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 non-relativistic Green’s function is first derived and then used to calculate the effective Hamiltonian, i.e. Pauli Hamiltonian, and effective velocity 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 then 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: magnetic 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 to replace the many-body Dirac equation by the Kohn-Sham-Dirac equation, which has an exact expression of the Dirac Green’s function.

$V_{\text{ext}}$ corresponds to the exchange coupling involving an effective magnetic field $B_{\text{eff}} = B_{\text{ext}} + B_{\text{xc}}$ where $B_{\text{ext}}$ is the external field and $B_{\text{xc}}$ the exchange-correlation field given by

The relativistic density functional theory is used to find the 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 non-relativistic Green’s function is first derived and then used to calculate the effective Hamiltonian, i.e. Pauli Hamiltonian, and effective velocity 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 then give a qualitative estimate of the order of magnitude of these spurious terms.

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For many-body systems, the relativistic density functional theory allows to replace the many-body Dirac equation by the Kohn-Sham-Dirac equation, which has the form

$$
H^A = c \alpha \cdot (p - e A_{\text{eff}}) + \beta mc^2 + V_{\text{eff}},
$$

where $\alpha$ and $\beta$ are the $(4 \times 4)$ Dirac matrices respectively related to the $(2 \times 2)$ Pauli matrix $\sigma$ and unit matrix

$$
\alpha \equiv \begin{pmatrix} 0 & \sigma \\ \sigma & 0 \end{pmatrix}, \quad \beta \equiv \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.
$$

The effective potential $V_{\text{eff}}$ and effective vector potential $A_{\text{eff}}$ are functions of external and internal fields as well as electron density $n(r) = \sum_{i=1}^{N} \psi_i^+(r) \psi_i(r)$ and current density $J(r) = \sum_{i=1}^{N} \psi_i^+(r) c \alpha \psi_i(r)$:

$$
V_{\text{eff}}(r) \equiv V_{\text{ext}}(r) + \int \frac{n(r')}{|r - r'|} d^3r' + \frac{\delta E_{\text{xc}}[n(r), J(r)]}{\delta n(r)},
$$

$$
A_{\text{eff}}(r) \equiv A_{\text{ext}}(r) + \frac{e^2}{4\pi \epsilon_0 c} \int \frac{J(r')}{|r - r'|} d^3r' + \frac{1}{c} \frac{\delta E_{\text{xc}}[n(r), J(r)]}{\delta J(r)}.
$$

where $V_{\text{ext}}$ and $A_{\text{ext}}$ are the external potential and vector potential and $E_{\text{xc}}$ is the exchange-correlation energy functional.

Neglecting orbital currents, a Gordon decomposition allows to rewrite $H^A$ as

$$
H^B = c (\alpha \cdot p) + \beta mc^2 + V_{\text{eff}} - \mu_B \beta (\sigma \cdot B_{\text{eff}}).
$$

The last term corresponds to the exchange coupling involving an effective magnetic field $B_{\text{eff}} = B_{\text{ext}} + B_{\text{xc}}$

$$
B_{\text{xc}}(r) = \frac{1}{\mu_B} \frac{\delta E_{\text{xc}}[n(r), m(r)]}{\delta m(r)}.
$$

where $m(r) = \sum_{i=1}^{N} \psi_i^+(r) \beta \sigma \psi_i(r)$ is the spin density and $\mu_B \equiv e\hbar/2m$. Expression for the Hamiltonian can also be derived and justified with the help of symmetry arguments.

To describe relativistic effects in magnetic systems, one could 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 system because we can include the magnetic interactions directly in the scalar potential which is then a sum of spin-independent and spin-dependent parts. Therefore, $H^B$ is generally used. However, there is no check on
the consistency of these two equations 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 an 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 then 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 terms of the non-relativistic Green’s function. Then, using this Dirac Green’s function as an alternative method to separate particles from anti-particles, we extract in Sec. III the effective Hamiltonian and effective velocity. 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 summarize in the Appendices. 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 summarize in the Appendix A. In order to simplify the notations, we introduce \( \pi \equiv p - eA_{\text{eff}} \), and write simply \( V \) instead of \( V_{\text{eff}} \). 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 \((2 \times 2)\) matrices as:

\[
H^A = \begin{pmatrix}
V & c(\sigma \cdot \pi) \\
c(\sigma \cdot \pi) & V - 2mc^2
\end{pmatrix},
\]

A series of algebraic manipulations allows to express the Dirac Green’s function \( G(z) \equiv (z - H^A)^{-1} \) in terms of the \((2 \times 2)\) non-relativistic Green’s function \( \tilde{G}(z) \equiv (z - H^A)^{-1} \) associated with the \((2 \times 2)\) non-relativistic Hamiltonian \( \tilde{H}^A \equiv (\sigma \cdot \pi)^2/2m + V \). We write first \( G \) as a product of matrices:

\[
G(z) = \left( \begin{array}{cccc}
z & -c(\sigma \cdot \pi) \\
-c(\sigma \cdot \pi) & 2mc^2
\end{array} \right)
+ \left( \begin{array}{c}
-V \\
0
\end{array} \right) (z - V)^{-1}
\]

\[
= \left( 1 + \mathcal{A}^{-1}(z) \left( \begin{array}{cc}
-V & 0 \\
0 & z - V
\end{array} \right) \right)^{-1} \mathcal{A}^{-1}(z),
\]

where we have introduced:

\[
\mathcal{A}(z) = \left( \begin{array}{cccc}
z & -c(\sigma \cdot \pi) \\
-c(\sigma \cdot \pi) & 2mc^2
\end{array} \right).
\]

The explicit inversion of \( \mathcal{A} \) yields

\[
\mathcal{A}^{-1}(z) = \left( \begin{array}{cccc}
\tilde{G}_0(z) & \frac{\sigma \cdot \pi}{2mc} \tilde{G}_0(z) \\
\frac{\sigma \cdot \pi}{2mc} \tilde{G}_0(z) & \frac{\sigma \cdot \pi}{2mc} \tilde{G}_0(z)
\end{array} \right),
\]

where \( \tilde{G}_0(z) \equiv (z - (\sigma \cdot \pi)^2/2m)^{-1} \). Using (10) in (9), we get

\[
G(z) = \left( 1 - \tilde{G}_0(z)V - \frac{\sigma \cdot \pi}{2mc} \tilde{G}_0(z)(z - V) \right)^{-1}
\times \left( \frac{\sigma \cdot \pi}{2mc} \tilde{G}_0(z) \right).
\]

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

\[
\left( \tilde{G}(z)\tilde{G}_0^{-1}(z) - \tilde{G}(z)\tilde{G}_0^{-1}(z)D(z)\sigma \cdot \pi \tilde{G}(z)\tilde{G}_0^{-1}(z) - \tilde{G}(z)\sigma \cdot \pi \tilde{G}(z)(z - V)D(z) \right)_{D(z)},
\]

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

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

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

\[
G(z) = \left( \tilde{G}(z) - \tilde{G}(z)\tilde{G}_0^{-1}(z)D(z)\sigma \cdot \pi \tilde{G}(z) \tilde{G}_0^{-1}(z)D(z)Q(z) \right).
\]
The corresponding expression associated with $H^B$ is given in the Appendix A by Eq. (A3). We have thus succeeded in expressing the $(4 \times 4)$ Dirac Green’s function in terms of $(2 \times 2)$ matrices, in particular the non-relativistic Green’s function $\tilde{G}$. This formulation provides an alternative method to separate particles from anti-particles in the weak-relativistic limit by a simple block diagonalization. More comments and illustration of this method are presented in Sec. III.

In absence of vector potential (which means $\pi = p$ in Eq. (4) or $\Theta = 0$ in Eq. (A3)), the method and result are similar to those presented earlier by Gesztesy et al. and more recently by Broder et al. for the non-magnetic case, although the form we give here is simpler than the one given by these authors. The generalization to magnetic cases suggested in Refs. 18,19 is not correct because the authors, who have started from $H^B$, have omitted the factor $\beta$ in the expression of the exchange coupling $-\mu_B \beta (\sigma \cdot B \lambda)$.

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

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

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

\begin{align}
G^{(0)}(z) &= \begin{pmatrix} \tilde{G}(z) & 0 \\ 0 & 0 \end{pmatrix}, \\
G^{(2k+1)}(z) &= \begin{pmatrix} 0 & \tilde{G}(z) \frac{\sigma \cdot \pi}{4mc^2} \left( \frac{z-v}{2mc^2} Q(z) \right)^k \\ \left(-Q(z) \frac{z-v}{2mc^2}\right)^k \tilde{G}(z) & 0 \end{pmatrix}, \\
G^{(2k+2)}(z) &= \begin{pmatrix} \tilde{G}(z) \frac{\sigma \cdot \pi}{4mc^2} (z-V) \left(-Q(z) \frac{z-v}{2mc^2}\right)^k & 0 \\ 0 & \frac{1}{2mc^2} \left(-Q(z) \frac{z-v}{2mc^2}\right)^k Q(z) \end{pmatrix},
\end{align}

for $k \geq 0$. 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 spin-orbit coupling $\lambda_{so}$ (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 $\lambda_{so}^3$ (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 $c\alpha$ 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 magnetooptical 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., non-relativistic) 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 which can be obtained through different ways such as for example the elimination of the lower components of the wave function or the Foldy-Wouthuysen transformation which requires a succession of canonical transformations. As we have the explicit expression of the Dirac Green’s function in term of $(2 \times 2)$ matrices, we do not need to used such methods to separate the particles from the antiparticles. Actually, a block diagonalization of (14) allows to cancel the terms which couple the upper and lower components and then to extract the effective Hamiltonian. Let us start with the non-relativistic limit ($c \to \infty$) which is obtained in a transparent manner. Indeed, in this limit, the matrix elements of $G = G^{(0)}(z)$ are different from zero only in the left upper part (see Eq. (14)). Then the separation between particles and antiparticles is naturally made: the Green’s function which described the particle are directly done by
and the effective Hamiltonian is \( \tilde{H}^A \). In the general case (arbitrary value of \( c \)), the separation between particles and antiparticles can be exactly made only for free electrons. For particles in a potential, like in our case, a development 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 after the second order with \( 1/c \): 

\[
G(z) \approx \left( \tilde{G}(z) - \tilde{G}(z) \frac{\sigma \cdot \pi}{2mc} \tilde{G}(z) \frac{\sigma \cdot \pi}{2mc} \tilde{G}(z) \frac{1}{2mc} \right). 
\]

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

\[
M = \left( 1 + \frac{(\sigma \cdot \pi)^2}{4m^2c^2} \right)^{-\frac{1}{2}} \begin{pmatrix} 1 & -\frac{\sigma \cdot \pi}{2mc} \\ \frac{\sigma \cdot \pi}{2mc} & 1 \end{pmatrix},
\]

and leads to the block diagonal Green’s function:

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

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

\[
g_+(z) = \tilde{G}(z) + \frac{(\sigma \cdot \pi)^2}{8m^2c^2} \tilde{G}(z) + \tilde{G}(z) \frac{\sigma \cdot \pi}{8m^2c^2} \tilde{G}(z),
\]

\[
-\tilde{G}(z) \frac{\sigma \cdot \pi}{2mc} \tilde{G}(z) - V \frac{\sigma \cdot \pi}{2mc} \tilde{G}(z),
\]

and

\[
g_-(z) = \frac{1}{2mc^2}.
\]

Since now, we restrict our study to the particles. Using the relations \( \tilde{G}z = 1 + \tilde{G}H^A = 1 + \tilde{H}^A \tilde{G} \), we can transform the last term in (22) and write \( g_+ \) under the form \( \tilde{G} + \tilde{H}^A_{rc} \tilde{G} \) where:

\[
H^A_{rc} = -\frac{(\sigma \cdot \pi)^4}{8m^4c^2} + \frac{1}{4m^2c^2}(\sigma \cdot \pi)V(\sigma \cdot \pi)
\]

\[
-\frac{(\sigma \cdot \pi)^2}{8m^2c^2} \tilde{V}(\sigma \cdot \pi)^2. \]

This expression corresponds to the relativistic corrections of order \( 1/c^2 \) to the non-relativistic Hamiltonian \( H^A \). Thus, the effective Hamiltonian, i.e. Pauli Hamiltonian, is \( H^A_{eff} = H^A + H^A_{rc} \) (it can also be derived from a block diagonalization of \( H^A \)). This result is exactly similar to that given in the literature in the case of a single particle Dirac equation (see for example Ref. [21]). In order to get a more usual expression of \( H^A_{rc} \), we have to perform some transformations which are detailed in Appendix B.

In the case of an uniform effective magnetic field and neglecting orbital magnetism, \( H^A_{rc} \) reduces to (see Eq. (B6) in Appendix B):

\[
H^A_{rc} = -\frac{\mu_B}{2m^2c^2}p^2(\sigma \cdot \mathbf{B}_{eff}),
\]

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 non-uniform 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 can now compare the relativistic corrections \( H^A_{rc} \) and \( H^B_{rc} \) obtained in the two descriptions in the case of a uniform effective magnetic field. Comparing (25) and (C7), we observe two differences: one in the spin-orbit coupling because the non-relativistic velocity \( \tilde{v} \) is equal to \( \pi/m \) in the first description whereas it is equal to \( \mathbf{p}/m \) in the second description; and another one in the relativistic corrections \( H^A_{rc} \) to the exchange coupling, given by the last term in Eqs. (25) and (C7). Actually, from (25) we have:

\[
H^A_{rc} = \frac{\mu_B}{2m^2c^2}p^2(\sigma \cdot \mathbf{B}_{eff}),
\]

whereas from (C7) we have:

\[
H^B_{rc} = \frac{\mu_B}{2m^2c^2}(\sigma \cdot \mathbf{p})(\mathbf{p} \cdot \mathbf{B}_{eff}).
\]

What is problematic is that \( H^A_{rc} \) and \( H^B_{rc} \) couple the spin and momentum in a quite different manner. Comments and consequences of this difference are presented in the section IV.

**B. Effective velocity operator**

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

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

where \( \mathbf{r} \) is the position operator. When we report the expression of the Hamiltonians \( H^A \) or \( H^B \), we get the simple form:
\[ \mathbf{v} = c \alpha = \begin{pmatrix} 0 & c \sigma \\ c \sigma & 0 \end{pmatrix}. \]  

(29)

The effective velocity 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 form \( H^A_{\text{eff}} \) using:

\[
\mathbf{v}^A_{\text{eff}} = \frac{1}{i \hbar} [\mathbf{r}, \mathbf{H}^A_{\text{eff}}] = \frac{1}{i \hbar} [\mathbf{r}, \tilde{\mathbf{H}}^A] + \frac{1}{i \hbar} [\mathbf{r}, \mathbf{H}^A_{\text{rec}}] = \tilde{\mathbf{v}}^A + \mathbf{v}^A_{\text{rec}},
\]

(30)

where \( \tilde{\mathbf{v}}^A \) is the non-relativistic velocity and \( \mathbf{v}^A_{\text{rec}} \) the relativistic corrections of order \( 1/c^2 \) to the velocity. From the expression of \( \tilde{\mathbf{H}}^A \), we get \( \tilde{\mathbf{v}}^A = \pi/m \) and from (B4) where diamagnetic terms are neglected, we get:

\[
\mathbf{v}^A_{\text{rec}} = -\frac{\mu}{2m^2c^2} + \frac{\hbar}{4m^2c^2} (\sigma \times \nabla V) + \frac{\mu_B}{2m^2c^2} (\mathbf{p} \cdot \mathbf{B}_{\text{eff}}) + (\sigma \cdot 
\]

\[
\mathbf{B}_{\text{eff}} \mathbf{p}) + \frac{e}{4m^2c^2} \mathbf{p}(\mathbf{A} + \mathbf{A} \cdot \mathbf{p}) + (\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p}) \mathbf{p}.
\]

(31)

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

\[
\mathbf{v}^A_{\text{rec}} = -\frac{\mu}{2m^2c^2} + \frac{\hbar}{4m^2c^2} (\sigma \times \nabla V) + \frac{\mu_B}{m^2c^2} (\mathbf{p} \cdot \mathbf{B}_{\text{eff}}).
\]

(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 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 Kühler justly notices, 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 \( H^A_{\text{rec}} \) (see Eq. (26)) is isotropic with respect to the direction of the momentum, \( H^B_{\text{rec}} \) (see Eq. (27)) is anisotropic because its amplitude depends, through the scalar product \( (\mathbf{p}, \mathbf{B}_{\text{eff}}) \), 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, \( H^B_{\text{rec}} \), 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 \( H^B_{\text{rec}} \).

![FIG. 1.](image-url) 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 \( H^B \). 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 \( \text{ab initio} \) calculations of magnetocrystalline anisotropy starting for the Dirac equation rely on the approximation (3), there validity can be questioned \textit{a priori}. Let us make a simple estimate of the orders of magnitude for the physical value of \( c \). Starting from \( H^A \), the magnetocrystalline anisotropy is \( \vdots \).
where $\lambda_{so}$ is the spin-orbit constant and $W$ the bandwidth. Starting from $H^B$ one hand, it is quite easy to calculate the magnetocrystalline anisotropy due to $H_{exc}^B$:

$$K^B \approx \frac{\varepsilon_F \Delta_{ex}}{2mc^2}. \quad (34)$$

where $\varepsilon_F$ is the Fermi level and $\Delta_{ex}$ 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 $c \rightarrow \infty$, we find that $K^B \ll K^A$ for the physical value $c \approx 3 \times 10^8$ m.s$^{-1}$. This result is visualized in Fig. 1. Therefore the quantitative results of first-principles calculations of the magnetocrystalline anisotropy is not 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.

**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 (5). It follows the same steps than 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 $\Theta = -\mu_B (\mathbf{\sigma} \cdot \mathbf{B}_{eff})$, then:

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

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

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

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

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

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

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

A semi-relativistic expansion $G(z) = \sum_{n=0}^{\infty} G^{(n)}(z)$ can also be given. The successive terms are
\[ G^{(0)}(z) = \begin{pmatrix} \tilde{G}(z) & 0 \\ 0 & 0 \end{pmatrix}, \]
\[ G^{(2k+1)}(z) = \begin{pmatrix} 0 & \tilde{G}(z) \frac{\sigma \cdot \mathbf{p}}{2mc} \left( -\frac{z - V + \Theta}{2mc^2} \right)^k \\ -\tilde{G}(z) \frac{\sigma \cdot \mathbf{p}}{2mc} \left( -\frac{z - V + \Theta}{2mc^2} \right)^k & 0 \end{pmatrix}, \]
\[ G^{(2k+2)}(z) = \begin{pmatrix} -\tilde{G}(z) \frac{\sigma \cdot \mathbf{p}}{2mc} (z - V + \Theta) \left( -\frac{z - V + \Theta}{2mc^2} \right)^k & \tilde{G}(z) \frac{\sigma \cdot \mathbf{p}}{2mc} \left( -\frac{z - V + \Theta}{2mc^2} \right)^k \\ 0 & \frac{1}{2mc^2} \left( -\frac{z - V + \Theta}{2mc^2} \right)^k \tilde{Q}(z) \end{pmatrix}, \]
for \( k \geq 0 \).

**APPENDIX B: EFFECTIVE HAMILTONIAN IN PRESENCE OF A POTENTIAL VECTOR**

Transformations of \( \tilde{H}^A \) and \( H^A_{rc} \) allow to get a more usual expression of the effective Hamiltonian \( H^A_{eff} \). Neglecting diamagnetic terms, we have:

\[ \tilde{H}^A = \frac{(\sigma \cdot \pi)^2}{2m} + V = \frac{p^2}{2m} + V - \mu_B (\sigma \cdot \mathbf{B}_{eff}) \]
\[ -\frac{e}{2m} (\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p}), \]
where we have used the identities:

\[ (\sigma \cdot \pi) (\sigma \cdot \pi) = \pi \cdot \pi + i \sigma \cdot (\pi \times \pi), \]
\[ \mathbf{p} \times \mathbf{A} + \mathbf{A} \times \mathbf{p} = -i \hbar \mathbf{B}_{eff}. \]

For a uniform effective field, \( \tilde{H}^A \) reduces to:

\[ \tilde{H}^A = \frac{p^2}{2m} + V - \mu_B ((\sigma + \mathbf{L}) \cdot \mathbf{B}_{eff}), \]
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 \( H^A_{rc} \). After transformation of (24) and neglecting the diamagnetic terms, we can rewrite \( H^A_{rc} \) as:

\[ H^A_{rc} = -\frac{p^4}{8m^3c^2} + \frac{\hbar^2}{8m^2c^2} \Delta V + \frac{\hbar}{4m^2c^2} \sigma \cdot (\nabla V \times \pi) \]
\[ + \frac{\mu_B}{4m^2c^2} \left( \frac{p^2}{m} (\sigma \cdot \mathbf{B}_{eff}) + (\sigma \cdot \mathbf{B}_{eff}) p^2 \right), \]
\[ + \frac{\mu_B}{8m^2c^2} \left( \frac{p^2}{m} (\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p}) + (\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p}) \frac{p^2}{m} \right) \]
\[ + (\mathbf{A} \cdot \mathbf{p} - \mathbf{p} \cdot \mathbf{A}) V - V (\mathbf{A} \cdot \mathbf{p} - \mathbf{p} \cdot \mathbf{A}), \]
where we have used the following identities:

\[ (\sigma \cdot \pi) V (\sigma \cdot \pi) = \pi V \cdot \pi + i \sigma \cdot (\pi V \times \pi), \]
\[ 2p \mathbf{V} \cdot \mathbf{p} = p^2 V + V p^2 + \hbar^2 \Delta V, \]
and replaced the momentum operator \( \mathbf{p} \) as well as the operator \( p^2 \), when they act only on the potential, respectively by the gradient \( \nabla = -\mathbf{p}/i\hbar \) and by the Laplacian \( \Delta = -p^2/\hbar^2 \). In the case of a uniform effective magnetic field, \( H^A_{rc} \) reduces to:

\[ H^A_{rc} = \frac{-p^4}{8m^3c^2} + \frac{\hbar^2}{8m^2c^2} \Delta V + \frac{\hbar}{4m^2c^2} \sigma \cdot (\nabla V \times \pi) \]
\[ + \frac{\mu_B}{2m^2c^2} \left( \frac{p^2}{m} (\sigma \cdot \mathbf{L}) + (\sigma \cdot \mathbf{L}) \mathbf{B}_{eff} \right). \]

**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 \( 1/\hbar \), then:

\[ G(z) \approx \left( \tilde{G}(z) - \tilde{G}(z) \frac{\sigma \cdot \mathbf{p}}{2mc} (z - V + \Theta) \frac{\sigma \cdot \mathbf{p}}{2mc} \tilde{G}(z) \right) \frac{1}{2mc^2} \tilde{Q}(z). \]
The block diagonalization $M^{-1}GM$ is obtained with:

$$M = \left(1 + \frac{p^2}{4m^2c^2} \right)^{-\frac{1}{2}} \left(\frac{1}{2mc} - \frac{\sigma \cdot p}{2mc} \frac{1}{1}\right),$$  

(C2)

and leads to:

$$g_+ = \tilde{G}(z) + \frac{p^2}{8m^2c^2} \tilde{G}(z) + \tilde{G}(z) \frac{p^2}{8m^2c^2}$$

$$- \tilde{G}(z) \frac{\sigma \cdot P}{2mc} (V + \Theta) \frac{\sigma \cdot P}{2mc} \tilde{G}(z),$$  

(C3)

which can be written under the form $\tilde{G} + \tilde{G}H_{rc}B\tilde{G}$ where:

$$H_{rc}B = -\frac{p^4}{8m^3c^2} + \frac{1}{4m^2c^2} (\sigma \cdot p) (V - \Theta) (\sigma \cdot p)$$

$$- \frac{p^2}{8m^2c^2} (V + \Theta) - (V + \Theta) \frac{p^2}{8m^2c^2}. $$  

(C4)

In order to get a more usual expression of $H_{rc}B$, we made some transformations of (C4) using identities (B5) where we have replaced $\pi$ by $p$ and the relation:

$$(\sigma \cdot p) (\sigma \cdot B_{eff}) (\sigma \cdot p) =$$

$$- p (\sigma \cdot B_{eff}) \cdot p + \hbar (\nabla \times B_{eff}) \cdot p$$

$$+ (\sigma \cdot p) (B_{eff} \cdot p) + (p \cdot B_{eff}) (\sigma \cdot p).  $$  

(C5)

Finally, we get:

$$H_{rc}B = -\frac{p^4}{8m^3c^2} + \frac{\hbar^2}{8m^2c^2} \Delta (V - \mu_B (\sigma \cdot B_{eff}))$$

$$+ \frac{\hbar}{4m^2c^2} \sigma \cdot (\nabla V \times p) + \frac{\hbar \mu_B}{4m^2c^2} (\nabla \times B_{eff}) \cdot p$$

$$+ \frac{\mu_B}{4m^2c^2} \left(\sigma \cdot p)(B_{eff} \cdot p) + (p \cdot B_{eff})(\sigma \cdot p)\right).  $$  

(C6)

This expression of the effective Hamiltonian in presence of the exchange coupling is equivalent to the one obtained by Kraft et al.[4] In the case of a uniform effective magnetic field, it reduces to:

$$H_{rc}B = -\frac{p^4}{8m^3c^2} + \frac{\hbar^2}{8m^2c^2} \Delta V + \frac{\hbar \mu_B}{4m^2c^2} \sigma \cdot (\nabla V \times p)$$

$$+ \frac{\mu_B}{2m^2c^2} (\sigma \cdot p)(B_{eff} \cdot p). $$  

(C7)

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 $H^B = p^2/2m + V + \Theta$, we get $\nabla^B = p/m$ and from the expression (C6) of $H_{rc}B$, we get:

$$v^B_{rc} = -\frac{p^2}{2m^2c^2} \sigma \cdot \nabla V + \frac{\hbar \mu_B}{4m^2c^2} (\nabla \times B_{eff})$$

$$+ \frac{\mu_B}{4m^2c^2} \left(\sigma (B_{eff} \cdot p) + (p \cdot B_{eff}) \sigma\right)$$

$$+ (\sigma \cdot p)B_{eff} + B_{eff} (\sigma \cdot p). $$  

(D1)

This expression differs from the result obtained by Kraft et al.[4] 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 [r, $\Delta V$] is equal to zero. In the case of a uniform effective magnetic field, (D1) reduces to:

$$v^B_{rc} = -\frac{p^2}{2m^2c^2} + \frac{\hbar}{4m^2c^2} \left(\sigma \times \nabla V\right)$$

$$+ \frac{\mu_B}{2m^2c^2} \left(\sigma (B_{eff} \cdot p) + B_{eff} (\sigma \cdot p)\right). $$  

(D2)

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