{pmatrix}, \tag{17}$$

$$G^{(2k+2)}(z)=\begin{pmatrix} -\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)\left[-Q(z)\dfrac{(z-V)}{2mc^2}\right]^k\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & 0 \\ 0 & \dfrac{1}{2mc^2}\left[-Q(z)\dfrac{(z-V)}{2mc^2}\right]^k Q(z) \end{pmatrix}, \tag{18}$$

for $k\geqslant0$. We remark that odd terms in the expansion of the Green's function in powers of $1/c$ are odd matrices whereas even terms are even matrices. Corresponding expressions for the Dirac Green's function expansion associated with H^B are given in Appendix A.

One advantage of the expression (15) is that we can directly identify and calculate the terms which gives rise to particular effect according to the order of this effect with $1/c$. Consider for example the magnetic anisotropies. In the case of a system with uniaxial anisotropy, the anisotropy energy is quadratic in the spin-orbit coupling λ_{so} (i.e., of order $1/c^4$). As the anisotropy energy is linear with respect to the Green's function, we have to consider the term $G^{(4)}$. For cubic anisotropy, the anisotropy energy is of order λ_{so}^4 (i.e., of order $1/c^8$), then we have to consider the term $G^{(8)}$. For galvanomagnetic and magneto-optical effects, one needs to calculate the conductivity tensor. The latter is expressed as a product of two Green's functions and two velocity operators, which are $c\boldsymbol{\alpha}$ in the relativistic theory. For the effects which are linear in spin-orbit coupling, (i.e., of order $1/c^2$), such as the anomalous Hall effect, and the Kerr and Faraday magneto-optical effects, one needs to calculate all terms up to $G^{(4)}$. For the effects that are quadratic in spin-orbit coupling (i.e., of order $1/c^4$), such as the AMR or the magnetic birefringence, one needs all terms up to $G^{(6)}$. Note that the usual (i.e., nonrelativistic) conductivity is obtained from the terms up to $G^{(2)}$.

III. WEAK-RELATIVISTIC LIMIT

A. Effective Hamiltonian

We turn now our attention to the effective Hamiltonian in the weak-relativistic limit. Different methods have been pro-

posed to obtain it: (i) the direct elimination of the lower components of the wave function;[20] (ii) the Foldy-Wouthuysen transformation which requires a succession of canonical transformations[21] and (iii) the expansion of the Dirac resolvent around its nonrelativistic limit.[18,22] A detailed comparison between these methods is beyond the purpose of this paper (we refer to, e.g., Ref. 23 and references therein). As we have the explicit expression of the Dirac Green's function in term of (2×2) matrices, we follow the last method but instead of using projection operators as it is done in Ref. 18, we extract the effective Hamiltonian from a block diagonalization and a Dyson-type expansion of the Dirac Green's function.

A block diagonalization of the Dirac Green's function allows to cancels the terms which couple the upper and lower components and then to extract the effective Hamiltonian. Let us start with the nonrelativistic limit $(c\rightarrow\infty)$ which is obtained in a transparent manner. Indeed, in this limit, only the upper left matrix element of $G=G^{(0)}$ is different from zero [see Eq. (16) for H^A or Eq. (A6) for H^B]. Then the separation between particles and antiparticles is naturally made: the Green's function which describes the particles is directly given by $\tilde{G}$ and the effective Hamiltonian is $\tilde{H}^A$ (respectively $\tilde{H}^B$). In the general case (arbitrary value of c), the decoupling of particles and antiparticles can be exactly made only for free electrons. For particles in a potential, like in our case, an expansion in powers of $1/c$ has to be performed which means a restriction to the weak-relativistic limit. Indeed, the block diagonalization of the Green's function is only possible if we cut the expansion (15) [respectively, Eq. (A3)]. Such limitation is a common characteristic for all methods.[3] Below, we present the calculation starting from H^A whereas the calculation starting from H^B is done in Appendix B. Since we restrict our calculations to the lowest order relativistic corrections to the Hamiltonian, we have to cut the expansion (15) of the Green's function after the second order in $1/c$:

$$G(z)\approx\begin{pmatrix}\tilde{G}(z)-\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\\ \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z) & \dfrac{1}{2mc^2}Q(z)\end{pmatrix}. \tag{19}$$

The block diagonalization of G corresponds to the change of basis $M^{-1}GM$ where the (4×4) unitary matrix M is given by

$$M=\left(1+\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{4m^2c^2}\right)^{-1/2}\begin{pmatrix}1 & -\dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\\ \dfrac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc} & 1\end{pmatrix}, \tag{20}$$

and leads to the block diagonal Green's function

$$M^{-1}G(z)M=\begin{pmatrix}g_+(z) & 0\\ 0 & g_-(z)\end{pmatrix}. \tag{21}$$

By means of this transformation, we have achieved a decoupling between the particles and the antiparticules: the (2×2) matrix g_+ describes the particles whereas the (2×2) matrix g_- describes the antiparticles. We get

$$g_+(z)=\tilde{G}(z)+\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{8m^2c^2}\tilde{G}(z)+\tilde{G}(z)\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{8m^2c^2}-\tilde{G}(z)\frac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}(z-V)\frac{\boldsymbol{\sigma}\cdot\boldsymbol{\pi}}{2mc}\tilde{G}(z), \tag{22}$$

and

$$g_-(z)=\frac{1}{2mc^2}. \tag{23}$$

From now, we restrict our study to the particles. Using the relations $\tilde{G}z=1+\tilde{G}\tilde{H}^A=1+\tilde{H}^A\tilde{G}$, we can transform the last term in Eq. (22) and write g_+ as a first-order perturbation expansion $\tilde{G}+\tilde{G}H^A_{rc}\tilde{G}$ where

$$H^A_{rc}=-\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^4}{8m^3c^2}+\frac{1}{4m^2c^2}(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})V(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})-\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{8m^2c^2}V-V\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{8m^2c^2}. \tag{24}$$

This expression corresponds to the relativistic corrections of order $1/c^2$ to the nonrelativistic Hamiltonian $\tilde{H}^A$. Thus the effective Hamiltonian, i.e., Pauli Hamiltonian, is $H^A_{eff}=\tilde{H}^A+H^A_{rc}$. This result is exactly similar to that given in the literature in the case of a single-particle Dirac equation (see, e.g., Refs. 18 and 24). In order to get a more familiar expression of H^A_{rc}, we have to perform some transformations which are detailed in Appendix B. In the case of a uniform effective magnetic field and neglecting orbital magnetism, H^A_{rc} reduces to [see Eq. (B6) in Appendix B]

$$H^A_{rc}=-\frac{p^4}{8m^3c^2}+\frac{\hbar^2}{8m^2c^2}\Delta V+\frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\boldsymbol{\pi})+\frac{\mu_B}{2m^2c^2}p^2(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff}). \tag{25}$$

We obtain the usual relativistic corrections (relativistic mass correction, Darwin term, and spin-orbit coupling) plus an additional contribution due to the presence of the exchange coupling. For a nonuniform effective magnetic field, which is the case in realistic problems, further relativistic corrections are obtained [see Eq. (B4)].

Similar calculations, presented in Appendix C, have been done starting from H^B by performing a block diagonalization of the Dirac Green's function calculated in Appendix A. We limit the comparison of the relativistic corrections H^A_{rc} and H^B_{rc} obtained in the two descriptions to the case of a uniform

effective magnetic field. Comparing Eqs. (25) and (C7), we observe two differences: one in the spin-orbit coupling because the nonrelativistic velocity $\tilde{\mathbf{v}}$ is equal to $\boldsymbol{\pi}/m$ in the first description whereas it is equal to $\mathbf{p}/m$ in the second description; and an other one in the relativistic corrections H_{rxc} to the exchange coupling, given by the last term in Eqs. (25) and (C7), respectively. Actually, from Eq. (25) we have

$$H_{rxc}^{A} \equiv \frac{\mu_B}{2m^2c^2} p^2(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff}), \tag{26}$$

whereas from Eq. (C7) we have

$$H_{rxc}^{B} \equiv \frac{\mu_B}{2m^2c^2}(\boldsymbol{\sigma}\cdot\mathbf{p})(\mathbf{p}\cdot\mathbf{B}_{eff}). \tag{27}$$

What is problematic is that H_{rxc}^{A} and H_{rxc}^{B} couple the spin and momentum in a quite different manner. Comments and consequences of this difference are presented in Sec. IV.

B. Effective velocity operator

To complete this study, we want to comment briefly on the velocity operator which is by definition

$$\mathbf{v} = \frac{1}{i\hbar}[\mathbf{r},H] = \frac{1}{i\hbar}(\mathbf{r}H - H\mathbf{r}) = \frac{\partial H}{\partial \mathbf{p}}, \tag{28}$$

where $\mathbf{r}$ is the position operator. When we insert the expression of the Hamiltonians H^A or H^B, we get the simple form

$$\mathbf{v} = c\,\boldsymbol{\alpha} = \begin{pmatrix} 0 & c\boldsymbol{\sigma} \\ c\boldsymbol{\sigma} & 0 \end{pmatrix}. \tag{29}$$

The effective velocity operator can be obtained from Eq. (29) by the change of basis $M^{-1}\mathbf{v}M$ but in order to get the corrections of order $1/c^2$ to the velocity, it would be necessary to expand M up to the order $1/c^4$ which is cumbersome. It can also be obtained from H_{eff}^{A} using

$$\mathbf{v}_{eff}^{A} = \frac{1}{i\hbar}[\mathbf{r},H_{eff}^{A}] = \frac{1}{i\hbar}[\mathbf{r},\tilde{H}^{A}] + \frac{1}{i\hbar}[\mathbf{r},H_{rc}^{A}] \equiv \tilde{\mathbf{v}}^{A} + \mathbf{v}_{rc}^{A}, \tag{30}$$

where $\tilde{\mathbf{v}}^{A}$ is the nonrelativistic velocity and $\mathbf{v}_{rc}^{A}$ the relativistic corrections of order $1/c^2$ to the velocity. From the expression of $\tilde{H}^{A}$, we get $\tilde{\mathbf{v}}^{A} = \boldsymbol{\pi}/m$ and from Eq. (B4) where diamagnetic terms are neglected, we get

$$\begin{aligned}\mathbf{v}_{rc}^{A} = &-\frac{p^2\mathbf{p}}{2m^3c^2} + \frac{\hbar}{4m^2c^2}(\boldsymbol{\sigma}\times\boldsymbol{\nabla}V) + \frac{\mu_B}{2m^2c^2}[\mathbf{p}(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff}) \\ &+ (\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})\mathbf{p}] + \frac{e}{4m^3c^2}[p^2\mathbf{A} + \mathbf{A}p^2 + \mathbf{p}(\mathbf{p}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{p}) \\ &+ (\mathbf{p}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{p})\mathbf{p}].\end{aligned} \tag{31}$$

In the case of a uniform effective magnetic field and in absence of orbital magnetism, it reduces to

$$\mathbf{v}_{rc}^{A} = -\frac{p^2\mathbf{p}}{2m^3c^2} + \frac{\hbar}{4m^2c^2}(\boldsymbol{\sigma}\times\boldsymbol{\nabla}V) + \frac{\mu_B}{m^2c^2}\mathbf{p}(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff}). \tag{32}$$

The first term is the contribution which comes from the relativistic mass correction. The second term, the so-called anomalous velocity, results from the spin-orbit coupling and can play an important role, for example, it leads to the side-jump mechanism in the anomalous Hall effect. The last term is due to the presence of the exchange coupling and has no specific name. Its symmetry is also different in comparison to the relativistic corrections to the velocity operator obtained starting from H^B [see Eq. (D2) in Appendix D].

IV. DISCUSSION

This study gives some clarification concerning the assumptions made when one replaces the Hamiltonian H^A by the Hamiltonian H^B. Even if the consequences of such approximation are not fully known, as Kübler justly notices,[25] it was generally believed that the transformation from H^A to H^B neglects only orbital magnetism effects. However, the calculations of the weak-relativistic limits of H^A and H^B made in Sec. III reveal an additional difference which corresponds to a different symmetry of the relativistic corrections to the exchange coupling: whereas H_{rxc}^{A} [see Eq. (26)] is isotropic with respect to the direction of the momentum, H_{rxc}^{B} [see Eq. (27)] is anisotropic because its amplitude depends, through the scalar product $(\mathbf{p}\cdot\mathbf{B}_{eff})$, on the angle between the directions of the momentum and the effective magnetic field. Thus the use of H^B can lead to anisotropic effects, such as the magnetocrystalline anisotropy or the AMR, which differ from those obtained from the exact Kohn-Sham-Dirac Hamiltonian H^A.

Let us first consider the magnetocrystalline anisotropy of a uniaxial system (e.g., a material with an hexagonal lattice, or an ultrathin film). If we start from H^A, the magnetic anisotropy arises only as a second-order perturbation due to the spin-orbit coupling, so that it is of order $1/c^4$. In contrast, if we use H^B, it is easy to see that the relativistic correction of the exchange interaction, H_{rxc}^{B}, gives rise to an additional contribution to the magnetocrystalline anisotropy, already in the first order of perturbation, i.e., of order $1/c^2$, which is unphysical.

Although the details are somewhat more complicated, a similar result is obtained when considering the AMR of a cubic system: starting from H^A, the AMR arises as a second-order perturbation due to the spin-orbit coupling, i.e., it is of order $1/c^4$, whereas starting from H^B, an additional AMR term of order $1/c^2$ arises as a first-order perturbation due to H_{rxc}^{B}.

In the limit of $c\to\infty$, these spurious terms of order $1/c^2$ dominate over the correct terms of order $1/c^4$, which is of course unacceptable. As all *ab initio* calculations of magnetocrystalline anisotropy[11–14] starting for the Dirac equation rely on the approximation (5), their validity can be questioned *a priori*. Let us make a simple estimate of the orders of magnitude for the physical value of c. Starting from H^A,

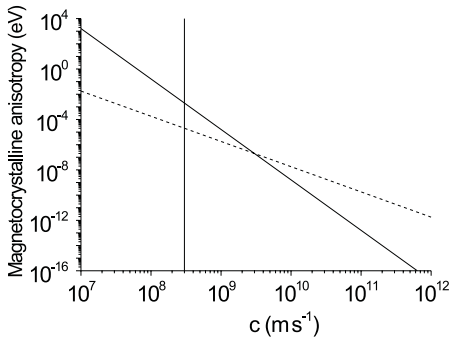

FIG. 1. This figure shows the typical variation of the magnetocrystalline anisotropy of a uniaxial ferromagnet as a function of c. The physical value of c is indicated by the vertical line. The solid curve is the correct value K^A obtained from H^A, whereas the dashed line represents the spurious contribution K^B obtained from H^B.

the magnetocrystalline anisotropy per atom is[26]

$$K^A \approx \frac{\lambda_{so}^2}{W} \sim \frac{1}{c^4}, \tag{33}$$

where λ_{so} is the spin-orbit constant and W the bandwidth. Starting from H^B, on the other hand, it is quite easy to calculate the magnetocrystalline anisotropy due to H^B_{rxc},

$$K^B \approx \frac{\varepsilon_F \Delta_{ex}}{2mc^2}, \tag{34}$$

where ε_F is the Fermi level and Δ_{ex} the exchange splitting. For a transition-metal ferromagnet, by taking typical values $\lambda_{so} \approx 0.1$ eV and $W \approx 5$ eV, one obtains $K^A \approx 2\times 10^{-3}$ eV. Taking $\varepsilon_F \approx 10$ eV, $\Delta_{ex} \approx 2$ eV, and $mc^2 \approx 500$ keV, we obtain $K^B \approx 2\times 10^{-5}$ eV. Therefore, in spite of the fact that $K^B \gg K^A$ in the limit $c \to \infty$, we find that $K^B \ll K^A$ for the the physical value $c \approx 3\times 10^8$ m s^{-1}. This result is visualized in Fig. 1. Then, the quantitative results of first-principles calculations of the magnetocrystalline anisotropy based on the approximate Kohn-Sham-Dirac Hamiltonian H^B should not be perturbed in a significant manner by the spurious contribution of order $1/c^2$. In spite of this fortunate circumstance, it would be desirable to develop a more satisfactory theoretical approach which is free from unphysical spurious contributions.

For a nonuniform effective magnetic field, additional relativistic corrections are obtained [see Eqs. (B4] and (C6)). The qualitative importance of these corrections remains to be investigated.

To conclude, we want to underline two aspects which represent the important results of this work. On the one hand, we have derived a convenient form of the Dirac Green's function which is valid for any value of c and has the advantage to be express in terms of (2×2) matrices, in particular the nonrelativistic Green's function. This is a quite general form which can be used to treat different problems. We have applied it as an alternative way to separate particles from antiparticles in order to extract the effective Hamiltonian and effective velocity in the weak-relativistic limit. On the other hand, we have performed a detailed comparison in the weak-relativistic limit between the Kohn-Sham-Dirac equation and its approximate form containing the exchange coupling. This study has revealed a difference of symmetry in the relativistic corrections to the exchange coupling which leads to artificial anisotropic effects. A qualitative estimation has shown that, in the case of transition metals, this difference is not significant.

APPENDIX A: DIRAC GREEN'S FUNCTION IN PRESENCE OF AN EXCHANGE COUPLING

In this appendix, we summarize the derivation of the Dirac Green's function starting from H^B given by Eq. (5). It follows the same steps as starting from H^A but involves different matrices and leads to a different expression of the final Dirac Green's function. To simplify the notations, we introduce $\Theta = -\mu_B(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})$, then

$$H^B = \begin{pmatrix} V+\Theta & c(\boldsymbol{\sigma}\cdot\mathbf{p}) \\ c(\boldsymbol{\sigma}\cdot\mathbf{p}) & V-2mc^2-\Theta \end{pmatrix}. \tag{A1}$$

The Dirac Green's function $G(z) \equiv (z-H^B)^{-1}$ can then be written as

$$G(z) = \left[1 + \mathcal{A}^{-1}(z)\begin{pmatrix} -V-\Theta & 0 \\ 0 & z-V+\Theta \end{pmatrix}\right]^{-1} \mathcal{A}^{-1}(z). \tag{A2}$$

$\mathcal{A}^{-1}$ is given by Eq. (10) where we replace $\boldsymbol{\pi}$ by $\mathbf{p}$ and define $\tilde{G}_0(z) \equiv (z-p^2/2m)^{-1}$. We perform the direct inversion of the matrices which appear in Eq. (A2). It leads to the final expression:

$$G(z) = \begin{pmatrix} \tilde{G}(z) - \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}(z-V+\Theta)D(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}Q^{-1}(z)D(z)Q(z) \\ D(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & \dfrac{1}{2mc^2}D(z)Q(z) \end{pmatrix}, \tag{A3}$$

where $\tilde{G}$ is the (2×2) nonrelativistic Green's function associated with the (2×2) nonrelativistic Hamiltonian $\tilde{H}^B = p^2/2m + V + \Theta$, and the operators D and Q are given by

$$D(z)=\left(1+Q(z)\frac{(z-V+\Theta)}{2mc^2}\right)^{-1}, \tag{A4}$$

$$Q(z)=1+\frac{(\boldsymbol{\sigma}\cdot\mathbf{p})\tilde{G}(z)(\boldsymbol{\sigma}\cdot\mathbf{p})}{2m}. \tag{A5}$$

A semirelativistic expansion $G(z)=\Sigma_{n=0}^{\infty}G^{(n)}(z)$ can also be given. The successive terms are

$$G^{(0)}(z)=\begin{pmatrix}\tilde{G}(z) & 0\\ 0 & 0\end{pmatrix}, \tag{A6}$$

$$G^{(2k+1)}(z)=\begin{pmatrix}0 & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\left[-\dfrac{(z-V+\Theta)}{2mc^2}Q(z)\right]^k\\ \left(-Q(z)\dfrac{(z-V+\Theta)}{2mc^2}\right)^k\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & 0\end{pmatrix}, \tag{A7}$$

$$G^{(2k+2)}(z)=\begin{pmatrix}-\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}(z-V+\Theta)\left[-Q(z)\dfrac{(z-V+\Theta)}{2mc^2}\right]^k\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & 0\\ 0 & \dfrac{1}{2mc^2}\left[-Q(z)\dfrac{(z-V+\Theta)}{2mc^2}\right]^k Q(z)\end{pmatrix}, \tag{A8}$$

for $k\geqslant 0$.

APPENDIX B: EFFECTIVE HAMILTONIAN IN PRESENCE OF A POTENTIAL VECTOR

Transformations of $\tilde{H}^A$ and H_{rc}^A allow to get a more familiar expression of the effective Hamiltonian H_{eff}^A. Neglecting diamagnetic terms, we have

$$\begin{aligned}\tilde{H}^A&=\frac{(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})^2}{2m}+V\\ &=\frac{p^2}{2m}+V-\mu_B(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})-\frac{e}{2m}(\mathbf{p}\cdot\mathbf{A}+\mathbf{A}\cdot\mathbf{p}),\end{aligned} \tag{B1}$$

where we have used the identities

$$(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})=\boldsymbol{\pi}\cdot\boldsymbol{\pi}+i\boldsymbol{\sigma}\cdot(\boldsymbol{\pi}\times\boldsymbol{\pi}),$$

$$\mathbf{p}\times\mathbf{A}+\mathbf{A}\times\mathbf{p}=-i\hbar\mathbf{B}_{eff}. \tag{B2}$$

For a uniform effective field, $\tilde{H}^A$ reduces to

$$\tilde{H}^A=\frac{p^2}{2m}+V-\mu_B[(\boldsymbol{\sigma}+\mathbf{L})\cdot\mathbf{B}_{eff}], \tag{B3}$$

where $\mathbf{L}=\mathbf{r}\times\mathbf{p}$ is the orbital momentum. In absence of orbital magnetism, this last expression is identical to $\tilde{H}^B$. We turn now our attention to the relativistic corrections H_{rc}^A. After transformation of Eq. (24) and neglecting the diamagnetic terms, we can rewrite H_{rc}^A as

$$\begin{aligned}H_{rc}^A=&-\frac{p^4}{8m^3c^2}+\frac{\hbar^2}{8m^2c^2}\Delta V+\frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\boldsymbol{\pi})\\ &+\frac{\mu_B}{4m^2c^2}[p^2(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})+(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})p^2]\\ &+\frac{e}{8m^2c^2}\left(\frac{p^2}{m}(\mathbf{p}\cdot\mathbf{A}+\mathbf{A}\cdot\mathbf{p})+(\mathbf{p}\cdot\mathbf{A}+\mathbf{A}\cdot\mathbf{p})\frac{p^2}{m}\right.\\ &\left.+(\mathbf{A}\cdot\mathbf{p}-\mathbf{p}\cdot\mathbf{A})V+V(\mathbf{p}\cdot\mathbf{A}-\mathbf{A}\cdot\mathbf{p})\right),\end{aligned} \tag{B4}$$

where we have used the following identities:

$$(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})V(\boldsymbol{\sigma}\cdot\boldsymbol{\pi})=\boldsymbol{\pi}V\cdot\boldsymbol{\pi}+i\boldsymbol{\sigma}\cdot(\boldsymbol{\pi}V\times\boldsymbol{\pi}),$$

$$2\mathbf{p}V\cdot\mathbf{p}=p^2V+Vp^2+\hbar^2\Delta V, \tag{B5}$$

and replaced the momentum operator $\mathbf{p}$ as well as the operator p^2, where they act only on the potential, respectively by the gradient $\boldsymbol{\nabla}=-\mathbf{p}/i\hbar$ and by the Laplacian $\Delta=-p^2/\hbar^2$. In the case of a uniform effective magnetic field, H_{rc}^A reduces to

$$\begin{aligned}H_{rc}^A=&-\frac{p^4}{8m^3c^2}+\frac{\hbar^2}{8m^2c^2}\Delta V+\frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\boldsymbol{\pi})\\ &+\frac{\mu_B}{2m^2c^2}p^2[(\boldsymbol{\sigma}+\mathbf{L})\cdot\mathbf{B}_{eff}].\end{aligned} \tag{B6}$$

The hermiticity of Eq. (B4) is manifest and the hermiticity of Eq. (B6) is assured by the fact that p^2 and $\mathbf{B}_{eff}$ commute in the case of a uniform effective magnetic field.

APPENDIX C: EFFECTIVE HAMILTONIAN IN PRESENCE OF AN EXCHANGE COUPLING

In this appendix, we summarize the derivation of the effective Hamiltonian starting from the Dirac Green's function (A3). We cut the expansion after the second order with $1/c$, then

$$G(z) \approx \begin{pmatrix} \tilde{G}(z)-\tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}(z-V+\Theta)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & \tilde{G}(z)\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc} \\ \dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z) & \dfrac{1}{2mc^2}Q(z) \end{pmatrix}. \tag{C1}$$

The block diagonalization $M^{-1}GM$ is obtained with

$$M=\left(1+\frac{p^2}{4m^2c^2}\right)^{-1/2}\begin{pmatrix} 1 & -\dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc} \\ \dfrac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc} & 1 \end{pmatrix}, \tag{C2}$$

and leads to

$$g_+(z)=\tilde{G}(z)+\frac{p^2}{8m^2c^2}\tilde{G}(z)+\tilde{G}(z)\frac{p^2}{8m^2c^2} - \tilde{G}(z)\frac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}(z-V+\Theta)\frac{\boldsymbol{\sigma}\cdot\mathbf{p}}{2mc}\tilde{G}(z), \tag{C3}$$

which can be written as $\tilde{G}+\tilde{G}H^B_{rc}\tilde{G}$ where

$$H^B_{rc}=-\frac{p^4}{8m^3c^2}+\frac{1}{4m^2c^2}(\boldsymbol{\sigma}\cdot\mathbf{p})(V-\Theta)(\boldsymbol{\sigma}\cdot\mathbf{p}) - \frac{p^2}{8m^2c^2}(V+\Theta)-(V+\Theta)\frac{p^2}{8m^2c^2}. \tag{C4}$$

In order to get a more usual expression of H^B_{rc}, we made some transformations of Eq. (C4) using identities (B5) where we have replaced $\boldsymbol{\pi}$ by $\mathbf{p}$ and the relation

$$(\boldsymbol{\sigma}\cdot\mathbf{p})(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})(\boldsymbol{\sigma}\cdot\mathbf{p})=-\mathbf{p}(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})\cdot\mathbf{p}+\hbar(\boldsymbol{\nabla}\times\mathbf{B}_{eff})\cdot\mathbf{p} + (\boldsymbol{\sigma}\cdot\mathbf{p})(\mathbf{B}_{eff}\cdot\mathbf{p})+(\mathbf{p}\cdot\mathbf{B}_{eff})(\boldsymbol{\sigma}\cdot\mathbf{p}). \tag{C5}$$

Finally, we get

$$H^B_{rc}=-\frac{p^4}{8m^3c^2}+\frac{\hbar^2}{8m^2c^2}\Delta[V-\mu_B(\boldsymbol{\sigma}\cdot\mathbf{B}_{eff})] + \frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\mathbf{p})+\frac{\hbar\mu_B}{4m^2c^2}(\boldsymbol{\nabla}\times\mathbf{B}_{eff})\cdot\mathbf{p} + \frac{\mu_B}{4m^2c^2}[(\boldsymbol{\sigma}\cdot\mathbf{p})(\mathbf{B}_{eff}\cdot\mathbf{p})+(\mathbf{p}\cdot\mathbf{B}_{eff})(\boldsymbol{\sigma}\cdot\mathbf{p})]. \tag{C6}$$

In the case of a uniform effective magnetic field, it reduces to

$$H^B_{rc}=-\frac{p^4}{8m^3c^2}+\frac{\hbar^2}{8m^2c^2}\Delta V+\frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\mathbf{p}) + \frac{\mu_B}{2m^2c^2}(\boldsymbol{\sigma}\cdot\mathbf{p})(\mathbf{B}_{eff}\cdot\mathbf{p}). \tag{C7}$$

The hermiticity of Eqs. (C4) and (C6) is manifest and the hermiticity of Eq. (C7) is assured by the fact that $\mathbf{p}$ and $\mathbf{B}_{eff}$ commute in the case of a uniform effective magnetic field.

APPENDIX D: EFFECTIVE VELOCITY OPERATOR IN PRESENCE OF AN EXCHANGE COUPLING

In this appendix, we summarize the derivation of the effective velocity starting from H^B. From the expression of $\tilde{H}^B=p^2/2m+V+\Theta$, we get $\tilde{\mathbf{v}}^B=\mathbf{p}/m$ and from the expression (C6) of H^B_{rc}, we get

$$\mathbf{v}^B_{rc}=-\frac{p^2\mathbf{p}}{2m^3c^2}+\frac{\hbar}{4m^2c^2}(\boldsymbol{\sigma}\times\boldsymbol{\nabla}V)+\frac{\hbar\mu_B}{4m^2c^2}(\boldsymbol{\nabla}\times\mathbf{B}_{eff}) + \frac{\mu_B}{4m^2c^2}[\boldsymbol{\sigma}(\mathbf{B}_{eff}\cdot\mathbf{p})+(\mathbf{p}\cdot\mathbf{B}_{eff})\boldsymbol{\sigma}+(\boldsymbol{\sigma}\cdot\mathbf{p})\mathbf{B}_{eff} + \mathbf{B}_{eff}(\boldsymbol{\sigma}\cdot\mathbf{p})]. \tag{D1}$$

This expression differs from the result obtained by Kraft *et al.*[15] on two points: they get a wrong coefficient (a factor 2 missing) for the contribution of the relativistic mass correction and they obtain a contribution from the Darwin term which should not appear because the commutator $[\mathbf{r},\Delta V]$ is equal to zero. In the case of a uniform effective magnetic field, Eq. (D1) reduces to

$$\mathbf{v}_{rc}^{B}=-\frac{p^2\mathbf{p}}{2m^3c^2}+\frac{\hbar}{4m^2c^2}(\boldsymbol{\sigma}\times\boldsymbol{\nabla}V)+\frac{\mu_B}{2m^2c^2}[\boldsymbol{\sigma}(\mathbf{B}_{eff}\cdot\mathbf{p})+\mathbf{B}_{eff}(\boldsymbol{\sigma}\cdot\mathbf{p})]. \tag{D2}$$

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[1] *The Effects of Relativity in Atoms, Molecules, and the Solid State*, edited by S. Wilson, I.P. Grant, and B.L. Gyorffy (Plenum, New York, 1991).

[2] *Spin-Orbit-Influenced Spectroscopies of Magnetic Solids*, edited by H. Ebert and G. Schütz (Springer-Verlag, Berlin and Heidelberg, 1996).

[3] P. Strange, *Relativistic Quantum Mechanics* (Cambridge University Press, Cambridge, England, 1998).

[4] H. Eschrig, *The Fundamentals of Density Functional Theory* (B.G. Teubner Verlagsgesellschaft Stuttgart-Leipzig, 1996), p. 159; H. Eschrig, G. Seifert, and P. Ziesche, Solid State Commun. **59**, 777 (1985).

[5] R.M. Dreizler and E.K.U. Gross, *Density Functional Theory* (Springer-Verlag, Berlin and Heidelberg, 1990).

[6] A.H. MacDonald and S.H. Vosko, J. Phys. C **12**, 2977 (1979).

[7] W. Gordon, Z. Phys. **50**, 630 (1928).

[8] A.K. Rajagopal and J. Callaway, Phys. Rev. B **7**, 1912 (1973).

[9] A formulation without the β factor in the exchange coupling: $H'^{B}=c(\boldsymbol{\alpha}\cdot\mathbf{p})+\beta mc^2+V_{eff}-\mu_B(\boldsymbol{\sigma}\cdot\mathbf{B}'_{eff})$ where $\mathbf{B}'_{eff}\equiv(1/\mu_B)\delta E_{xc}[n(\mathbf{r}),\mathbf{m}'(\mathbf{r})]/\delta\mathbf{m}'(\mathbf{r})$ and $\mathbf{m}'(\mathbf{r})\equiv\Sigma_{\varepsilon_i\leqslant\varepsilon_F}\psi_i^{\dagger}(\mathbf{r})\boldsymbol{\sigma}\psi_i(\mathbf{r})$, was later proposed by M.V. Ramana and A.K. Rajagopal, J. Phys. C **14**, 4291 (1981), but without any explicit derivation. It can be shown that like H^B (see the discussion in Sec. IV), H'^{B} yields to relativistic corrections to the exchange coupling which give an unphysical effect. However, we do not present the details of this calculation as to our knowledge the Hamiltonian H'^{B} has no justification.

[10] P. Strange, J. Staunton, and B.L. Gyorffy, J. Phys. C **17**, 3355 (1984).

[11] H. Ebert, Phys. Rev. B **38**, 9390 (1988).

[12] L. Szunyogh, B. Újfalussy, and P. Weinberger, Phys. Rev. B **51**, 9552 (1995); B. Újfalussy, L. Szunyogh, P. Bruno, and P. Weinberger, Phys. Rev. Lett. **77**, 1805 (1996); L. Szunyogh, B. Újfalussy, C. Blaas, U. Pustogowa, C. Sommers, and P. Weinberger, Phys. Rev. B **56**, 14 036 (1997).

[13] S.S.A. Razee, J.B. Staunton, and F.J. Pinski, Phys. Rev. B **56**, 8082 (1997).

[14] S.V. Beiden, W.M. Temmerman, Z. Szotek, G.A. Gehring, G.M. Stocks, Y. Wang, D.M.C. Nicholson, W.A. Shelton, and H. Ebert, Phys. Rev. B **57**, 14 247 (1998).

[15] T. Kraft, P.M. Oppeneer, V.N. Antonov, and H. Eschrig, Phys. Rev. B **52**, 3561 (1995)

[16] H. Ebert, Rep. Prog. Phys. **59**, 1665 (1996).

[17] H. Ebert, A. Vernes, and J. Banhart, Phys. Rev. B **54**, 8479 (1996).

[18] F. Gesztesy, H. Grosse, and B. Thaller, Ann. I.H.P. Phys. Theor. **40**, 159 (1984); F. Gesztesy, B. Thaller, and H. Grosse, Phys. Rev. Lett. **50**, 625 (1983).

[19] C. Brouder, M. Alouani, and K.H. Bennemann, Phys. Rev. B **54**, 7334 (1996); C. Brouder, in *Magnetism and Synchrotron Radiation, Mittelwihr 1996*, edited by E. Beaurepaire, B. Carrière, and J-P. Kappler (Les Editions de Physique, Les Ulis, 1997), pp. 55-64.

[20] W. Pauli, Z. Phys. **43**, 601 (1927).

[21] L.L. Foldy and S.A. Wouthuysen, Phys. Rev. **78**, 29 (1950).

[22] B. Thaller, *The Dirac Equation* (Springer, Berlin, 1992).

[23] D.R. Grigore, G. Nenciu, and R. Purice, Ann. I.H.P. Phys. Theor. **51**, 231 (1989).

[24] A. Messiah, *Quantum Mechanics* (North-Holland Amsterdam, 1962), Vol. 2, p. 946.

[25] J. Kübler, *Theory of Itinerant Electron Magnetism* (Clarendon Press, Oxford, 2000).

[26] P. Bruno, in *Magnetismus von Festkörpern und Grenzflächen*, edited by P.H. Dederichs, P. Grünberg, and W. Zinn (24. IFF-Ferienkurs, Forschungszentrum Jülich, 1993), pp. 24.1-24.28.