

**First-principles relativistic study of spin waves in thin magnetic films**L. Udvardi,<sup>1,2</sup> L. Szunyogh,<sup>1,2</sup> K. Palotás,<sup>2</sup> and P. Weinberger<sup>2</sup><sup>1</sup>*Department of Theoretical Physics and Center for Applied Mathematics and Computational Physics, Budapest University of Technology and Economics, Budafoki út 8, H-1111, Budapest, Hungary*<sup>2</sup>*Center for Computational Materials Science, Technical University Vienna, Gumpendorfer Str. 1a, A-1060 Vienna, Austria*

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In order to study spin-wave excitations of itinerant ferromagnets a relativistic first-principles method based on the adiabatic approach is presented. The derivatives of the free energy up to second order with respect of the polar and azimuthal angles are derived within the framework of the magnetic force theorem and the fully relativistic Korringa-Kohn-Rostoker method. Exchange and spin-orbit coupling are thus incorporated on equal footing in the Hamiltonian. Furthermore, a detailed comparison to classical spin Hamiltonians is given and it is shown that the magnetocrystalline anisotropy energy contains contributions from both the on-site anisotropy and the off-site exchange coupling terms. The method is applied to an Fe monolayer on Cu(001) and Au(001) surfaces and for a Co monolayer on Cu(001). The calculations provide with the gap at zero wave number due to the spin-orbit coupling and uniaxial anisotropy energies in good agreement with the results of the band energy difference method. It is pointed out that the terms in the spin-wave Hamiltonian related to the mixed partial derivatives of the free energy, absent within a nonrelativistic description, introduce an asymmetry in the magnon spectrum with respect to two in-plane easy axes. Moreover, in the case of an in-plane magnetized system the long-wavelength magnons are elliptically polarized due to the difference of the second-order uniaxial and fourth-order in-plane magnetic anisotropy.

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**I. INTRODUCTION**

In recent years the magnetic properties of thin films have attracted much interest both experimentally as well as theoretically mainly due to possible applications in magnetic storage technology. In thin magnetic films relativistic effects such as magnetic anisotropy are more pronounced than in bulk materials. The spin-orbit coupling plays a key role in low-energy magnetic excitations of such materials as it opens a gap at zero wave number which in turn is necessary for the formation of long-range magnetic order. First-principles calculations of the spin-wave excitations can help a lot in understanding the details of the underlying physics and in tailoring the properties of different ultrathin magnetic films.

Spin-wave excitation spectra can be determined by calculating the dynamical spin susceptibilities using linear response theory<sup>1</sup> or by using the adiabatic approach.<sup>2-6</sup> Within the adiabatic approach the fast motion of itinerant electrons is decoupled from the slow motion of the spins. Obviously, the condition for the validity of the adiabatic approximation is that the time scale of the precession of the magnetic moments should be sufficiently larger than the characteristic (hopping) time of the motion of electrons. This condition corresponds to the criterion that the spin-wave energy must be small as compared to the bandwidth and to the exchange splitting. It has been shown that in terms of the rigid-spin approximation; i.e., by choosing a uniquely defined spin-quantization axis within each cell, the general equation of motion for the spin density reduces to the semiclassical Landau-Lifshitz equation,<sup>2,6</sup> describing the orientational motion of the spin moments, whereas the magnitude of the spin magnetization can be evaluated self-consistently within the framework of a constrained density functional theory.<sup>7</sup> A non-self-consistent theory such as employed in the present

work allows one to trace the orientational motion of the magnetic moments, describing thus transverse spin-wave excitations only. It should be mentioned that a conceptually clear description of spin dynamics within a time-dependent density functional theory is matter of ongoing investigation<sup>8</sup> and yet is not in the state of being used in practical terms.

In the nonrelativistic case the adiabatic approach leads within the harmonic approximation to a classical Heisenberg Hamiltonian,

$$\mathcal{H} = \frac{1}{2} \sum_{i \neq j} J_{ij} \boldsymbol{\sigma}_i \boldsymbol{\sigma}_j, \quad (1)$$

where  $\boldsymbol{\sigma}_i$  is a classical unit vector parallel to the magnetization at site  $i$  and  $J_{ij}$  is the exchange interaction energy between sites  $i$  and  $j$ . Basically two methods are used to determine the exchange interaction energy  $J_{ij}$ . One of these is the so-called frozen magnon approximation,<sup>4,9</sup> in which the spin configuration is constrained to a spin wave of wave vector  $\mathbf{q}$  and the energy of this spin wave is calculated by employing the generalized Bloch theorem for a spin-spiral configuration.<sup>10</sup> Since within the particular model described by Eq. (1) the energy of a spin wave with respect to a ferromagnetic ground state can be expressed as

$$\varepsilon(\mathbf{q}) = \sum_{j \neq 0} J_{0j} (e^{i\mathbf{q} \cdot \mathbf{R}_{0j}} - 1), \quad (2)$$

where  $R_{ij}$  is the relative position vector connecting sites  $i$  and  $j$ , and  $q$  is the wave vector of the spin wave, the exchange coupling parameters  $J_{ij}$  are obtained from the inverse Fourier transform of the spin-wave dispersion law.

In the second approach which is frequently referred to as the torque method or the method of infinitesimal rotations,<sup>11</sup>

the coupling constants  $J_{ij}$  are calculated directly from the change of the energy associated with constrained rotations of the spin-polarization axes at sites  $i$  and  $j$ . Using the magnetic force theorem<sup>11,12</sup> the change of the total energy is approximated by the corresponding change of the one-particle energies, remarkably simplifying the calculations. A renormalization of this approach was recently proposed by Bruno<sup>13</sup> and Antropov<sup>14</sup> which considerably improved the calculated spectra in the region of shorter wavelengths.

For magnetic systems with noncubic symmetry such as hcp Co or for layered systems the importance of relativistic effects is enhanced. In a relativistic formalism the effective Hamiltonian, Eq. (1), no longer applies since the invariance of the Hamiltonian against a global rotation of the spins is lost. So far, when calculating spin-wave spectra, relativistic effects such as the spin-orbit coupling and the magnetic dipole-dipole interaction were taken into account as additive terms in the Hamiltonian (1), while the exchange coupling parameters  $J_{ij}$  were calculated nonrelativistically or were estimated from experiments.<sup>15–19</sup>

The aim of the present paper is to provide with a relativistic description of spin waves in itinerant, transition-metal ferromagnets from first principles. Our method relies on the adiabatic decoupling of the local spin-density functional theory and the rigid-spin approximation, whereby a fully relativistic treatment of the electrons in terms of the Kohn-Sham-Dirac equation is used. In the next section we describe the main points of our theory based on the Landau-Lifshitz equation and a harmonic approximation to the free energy. It will be shown that due to the relativistic treatment the spin-wave Hamiltonian contains off-diagonal elements. Closed formulas for the derivatives of the free energy up to second order with respect of the polar and azimuthal angles are derived by means of the fully relativistic Korringa-Kohn-Rostoker (KKR) method.<sup>20</sup> The relationship of our theory to classical spin Hamiltonians will also be given. Applications are then presented to typical ultrathin magnetic films with out-of-plane, Fe<sub>1</sub>Au(001), and in-plane, Co<sub>1</sub>Cu(001), ground-state magnetic orientation. In the case of Fe<sub>1</sub>Cu(001) the effect of the magnetic dipole-dipole interaction to the spin-wave spectrum is emphasized.

## II. THEORY

### A. General formalism

As shown in Refs. 2 and 6, within the rigid-spin approximation the adiabatic dynamics of local spin moments is described by the Landau-Lifshitz equation

$$M_{ri} \dot{\boldsymbol{\sigma}}_{ri} = -\frac{2\mu_B}{\hbar} \frac{\delta \mathcal{F}}{\delta \boldsymbol{\sigma}_{ri}} \times \boldsymbol{\sigma}_{ri}, \quad (3)$$

where  $M_{ri}$  is the magnitude of the spin moment and  $\boldsymbol{\sigma}_{ri}$  is a unit vector pointing along the spin-quantization axis in the atomic cell at the site  $i$  of layer  $r$ ,

$$\boldsymbol{\sigma}_{ri} = (\sin \vartheta_{ri} \cos \varphi_{ri}, \sin \vartheta_{ri} \sin \varphi_{ri}, \cos \vartheta_{ri}), \quad (4)$$

with the polar and azimuthal angles  $\vartheta_{ri}$  and  $\varphi_{ri}$ , respectively, and  $\mathcal{F}$  is the free energy of the system. For the case of

transverse magnons (see Introduction), the angles  $\vartheta_{ri}$  and  $\varphi_{ri}$  depend on time, whereas, by supposing two-dimensional translational invariance for the ground state, the time-independent magnitudes  $M_{ri}$  depend only on the layer index; i.e.,  $M_{ri} = M_r$  for all sites  $i$  in a particular layer  $r$ .

Rewriting Eq. (3) into spherical coordinates, the equations of motion for the angles  $\vartheta_{ri}$  and  $\varphi_{ri}$  are given by

$$M_r \dot{\varphi}_{ri} \sin \vartheta_{ri} = \frac{2\mu_B}{\hbar} \frac{\partial \mathcal{F}}{\partial \vartheta_{ri}}, \quad (5)$$

$$-M_r \dot{\vartheta}_{ri} \sin \vartheta_{ri} = \frac{2\mu_B}{\hbar} \frac{\partial \mathcal{F}}{\partial \varphi_{ri}}. \quad (6)$$

Choosing the polar ( $z$ ) axis of the reference system to be perpendicular to the magnetization in the ferromagnetic ground state, Eqs. (5) and (6) can easily be linearized,

$$M_r \dot{\varphi}_{ri} = \frac{2\mu_B}{\hbar} \frac{\partial \mathcal{F}}{\partial \vartheta_{ri}} \Big|_{\vartheta=\pi/2, \varphi=0}, \quad (7)$$

$$-M_r \dot{\vartheta}_{ri} = \frac{2\mu_B}{\hbar} \frac{\partial \mathcal{F}}{\partial \varphi_{ri}} \Big|_{\vartheta=\pi/2, \varphi=0}, \quad (8)$$

where the constraint  $\vartheta = \pi/2$ ,  $\varphi = 0$  denotes that the partial derivatives have to be taken at  $\vartheta_{ri} = \pi/2$ , and  $\varphi_{ri} = 0$  for all  $r$  and  $i$ . The linearized version of the Landau-Lifshitz equations, Eqs. (7) and (8), are the canonical equations for the generalized coordinates  $q_{ri} \equiv (M_r/\mu_B)^{1/2} \varphi_{ri}$  and momenta  $p_{ri} \equiv (M_r/\mu_B)^{1/2} \vartheta_{ri}$ . Adopting the harmonic approximation—i.e., expanding the free energy up to second order in the angular variables—the corresponding Hamilton function can be written as

$$\mathcal{H} = \frac{1}{\hbar} \sum_{ri,sj} (q_{ri} A_{ri,sj} q_{sj} + q_{ri} B_{ri,sj} p_{sj} + p_{ri} B_{sj,ri} q_{sj} + p_{ri} C_{ri,sj} p_{sj}), \quad (9)$$

with

$$A_{ri,sj} = (M_r/\mu_B)^{-1/2} \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \varphi_{sj}} \Big|_{\vartheta=\pi/2, \varphi=0} (M_s/\mu_B)^{-1/2}, \quad (10)$$

$$B_{ri,sj} = (M_r/\mu_B)^{-1/2} \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \vartheta_{sj}} \Big|_{\vartheta=\pi/2, \varphi=0} (M_s/\mu_B)^{-1/2}, \quad (11)$$

$$C_{ri,sj} = (M_r/\mu_B)^{-1/2} \frac{\partial^2 \mathcal{F}}{\partial \vartheta_{ri} \partial \vartheta_{sj}} \Big|_{\vartheta=\pi/2, \varphi=0} (M_s/\mu_B)^{-1/2}. \quad (12)$$

It is straightforward now to quantize the system using the usual commutation rules  $[q_{ri}, p_{sj}] = (\hbar/i) \delta_{ij} \delta_{rs}$  for the generalized coordinates and momenta. By introducing bosonic annihilation and creation operators  $a_{ri} = (1/\sqrt{2\hbar})(q_{ri} + ip_{ri})$  and  $a_{ri}^\dagger = (1/\sqrt{2\hbar})(q_{ri} - ip_{ri})$ , the Hamilton operator of magnetic excitations can be written as

$$\begin{aligned} \mathcal{H} = & \frac{1}{2} \sum_{rs,ij} (A_{ri,sj} - iB_{ri,sj} + iB_{sj,ri} + C_{ri,sj}) a_{ri}^\dagger a_{sj} + \text{H.c.}, \\ & + \frac{1}{2} \sum_{rs,ij} (A_{ri,sj} + iB_{ri,sj} + iB_{sj,ri} - C_{ri,sj}) a_{ri}^\dagger a_{sj}^\dagger + \text{H.c.} \end{aligned} \quad (13)$$

By utilizing the two-dimensional translational symmetry of the coefficients  $A_{ri,sj}$ ,  $B_{ri,sj}$ , and  $C_{ri,sj}$  one can rewrite the above Hamiltonian as

$$\begin{aligned} \mathcal{H} = & \frac{1}{2} \sum_{\mathbf{q},rs} h_{rs}(\mathbf{q}) [a_r^\dagger(\mathbf{q}) a_s(\mathbf{q}) + a_s(\mathbf{q}) a_r^\dagger(\mathbf{q})] \\ & + \frac{1}{2} \sum_{\mathbf{q},rs} [w_{rs}(\mathbf{q}) a_r^\dagger(\mathbf{q}) a_s^\dagger(-\mathbf{q}) + w_{sr}^*(\mathbf{q}) a_r(\mathbf{q}) a_s(-\mathbf{q})], \end{aligned} \quad (14)$$

where

$$a_r(\mathbf{q}) = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{q} \cdot \mathbf{T}_i} a_{ri} \quad (15)$$

and

$$h_{rs}(\mathbf{q}) = \sum_i (A_{r0,sj} - iB_{r0,sj} + iB_{sj,r0} + C_{r0,sj}) e^{i\mathbf{q}(\mathbf{T}_j + \mathbf{C}_s - \mathbf{C}_r)}, \quad (16)$$

$$w_{rs}(\mathbf{q}) = \sum_i (A_{r0,sj} + iB_{r0,sj} + iB_{sj,r0} - C_{r0,sj}) e^{i\mathbf{q}(\mathbf{T}_j + \mathbf{C}_s - \mathbf{C}_r)}, \quad (17)$$

with the two-dimensional translational vectors  $\mathbf{T}_i$  and the layer generating vectors  $\mathbf{C}_r$  ( $\mathbf{R}_{ri} = \mathbf{C}_r + \mathbf{T}_i$ ).

The same structure of the Hamiltonian has been obtained by Erickson and Mills<sup>16</sup> in terms of the Holstein-Primakoff transformation of a Heisenberg Hamiltonian with additional magnetic dipole-dipole interactions. Following their procedure, in order to diagonalize the Hamiltonian in Eq. (14) we introduce a new set of creation and annihilation operators  $\alpha_s^\dagger(\mathbf{q})$  and  $\alpha_s(\mathbf{q})$ , respectively, defined by the transformations

$$a_r(\mathbf{q}) = \sum_s S_{rs}(\mathbf{q}) \alpha_s(\mathbf{q}) + T_{rs}^*(\mathbf{q}) \alpha_s^\dagger(-\mathbf{q}), \quad (18)$$

$$a_r^\dagger(\mathbf{q}) = \sum_s S_{rs}^*(\mathbf{q}) \alpha_s^\dagger(\mathbf{q}) + T_{rs}(\mathbf{q}) \alpha_s(-\mathbf{q}), \quad (19)$$

where the transformation matrices  $\mathbf{S}(\mathbf{q})$  and  $\mathbf{T}(\mathbf{q})$  satisfy the eigenvalue equation

$$\begin{pmatrix} \mathbf{h}(\mathbf{q}) & \mathbf{w}(\mathbf{q}) \\ -\mathbf{w}^\dagger(\mathbf{q}) & -\mathbf{h}^\dagger(\mathbf{q}) \end{pmatrix} \begin{pmatrix} \mathbf{S}(\mathbf{q}) & \mathbf{T}^*(\mathbf{q}) \\ \mathbf{T}(-\mathbf{q}) & \mathbf{S}^*(-\mathbf{q}) \end{pmatrix} = \begin{pmatrix} \mathbf{S}(\mathbf{q}) & \mathbf{T}^*(\mathbf{q}) \\ \mathbf{T}(-\mathbf{q}) & \mathbf{S}^*(-\mathbf{q}) \end{pmatrix} \begin{pmatrix} \boldsymbol{\varepsilon}(\mathbf{q}) \\ -\boldsymbol{\varepsilon}(\mathbf{q}) \end{pmatrix}. \quad (20)$$

Quite clearly, the diagonal matrix  $\boldsymbol{\varepsilon}(\mathbf{q})$  contains the excitation energies of the magnons.

So far we have not made any distinction between a relativistic and a nonrelativistic theory. Relativistic effects enter through the parameters  $A_{ri,sj}$ ,  $B_{ri,sj}$ , and  $C_{ri,sj}$  defined by Eqs. (10), (11), and (12), respectively. However, it is easy to see that in a nonrelativistic case the mixed partial derivatives of the free energy,  $B_{ri,sj}$ , vanish and there is no difference between the second derivatives with respect to the azimuthal and polar angles, i.e.,  $A_{ri,sj} = C_{ri,sj}$ . Consequently, the second term in Eq. (14) disappears and the Hamiltonian is of the form of a simple harmonic oscillator. In a relativistic treatment the second term in Eq. (14) appears to be nonzero due to the magnetic dipole-dipole interaction and spin-orbit coupling. The effect of the magnetic dipole-dipole interaction is described in detail in Refs. 15–17, but according to our knowledge the effect of the spin-orbit coupling on the spin-wave spectra has not yet been studied from first principles.

### B. Evaluation in terms of the Korringa-Kohn-Rostoker method

Expressions for the derivatives of the free energy with respect to the orientation of the magnetic moments can be derived in the framework of the fully relativistic KKR method which was successfully applied to calculations of magnetic anisotropy of thin films.<sup>20</sup> Within the magnetic force theorem, the free energy (grand potential) at zero temperature is approximated by

$$\mathcal{F} = \int_{-\infty}^{E_F} d\varepsilon (\varepsilon - E_F) n(\varepsilon) = - \int_{-\infty}^{E_F} d\varepsilon N(\varepsilon), \quad (21)$$

where  $E_F$  denotes the Fermi energy of the system,  $n(\varepsilon)$  is the density of states (DOS), and  $N(\varepsilon)$  is the integrated DOS. Employing Lloyd's formula,<sup>21</sup> apart from a constant term corresponding to the potential-free system, the free energy can further be written as

$$\mathcal{F} = - \frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr} \ln \boldsymbol{\tau}(\varepsilon), \quad (22)$$

where the site representation of the scattering path operator  $\boldsymbol{\tau}(\varepsilon) = \{\tau_{ij}(\varepsilon)\}$  can be expressed in terms of a site-diagonal single-site scattering matrix  $\mathbf{t}(\varepsilon) = \{t_i(\varepsilon) \delta_{ij}\}$  and the structure constants  $\mathbf{G}_0(\varepsilon) = \{G_{0,ij}(\varepsilon) (1 - \delta_{ij})\}$  as

$$\boldsymbol{\tau}(\varepsilon) = [\mathbf{t}^{-1}(\varepsilon) - \mathbf{G}_0(\varepsilon)]^{-1}. \quad (23)$$

In here,  $\tau_{ij}(\varepsilon)$ ,  $t_i(\varepsilon)$ , and  $G_{ij}(\varepsilon)$  are matrices in angular momentum space.

The change of the free energy has to be expressed up to second order with respect to a small deviation of the orientation of the magnetizations at sites  $i$  and  $j$  relative to the ferromagnetic ground-state orientation. As it is explained in Ref. 20 the orientational dependence of the single-site  $t$  matrix corresponds to a similarity transformation that rotates the  $z$  axis of the reference system to the desired orientation given by the angles  $\vartheta_i$  and  $\varphi_i$ ,

$$t_i^{-1} = m_i = R(\vartheta_i, \varphi_i) m_i^0 R^\dagger(\vartheta_i, \varphi_i), \quad (24)$$

where  $m_i^0$  denotes the inverse of the  $t$  matrix at site  $i$  in a local frame in which the  $z$  axis coincides with the axis of the spin quantization (magnetization). Note that for brevity we have dropped the energy argument from the corresponding matrices. The change of  $m_i$  up to second order in  $\vartheta_i$  and  $\varphi_i$ ,

$$\Delta m_i^{(1)} = m_i^{\vartheta} d\vartheta_i + m_i^{\varphi} d\varphi_i, \quad (25)$$

$$\Delta m_i^{(2)} = \frac{1}{2} m_i^{\vartheta\vartheta} d\vartheta_i d\vartheta_i + m_i^{\varphi\vartheta} d\vartheta_i d\varphi_i + \frac{1}{2} m_i^{\varphi\varphi} d\varphi_i d\varphi_i, \quad (26)$$

can easily be expressed by means of the derivatives of the rotation matrices  $R(\vartheta_i, \varphi_i)$ ,

$$m_i^{\vartheta} \equiv \frac{\partial m_i}{\partial \vartheta_i} = \frac{\partial R_i}{\partial \vartheta_i} m_i^0 R_i^{\dagger} + R_i m_i^0 \frac{\partial R_i^{\dagger}}{\partial \vartheta_i}, \quad (27)$$

$$m_i^{\varphi} \equiv \frac{\partial m_i}{\partial \varphi_i} = \frac{\partial R_i}{\partial \varphi_i} m_i^0 R_i^{\dagger} + R_i m_i^0 \frac{\partial R_i^{\dagger}}{\partial \varphi_i}, \quad (28)$$

$$m_i^{\vartheta\vartheta} \equiv \frac{\partial^2 m_i}{\partial \vartheta_i \partial \vartheta_i} = \frac{\partial^2 R_i}{\partial \vartheta_i^2} m_i^0 R_i^{\dagger} + R_i m_i^0 \frac{\partial^2 R_i^{\dagger}}{\partial \vartheta_i^2} + 2 \frac{\partial R_i}{\partial \vartheta_i} m_i^0 \frac{\partial R_i^{\dagger}}{\partial \vartheta_i}, \quad (29)$$

$$m_i^{\varphi\vartheta} \equiv \frac{\partial^2 m_i}{\partial \varphi_i \partial \vartheta_i} = \frac{\partial^2 R_i}{\partial \varphi_i \partial \vartheta_i} m_i^0 R_i^{\dagger} + \frac{\partial R_i}{\partial \varphi_i} m_i^0 \frac{\partial R_i^{\dagger}}{\partial \vartheta_i} + \frac{\partial R_i}{\partial \vartheta_i} m_i^0 \frac{\partial R_i^{\dagger}}{\partial \varphi_i} + R_i m_i^0 \frac{\partial^2 R_i^{\dagger}}{\partial \varphi_i \partial \vartheta_i}, \quad (30)$$

$$m_i^{\varphi\varphi} \equiv \frac{\partial^2 m_i}{\partial \varphi_i \partial \varphi_i} = \frac{\partial^2 R_i}{\partial \varphi_i^2} m_i^0 R_i^{\dagger} + R_i m_i^0 \frac{\partial^2 R_i^{\dagger}}{\partial \varphi_i^2} + 2 \frac{\partial R_i}{\partial \varphi_i} m_i^0 \frac{\partial R_i^{\dagger}}{\partial \varphi_i}, \quad (31)$$

where for simplicity we used the abbreviation  $R_i \equiv R(\vartheta_i, \varphi_i)$ .

First we evaluate the diagonal terms of the second derivative tensor of the free energy by introducing a change of the orientation of the magnetization only on site  $i$ . The logarithm of the scattering path operator appearing in Eq. (22) can now be written as

$$\ln \tau' = \ln(\mathbf{m} + \Delta \mathbf{m}_i - \mathbf{G}_0)^{-1} = \ln \tau - \ln(\mathbf{1} + \tau \Delta \mathbf{m}_i). \quad (32)$$

Quite clearly, the site-diagonal matrix  $\Delta \mathbf{m}_i$  has only the block corresponding to site  $i$  with nonzero elements. Expanding the logarithm in Eq. (32) and keeping the terms up to second order one obtains

$$\ln \tau' - \ln \tau = -\tau \Delta \mathbf{m}_i^{(1)} - \tau \Delta \mathbf{m}_i^{(2)} + \frac{1}{2} \tau \Delta \mathbf{m}_i^{(1)} \tau \Delta \mathbf{m}_i^{(1)}. \quad (33)$$

The first term on the right-hand side of the above equation contributes to the gradient of the free energy, while the site-diagonal elements of its second derivative tensor are related to the second and third terms of Eq. (33).

In order to find an expression for the site-off-diagonal part of the second derivative of the free energy, the orientation of the magnetization has to be altered simultaneously at two different sites  $i$  and  $j$  ( $i \neq j$ ),

$$\begin{aligned} \ln \tau' &= \ln(\mathbf{m} + \Delta \mathbf{m}_i + \Delta \mathbf{m}_j - \mathbf{G}_0)^{-1} \\ &= \ln \tau - [\mathbf{1} + \tau(\Delta \mathbf{m}_i + \Delta \mathbf{m}_j)], \end{aligned} \quad (34)$$

which can be rewritten into the form

$$\ln \tau' = \ln \tau - \ln(\mathbf{1} + \tau \Delta \mathbf{m}_i) - \ln(\mathbf{1} + \tau \Delta \mathbf{m}_j) - \ln(\mathbf{1} - \tau \Delta_i \tau \Delta_j), \quad (35)$$

where we introduced the notation  $\Delta_i \equiv \Delta \mathbf{m}_i (\mathbf{1} + \tau \Delta \mathbf{m}_i)^{-1}$ . Expanding Eq. (35) up to second order gives

$$\ln \tau' - \ln \tau = -\tau(\Delta \mathbf{m}_i^{(1)} + \Delta \mathbf{m}_j^{(1)}) + \tau \Delta \mathbf{m}_i^{(1)} \tau \Delta \mathbf{m}_j^{(1)}. \quad (36)$$

Similar to the site-diagonal case the second derivatives can be deduced from the second term of the right-hand side (RHS) of Eq. (36).

In the following a *layered ferromagnetic order* is assumed; i.e., the orientation of the magnetization is the same within each layer but the magnetic moment can be different in different layers. With these conditions the single-site  $t$  matrix depends only on the layer index. Substituting Eqs. (25)–(31) into Eqs. (33) and (36) and applying Eq. (22), the second derivative tensor of the free energy with respect to the site-dependent orientations of the magnetization can be written as follows:

*diagonal terms:*

$$\begin{aligned} \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \varphi_{ri}} &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[-\tau_{r0,r0}(\varepsilon) m_r^{\varphi\varphi}(\varepsilon) \\ &\quad + \tau_{r0,r0}(\varepsilon) m_r^{\varphi}(\varepsilon) \tau_{r0,r0}(\varepsilon) m_r^{\varphi}(\varepsilon)], \end{aligned} \quad (37)$$

$$\begin{aligned} \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \vartheta_{ri}} &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[-\tau_{r0,r0}(\varepsilon) m_r^{\varphi\vartheta}(\varepsilon) \\ &\quad + \tau_{r0,r0}(\varepsilon) m_r^{\varphi}(\varepsilon) \tau_{r0,r0}(\varepsilon) m_r^{\vartheta}(\varepsilon)], \end{aligned} \quad (38)$$

$$\begin{aligned} \frac{\partial^2 \mathcal{F}}{\partial \vartheta_{ri} \partial \vartheta_{ri}} &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[-\tau_{r0,r0}(\varepsilon) m_r^{\vartheta\vartheta}(\varepsilon) \\ &\quad + \tau_{r0,r0}(\varepsilon) m_r^{\vartheta}(\varepsilon) \tau_{r0,r0}(\varepsilon) m_r^{\vartheta}(\varepsilon)]; \end{aligned} \quad (39)$$

*off-diagonal terms:*

$$\begin{aligned} \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \varphi_{sj}} &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[\tau_{sj,ri}(\varepsilon) m_r^{\varphi}(\varepsilon) \tau_{ri,sj}(\varepsilon) m_s^{\varphi}(\varepsilon)], \end{aligned} \quad (40)$$

$$\begin{aligned} & \frac{\partial^2 \mathcal{F}}{\partial \varphi_{ri} \partial \vartheta_{sj}} \\ &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[\tau_{sj,ri}(\varepsilon) m_r^\varphi(\varepsilon) \tau_{ri,sj}(\varepsilon) m_s^\vartheta(\varepsilon)], \end{aligned} \quad (41)$$

$$\begin{aligned} & \frac{\partial^2 \mathcal{F}}{\partial \vartheta_{ri} \partial \vartheta_{sj}} \\ &= -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[\tau_{sj,ri}(\varepsilon) m_r^\vartheta(\varepsilon) \tau_{ri,sj}(\varepsilon) m_s^\vartheta(\varepsilon)], \end{aligned} \quad (42)$$

where  $\tau_{ri,sj}$  is the block of the real-space scattering path operator that corresponds to site  $i$  of layer  $r$  and site  $j$  of layer  $s$ . In Eqs. (37)–(42) the trace has to be performed in angular momentum space.

### C. Relation to classical spin Hamiltonians

In order to study, e.g., magnetic properties at finite temperatures the energy of an itinerant electron system is often mapped onto a simple classical spin Hamiltonian. Using a quadratic approximation of the (effective) spin-spin interaction, such a Hamiltonian can be written in the most general form as

$$\mathcal{H} = \sum_i K(\boldsymbol{\sigma}_i) + \frac{1}{2} \sum_{ij} \boldsymbol{\sigma}_i \mathcal{J}_{ij} \boldsymbol{\sigma}_j, \quad (43)$$

where the first term stands for the on-site anisotropy energy and the  $\mathcal{J}_{ij}$  in the second term are  $3 \times 3$  matrices. It is evident that the matrices  $\mathcal{J}_{ij}$  ( $i \neq j$ ) can be chosen such that  $\mathcal{J}_{ij} = \mathcal{J}_{ji}^t$ , where  $t$  refers to the transposed matrix. In order to relate the Hamiltonian (43) to physically significant interactions,  $\mathcal{J}_{ij}$  has to be decomposed as

$$\mathcal{J}_{ij} = J_{ij} \mathcal{I} + \mathcal{J}_{ij}^S + \mathcal{J}_{ij}^A, \quad (44)$$

where  $\mathcal{I}$  is the unit matrix,

$$J_{ij} = \frac{1}{3} \text{Tr}(\mathcal{J}_{ij}), \quad (45)$$

and the symmetric, traceless part of  $\mathcal{J}_{ij}$ ,  $\mathcal{J}_{ij}^S$ , is defined by

$$\mathcal{J}_{ij}^S = \frac{1}{2} (\mathcal{J}_{ij} + \mathcal{J}_{ij}^t) - J_{ij} \mathcal{I}, \quad (46)$$

while the antisymmetric part of  $\mathcal{J}_{ij}$ ,  $\mathcal{J}_{ij}^A$ , by

$$\mathcal{J}_{ij}^A = \frac{1}{2} (\mathcal{J}_{ij} - \mathcal{J}_{ij}^t). \quad (47)$$

Clearly, therefore, a typical intersite interaction consists of the three terms<sup>22</sup>

$$\boldsymbol{\sigma}_i \mathcal{J}_{ij} \boldsymbol{\sigma}_j = J_{ij} \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j + \boldsymbol{\sigma}_i \mathcal{J}_{ij}^S \boldsymbol{\sigma}_j + \mathbf{D}_{ij} (\boldsymbol{\sigma}_i \times \boldsymbol{\sigma}_j), \quad (48)$$

where the first and second terms on the RHS are the isotropic and the symmetric anisotropic exchange interactions, respectively, while the third term represents the Dzyaloshinsky-Moriya (DM) interaction,<sup>22,23</sup> the vector  $\mathbf{D}_{ij}$  being defined as

$$\begin{aligned} D_{ij}^x &= \frac{1}{2} (J_{ij}^{yz} - J_{ij}^{zy}), & D_{ij}^y &= \frac{1}{2} (J_{ij}^{xz} - J_{ij}^{zx}), \\ D_{ij}^z &= \frac{1}{2} (J_{ij}^{xy} - J_{ij}^{yx}). \end{aligned} \quad (49)$$

Since the on-site term in Eq. (43),  $\boldsymbol{\sigma}_i \mathcal{J}_{ii} \boldsymbol{\sigma}_i$ , contains only symmetric contributions, it can readily be included as a second-order term to the magnetic anisotropy  $K(\boldsymbol{\sigma}_i)$ . Consequently, the summation in the second term on the RHS of Eq. (43) has to run over  $i \neq j$  only. In what follows we show how the parameters of the Hamiltonian (9) can be related to those in Eq. (43), the magnetocrystalline anisotropy and the generalized exchange interactions occurring in Eq. (48) being thus automatically included in our Hamiltonian for transverse magnons.

We start by calculating the second derivatives of the Hamiltonian in Eq. (43) with respect to the polar and azimuthal angles,

$$\begin{aligned} \frac{\partial^2 \mathcal{H}}{\partial \alpha_i \partial \beta_k} &= \delta_{ik} K^{\alpha\beta}(\boldsymbol{\sigma}_i) + \delta_{ik} \sum_{j(\neq i)} \boldsymbol{\sigma}_i^{\alpha\beta} \mathcal{J}_{ij} \boldsymbol{\sigma}_j \\ &+ (1 - \delta_{ik}) \boldsymbol{\sigma}_i^\alpha \mathcal{J}_{ik} \boldsymbol{\sigma}_k^\beta, \end{aligned} \quad (50)$$

where  $\alpha$  and  $\beta$  can be either  $\vartheta$  or  $\varphi$  and

$$K^{\alpha\beta}(\boldsymbol{\sigma}_i) = \frac{\partial^2 K(\boldsymbol{\sigma}_i)}{\partial \alpha_i \partial \beta_i}, \quad \boldsymbol{\sigma}_i^\alpha = \frac{\partial \boldsymbol{\sigma}_i}{\partial \alpha_i}, \quad \boldsymbol{\sigma}_i^{\alpha\beta} = \frac{\partial^2 \boldsymbol{\sigma}_i}{\partial \alpha_i \partial \beta_i}.$$

Note that, according to Eqs. (7) and (8), all the derivatives have to be taken at  $\vartheta_i = \pi/2$  and  $\varphi_i = 0$  in a coordinate system with the  $z$  axis normal to the reference orientation of the magnetization.

If the magnetization points along the  $x$  axis of the global coordinate system, i.e., to which the matrices  $\mathcal{J}_{ij}$  are related, Eq. (4) yields,

$$\begin{aligned} \boldsymbol{\sigma}_i &= \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} & \boldsymbol{\sigma}_i^\varphi &= \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} & \boldsymbol{\sigma}_i^\vartheta &= \begin{pmatrix} 0 \\ 0 \\ -1 \end{pmatrix} & \boldsymbol{\sigma}_i^{\varphi\varphi} &= \begin{pmatrix} -1 \\ 0 \\ 0 \end{pmatrix} \\ \boldsymbol{\sigma}_i^{\vartheta\vartheta} &= \begin{pmatrix} -1 \\ 0 \\ 0 \end{pmatrix} & \boldsymbol{\sigma}_i^{\varphi\vartheta} &= \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \end{aligned} \quad (51)$$

which, when substituted into Eq. (50), leads to

$$\frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \varphi_k} = \delta_{ik} K^{\varphi\varphi}(\boldsymbol{\sigma}_i) - \delta_{ik} \sum_{j(\neq i)} J_{ij}^{xx} + (1 - \delta_{ik}) J_{ik}^{yy}, \quad (52)$$

$$\frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_k} = \delta_{ik} K^{\vartheta\vartheta}(\boldsymbol{\sigma}_i) - \delta_{ik} \sum_{j(\neq i)} J_{ij}^{xx} + (1 - \delta_{ik}) J_{ik}^{zz}, \quad (53)$$

$$\frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \vartheta_k} = \delta_{ik} K^{\varphi \vartheta}(\boldsymbol{\sigma}_i) - (1 - \delta_{ik}) J_{ik}^{yz}, \quad (54)$$

$$\frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \varphi_k} = \delta_{ik} K^{\vartheta \varphi}(\boldsymbol{\sigma}_i) - (1 - \delta_{ik}) J_{ik}^{zy}. \quad (55)$$

Assuming for matters of simplicity second-order uniaxial magnetic anisotropy

$$K(\boldsymbol{\sigma}_i) = -K_{2,i} \sin^2(\vartheta_i), \quad (56)$$

the diagonal components of the second derivative tensor are given by

$$\begin{aligned} \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \varphi_i} &= - \sum_{j(\neq i)} J_{ij}^{xx}, & \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_i} &= 2K_{2,i} - \sum_{j(\neq i)} J_{ij}^{xx}, \\ \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \varphi_i} &= \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \vartheta_i} = 0, \end{aligned} \quad (57)$$

while for the off-diagonal components ( $i \neq k$ ) we get

$$\begin{aligned} \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \varphi_k} &= J_{ik}^{yy}, & \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_k} &= J_{ik}^{zz}, & \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \vartheta_k} &= -J_{ik}^{yz}, \\ \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \varphi_k} &= -J_{ik}^{zy}. \end{aligned} \quad (58)$$

If the magnetization points parallel to the  $z$  axis of the global coordinate system, the corresponding derivatives can be obtained by applying the unitary transformation

$$U = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ -1 & 0 & 0 \end{pmatrix}, \quad (59)$$

whereas also the uniaxial magnetic anisotropy, Eq. (56), has to be transformed to

$$K(\boldsymbol{\sigma}_i) = -K_{2,i} \sin^2(\vartheta_i) \cos^2(\varphi_i). \quad (60)$$

Thus, one ends up with the expressions for the on-site terms,

$$\begin{aligned} \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \varphi_i} &= \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_i} = 2K_{2,i} - \sum_{j(\neq i)} J_{ij}^{zz}, \\ \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \varphi_i} &= \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \vartheta_i} = 0, \end{aligned} \quad (61)$$

and for  $i \neq k$  with

$$\begin{aligned} \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \varphi_k} &= J_{ik}^{yy}, & \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_k} &= J_{ik}^{xx}, & \frac{\partial^2 \mathcal{H}}{\partial \varphi_i \partial \vartheta_k} &= J_{ik}^{yx}, \\ \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \varphi_k} &= J_{ik}^{xy}. \end{aligned} \quad (62)$$

Evidently, the magnetocrystalline anisotropy constants  $K_{2,i}$  and the elements of the matrices  $\mathcal{J}_{ij}$  can be calculated from Eqs. (57), (58), (61), or (62) using Eqs. (37)–(42). (The missing elements  $J_{ij}^{xz}$  and  $J_{ij}^{zx}$  can be derived by choosing the orientation of the magnetization along the  $y$  axis of the global coordinate system.)

It is, however, obvious that this mapping between the Hamiltonians, Eq. (9) and Eq. (43), is homomorphic: the

above procedure can be repeated for any three independent reference orientations of the magnetization, resulting in sets of parameters  $K_{2,i}$  and  $\mathcal{J}_{ij}$  that are not necessarily unique. This follows from the fact that the spin-wave Hamiltonian, Eq. (9), has been derived from a second-order expansion of the exact free energy [see Eq. (22)] in the vicinity of a given magnetic configuration; therefore, it cannot provide an overall good approximation to the energy of the system for arbitrary spin configurations. In other words, when expanded with respect to spin variables, Eq. (22) contains also spin-spin interaction terms beyond the quadratic approximation.<sup>24</sup> Consequently, a mapping to a Hamiltonian of the form of Eq. (43) leads to parameters that depend on the reference magnetic configuration. Using the accidental degeneracy of the mapping [see, e.g., Eqs. (58) and (62) for  $J_{ik}^{yy}$ ], we estimated for a Co monolayer on Cu(001) an uncertainty of about  $5 \mu\text{Ry}$  for the nearest-neighbor interactions, which in turn corresponds to a relative accuracy of about 0.1%.

In the absence of spin-orbit coupling the off-diagonal elements vanish and the diagonal elements become identical: this situation clearly corresponds to isotropic exchange coupling. In this particular case, the similarity transformation in Eq. (24) can be performed in spin space and the off-diagonal terms (40) or (42) as well as the diagonal terms (37) or (39) become identical to the expressions derived by Liechtenstein *et al.*<sup>11</sup> for the effective exchange interaction parameters  $J_{ij}$  and for the on-site rotation terms  $J_i$ , respectively. Since in the nonrelativistic case there is no preferred orientation of the magnetization, the energy of the system is invariant against a global rotation of all the magnetic moments, and the following condition has to be satisfied:

$$\frac{\partial^2 \mathcal{H}}{\partial \alpha_i \partial \alpha_i} + \sum_{j(\neq i)} \frac{\partial^2 \mathcal{H}}{\partial \alpha_i \partial \alpha_j} = 0 \quad (\alpha = \vartheta \text{ or } \varphi), \quad (63)$$

which is equivalent to the condition for the exchange interaction parameters,<sup>11</sup>

$$J_i = \sum_{j(\neq i)} J_{ij}. \quad (64)$$

In the relativistic case this sum rule no longer applies. Indeed, the expression on the LHS of Eq. (63) can be used to define an effective second-order magnetic anisotropy constant  $\lambda_i$ . For the case of a magnetization parallel to the  $z$  axis of the global coordination frame, Eqs. (61) and (62) imply

$$\begin{aligned} \lambda_i &\equiv \frac{1}{2} \left( \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_i} + \sum_{j(\neq i)} \frac{\partial^2 \mathcal{H}}{\partial \vartheta_i \partial \vartheta_j} \right) \\ &= K_{2,i} - \frac{1}{2} \sum_{j(\neq i)} (\mathcal{J}_{ij}^{zz} - \mathcal{J}_{ij}^{xx}). \end{aligned} \quad (65)$$

The physical content of the above equation is obvious: beyond the on-site anisotropy term the magnetocrystalline anisotropy energy of the system contains contributions that arise from the anisotropy of the intersite exchange interactions.

Going back now to Eqs. (39) and (42) and using the notation used in Sec. II B, one obtains, for the layer-resolved anisotropies,

$$\begin{aligned}
\lambda_r = & -\frac{1}{2\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[-\tau_{r0,r0}(\varepsilon) m_r^{\dagger\ddagger}(\varepsilon) \\
& + \tau_{r0,r0}(\varepsilon) m_r^{\ddagger}(\varepsilon) \tau_{r0,r0}(\varepsilon) m_r^{\dagger}(\varepsilon)] \\
& - \sum_{sj} \frac{1}{2\pi} \text{Im} \int_{-\infty}^{E_F} d\varepsilon \text{Tr}[\tau_{sj,r0}(\varepsilon) m_r^{\dagger}(\varepsilon) \tau_{r0,sj}(\varepsilon) m_s^{\ddagger}(\varepsilon)],
\end{aligned} \tag{66}$$

which for practical purposes can be rewritten in terms of reciprocal-space summations as

$$\begin{aligned}
\lambda_r = & \frac{1}{2\pi\Omega} \text{Im} \int_{-\infty}^{E_F} d\varepsilon d\mathbf{k}_{\parallel} \text{Tr}[\tau_{rr}(\varepsilon, \mathbf{k}_{\parallel}) m_r^{\dagger\ddagger}(\varepsilon)] \\
& - \frac{1}{2\pi\Omega} \sum_s \text{Im} \int_{-\infty}^{E_F} d\varepsilon d\mathbf{k}_{\parallel} \text{Tr}[\tau_{rs}(\varepsilon, \mathbf{k}_{\parallel}) m_r^{\dagger}(\varepsilon) \\
& \times \tau_{rs}(\varepsilon, \mathbf{k}_{\parallel}) m_s^{\ddagger}(\varepsilon)],
\end{aligned} \tag{67}$$

where the  $\mathbf{k}$  integral is performed in the first two-dimensional (2D) Brillouin zone, the area of which is denoted by  $\Omega$ . Note that Eq. (67) can be regarded as an extension of the expression for the anisotropy energy derived in Refs. 25 and 26 to the case of two-dimensional translational symmetry.

### III. RESULTS

We demonstrate our method for three systems: an Fe and a Co atomic monolayer on a Cu(001) surface and an Fe monolayer on Au(001). The calculations were performed supposing perfect epitaxial structure by neglecting any relaxation of the lattice. The energy integrals in Eqs. (37)–(42) were performed on a semicircle contour using a 16-point Gaussian quadrature. The integration over the full Brillouin zone was performed by taking up to  $9 \times 10^4$   $\mathbf{k}$  points at the energy closest to the Fermi level, and the number of  $\mathbf{k}$  points then gradually decreased for energy points more distant from the real axis in the complex plane and towards the bottom of the band. The second derivatives, Eqs. (40)–(42), were calculated within a sphere with a radius of up to 29 times the 2D lattice constant. The induced moments of the nonmagnetic substrate and vacuum layers were fairly small as compared to the magnetic moments of the ferromagnetic layers. However, the neglect of these small moments—i.e., missing summation over the corresponding sites—introduced an error comparable to the gap at  $\mathbf{q}=0$  due to spin-orbit coupling. In our calculation, therefore, beyond the ferromagnetic layer the nearest and next-nearest nonmagnetic layers are involved. In order to test the accuracy of our procedure, the sum rule, Eq. (63), was checked with spin-orbit coupling turned off.<sup>27</sup> Assuringly, the deviation was always at least two orders of magnitude smaller than the corresponding gap in the magnon spectrum. The magnetic dipole-dipole interaction was taken into account as an additional term to the band energy contribution (22),

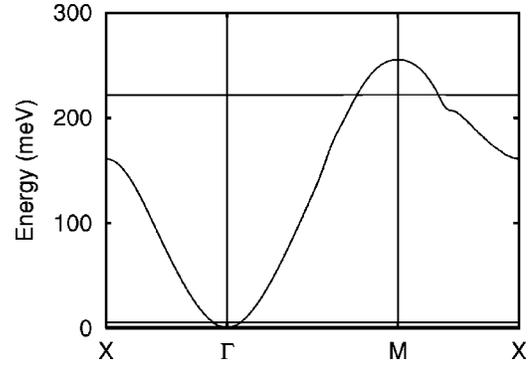


FIG. 1. Spin-wave spectrum for Fe<sub>1</sub>/Au(001). The almost dispersion-less bands belong to the two Au layers adjacent the Fe monolayer.

$$E_{dip} = \frac{1}{c^2} \sum_{i \neq j} \frac{R_{ij}^2 \mathbf{M}_i \cdot \mathbf{M}_j - 3(\mathbf{M}_i \cdot \mathbf{R}_{ij})(\mathbf{R}_{ij} \cdot \mathbf{M}_j)}{R_{ij}^5}. \tag{68}$$

As has been shown by Szunyogh *et al.*<sup>20</sup> the orientation of the magnetization for an Fe monolayer on Au(001) is perpendicular to the surface due to the relatively strong band energy anisotropy. In the case of an Fe monolayer on Cu(001) the magnetic dipole-dipole interaction overrides the positive band energy contribution (out-of-plane orientation), resulting in an in-plane ferromagnetic order.<sup>28</sup> For a cobalt monolayer on Cu(001) surface both the magnetic anisotropy and the magnetic dipole-dipole interaction favor the in-plane orientation of the magnetization.<sup>29</sup> The magnon spectra of the three systems should therefore account for these three different situations.

The calculated magnon spectra along the high-symmetry directions of the Brillouin zone are depicted in Figs. 1–3. The almost dispersion-less bands belong to the nonmagnetic nearest- and next-nearest-neighbor layers. There are anti-crossings between the bands which are the most pronounced in the case of Co/Cu(001), indicating the largest interactions between the magnetic and nonmagnetic layers. For Fe/Cu(001), the dispersion relation is very similar to that ob-

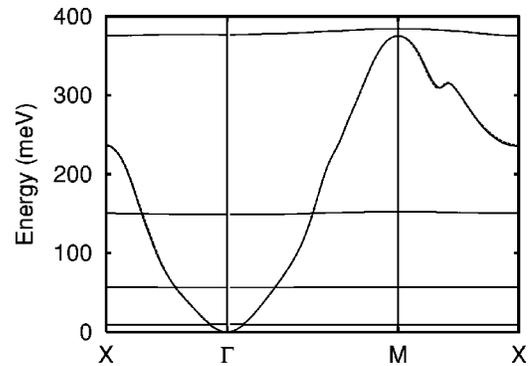


FIG. 2. Spin-wave spectrum for Fe<sub>1</sub>/Cu(001) with an in-plane ground-state magnetization. Four additional Cu layers indicated by bands of very low dispersion have been taken into account. The almost identical solid and dashed lines represent the (100) and (010) directions of the magnetization, respectively.

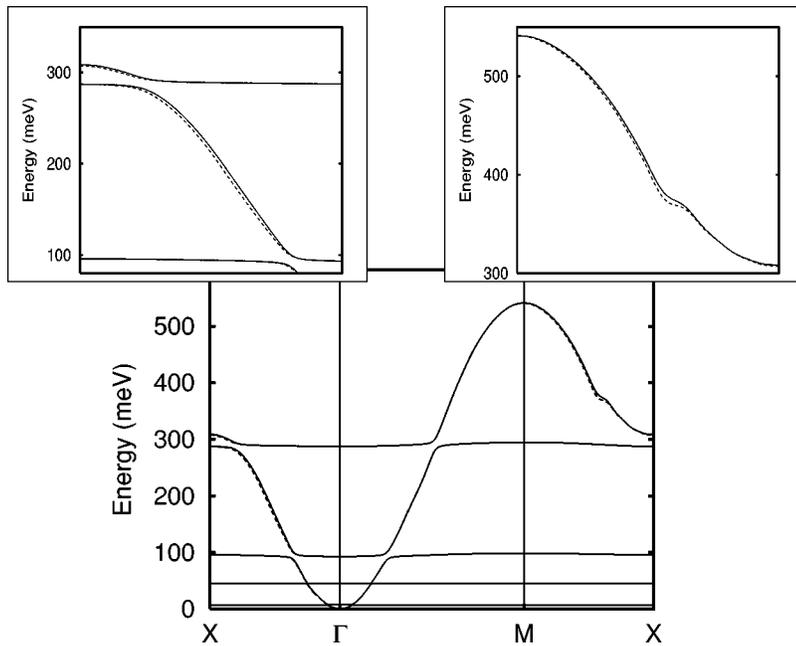


FIG. 3. Spin-wave spectrum for  $\text{Co}_1/\text{Cu}(001)$  with an in-plane ground-state magnetization. The four additional Cu layers considered in the calculations are coupled relatively strongly to the Co monolayer as indicated by the noncrossing behavior (hybridization) of the corresponding bands. The solid and dashed lines represent the (100) and (010) directions of the magnetization, respectively. The spectrum between the symmetry points  $\text{X}$  and  $\Gamma$  as well as between  $\text{M}$  and  $\text{X}$  is shown on an enlarged scale in upper left and upper right insets, respectively.

tained by Pajda *et al.*<sup>18</sup> using the method of infinitesimal rotations combined with the nonrelativistic tight-binding linear muffin-tin orbital (TB-LMTO) approach. A local minimum observed between the points  $\text{X}$  and  $\text{M}$  is an indication for a so-called Kohn anomaly<sup>4</sup> which is the consequence of the long-range RKKY-like behavior of the exchange interactions.

In Fig. 4 the spectra in the vicinity of the  $\Gamma$  point are shown for  $\text{Fe}/\text{Au}(001)$  as calculated for an in-plane and for an out-of-plane magnetic orientation, and also for the case of neglecting spin-orbit coupling. In the absence of spin-orbit coupling the dispersion curve starts from zero as has to be expected. Inclusion of spin-orbit coupling opens up a gap of  $\Delta = 43 \mu\text{Ry}$  for the normal-to-plane orientation. For an in-plane magnetization the relativistic calculation resulted in an imaginary magnon energy close to the  $\Gamma$  point, indicating that the in-plane ferromagnetic order corresponds to a local maximum in the free energy and the easy axis is perpendicular to the surface.

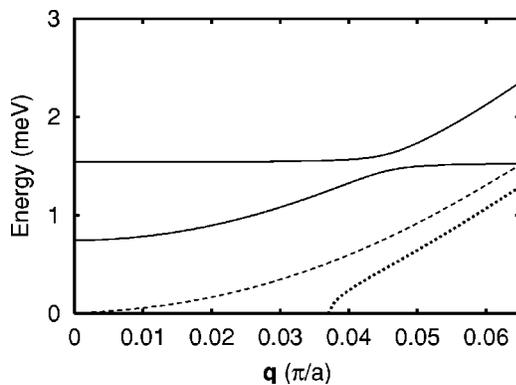


FIG. 4. Spin-wave spectrum for  $\text{Fe}_1/\text{Au}(100)$  in the vicinity of the  $\Gamma$  point. The solid lines correspond to the normal-to-plane orientation of the magnetization, the dotted line represents the spectrum for an in-plane orientation, and the dashed line denotes the magnon spectrum with spin-orbit-coupling turned off.

The long-wavelength parts of the relativistic spectra of the  $\text{Fe}/\text{Cu}(001)$  monolayer without and with inclusion of the magnetic dipole-dipole interaction are shown in Fig. 5. Considering only the band-energy contributions the easy axis turned out to be perpendicular to the surface and a gap of  $\Delta = 17.5 \mu\text{Ry}$  appeared in the corresponding magnon spec-

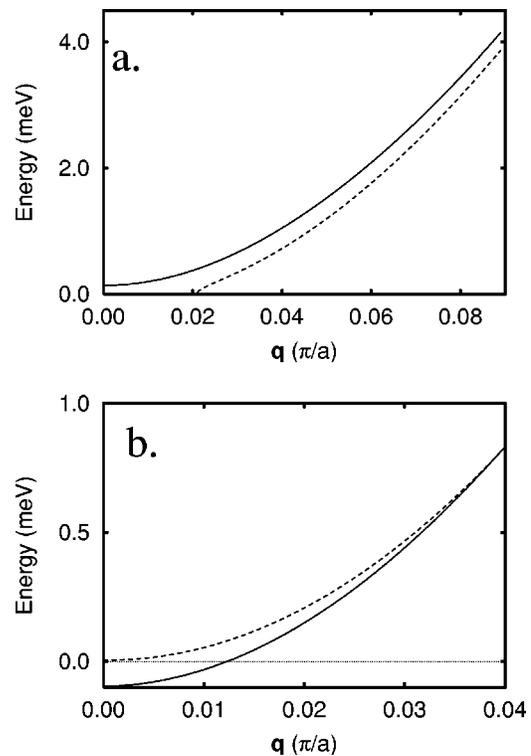


FIG. 5. Spin-wave spectrum in the vicinity of the  $\Gamma$  point for  $\text{Fe}_1/\text{Cu}(100)$  neglecting the magnetic dipole-dipole interaction (a) and including the magnetic dipole-dipole interaction (b). The solid and dashed lines represent the spectra corresponding to the normal-to-plane and to the in-plane magnetic orientations, respectively.

trum, while, similar to the previous case of Fe/Au(001), there are imaginary excitation energies in the in-plane magnon spectrum at small wave numbers. Inclusion of the magnetic dipole-dipole term, Eq. (68), reverses the situation: there is a tiny gap for the in-plane magnetized film and the magnon spectrum becomes negative for the out-of-plane directions.

In the case of a thin film with an easy axis perpendicular to the layers the gap is determined by the second-order uniaxial anisotropy. For a magnetic layer with an in-plane easy axis the lowest energy of the magnetic excitations—i.e., the gap at  $\mathbf{q}=0$ —lies in the order of the in-plane anisotropy energy which for the present systems of  $c_{4v}$  symmetry is at least by an order of magnitude smaller than the uniaxial one. Our calculation resulted in a gap of  $\Delta=0.3 \mu\text{Ry}$  for the Fe/Cu(001) monolayer and the easy axis turned out to be parallel to the (110) direction. We have to mention that such a small value is very close to the numerical accuracy of our calculations inherent mainly to the Brillouin zone sampling.

A similar drop of the gap has, however, been indicated in the experiments for a thermally deposited three-monolayer film of Fe on Cu(001). In contrast to the pulsed-laser-deposited (PLD) samples having an in-plane ground-state magnetization<sup>30,31</sup> thermal deposition (TD) produces Fe films on Cu(001) with an out-of-plane easy axis.<sup>32</sup> In their Brillouin light scattering experiment Dutcher *et al.*<sup>32</sup> studied the lowest-lying spin-wave mode in the presence of an external magnetic field parallel to the substrate. They found that the frequency of this mode has a minimum as the magnetic field passes a critical value where the spins become parallel to the film.

In the case of an in-plane magnetization the  $c_{4v}$  symmetry of the fcc (001) surface is lifted and the spin-wave spectra for the (001) and (010) directions of the magnetization become different as is shown in Fig. 3 for Co/Cu(001). A splitting of a few meV due to spin-orbit coupling can clearly be seen in the insets in Fig. 3. This splitting of the spectrum can also be interpreted in terms of the Dzyaloshinsky-Moriya interaction.<sup>22,23</sup> On the one hand, it is straightforward to show that transverse magnons are affected only by the component of the DM vector,  $D_{ij}$  [see Eq. (49)], parallel to the ground-state magnetization. On the other hand, in agreement with the symmetry rules set up by Moriya,<sup>22</sup> we observed noticeable parallel components of the DM vectors only if the vector  $R_{ij}$  connecting the two interacting spins is aligned close to a direction perpendicular to the magnetization, while the DM interactions along a direction parallel to the ground-state magnetization turned out to be practically zero. Evidently, the above real-lattice asymmetry of the DM interactions shows up as a corresponding asymmetry in the spin-wave spectrum for wave vectors parallel and normal to the magnetization.

There is another interesting feature of the in-plane magnons. The long-wavelength part of the spin-wave spectrum is determined by the anisotropy fields which are considerably different for the out-of-plane and for the in-plane motion of the magnetic moments and, consequently, the spin waves are elliptically polarized. In our theoretical picture the out-of-plane and in-plane motions are assigned to the angles  $\vartheta_{ri}$  and  $\varphi_{ri}$ , respectively. In order to characterize the ellipticity of a

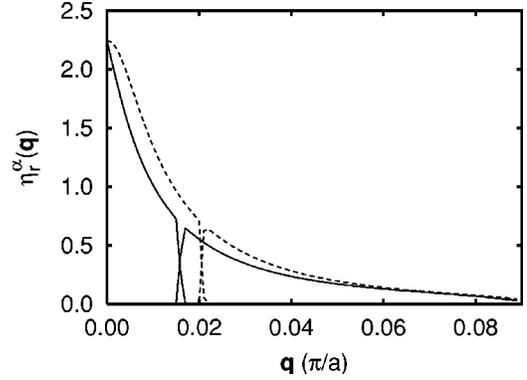


FIG. 6. Spectral resolution of the ellipticity,  $\eta_r^\alpha(\mathbf{q})$ , for the magnon band having the largest weight at the Co monolayer in  $\text{Co}_1/\text{Cu}(001)$ . The solid and dashed lines refer to the (100) and (010) directions of the magnetization, respectively.

magnon in a specific mode (band)  $\alpha$ , the difference of the mean-square deviation of the out-of-plane and in-plane components of the magnetization,

$$\begin{aligned} \eta^\alpha &= (\Delta M_\alpha^\perp)^2 - (\Delta M_\alpha^\parallel)^2 \simeq \left\langle \sum_{ri} M_r^2 (\vartheta_{ri}^2 - \varphi_{ri}^2) \right\rangle_\alpha \\ &= \mu_B \left\langle \sum_{ri} (q_{ri}^2 - p_{ri}^2) \right\rangle_\alpha, \end{aligned} \quad (69)$$

is introduced, where the notation  $\langle \rangle_\alpha$  refers to a quantum-mechanical expectation value with respect to the eigenstate labeled by  $\alpha$  of the Hamiltonian (14). The ellipticity (69) can obviously be expressed as a sum in reciprocal space,

$$\eta^\alpha = \sum_{r,\mathbf{q}} \eta_r^\alpha(\mathbf{q}), \quad (70)$$

$$\eta_r^\alpha(\mathbf{q}) = \langle a_r^\dagger(\mathbf{q}) a_r^\dagger(-\mathbf{q}) + a_r(\mathbf{q}) a_r(-\mathbf{q}) \rangle_\alpha, \quad (71)$$

while  $\eta_r^\alpha(\mathbf{q})$  can easily be calculated using the matrices in Eq. (20),

$$\eta_r^\alpha(\mathbf{q}) = S_{r\alpha}(\mathbf{q}) T_{r\alpha}(-\mathbf{q}) + T_{r\alpha}^*(\mathbf{q}) S_{r\alpha}^*(-\mathbf{q}). \quad (72)$$

In Fig. 6,  $\eta_r^\alpha(\mathbf{q})$  is shown for the band having the largest weight in the cobalt layer in the case of Co/Cu(001) along the (001) and (010) directions. As the wave number is increased the isotropic exchange interaction plays an increasingly important role in the formation of the magnetic excitations and the ellipticity rapidly tends to zero. The band crossing and the asymmetry are also reflected by the ellipticity.

Finally in Table I we summarize the uniaxial anisotropy constants provided by Eq. (67) together with those obtained by taking the band-energy difference between the cases of an in-plane and a normal-to-plane magnetization.<sup>20,28,29</sup> The results of the two types of calculations are in very good agreement. It is worth mentioning that the present method naturally supplies layer-resolved anisotropy constants that can

TABLE I. The gap at the  $\Gamma$  point for monolayers with out-of-plane magnetization and the uniaxial magnetic anisotropy constants given by the present method and by the band energy difference method.

System	Gap ( $q=0$ )	$\lambda$ Present method	$\lambda$ Band energy diff.
Fe <sub>1</sub> /Au(001)	56.8 $\mu$ Ry	42.8 $\mu$ Ry	42.9 $\mu$ Ry
Fe <sub>1</sub> /Cu(001)	17.5 $\mu$ Ry <sup>a</sup>	12.1 $\mu$ Ry	12.2 $\mu$ Ry
Co <sub>1</sub> /Cu(001)		-30.6 $\mu$ Ry	-31.3 $\mu$ Ry

<sup>a</sup>Magnetic dipole-dipole interaction is not included.

differ from the projection of the band-energy anisotropies to the layers as applied so far in magnetic anisotropy calculations.

#### IV. SUMMARY

In order to study low-energy spin-wave excitations of itinerant ferromagnets we have developed a relativistic first-principles method based on the adiabatic decoupling and rigid-spin approximation. A spin-wave Hamiltonian has been constructed starting from the Landau-Lifshitz equation and using a harmonic approximation for the free energy. Closed formulas for the derivatives of the free energy up to second order with respect of the polar and azimuthal angles have been derived within the framework of the magnetic force theorem by means of the fully relativistic Korringa-Kohn-Rostoker method. Exchange and spin-orbit couplings have thus been treated in equal terms. We discussed in details the relevance of classical spin models with respect to the Hamiltonian of transverse magnons. The method has been applied to an Fe monolayer on Cu(001) and Au(001) surfaces and for a Co monolayer on Cu(001). Our calculations reproduced the gap at zero wave number due to spin-orbit coupling and provided uniaxial anisotropy energies in good agreement with the results of the band-energy difference method. We

pointed out that a relativistic treatment gives rise to an asymmetry in the magnon spectra with respect to magnetic orientations along two in-plane easy axes. We also emphasized that in the case of an in-plane magnetized system the long-wavelength magnons are elliptically polarized due to the difference of the second-order uniaxial and fourth-order in-plane magnetic anisotropy.

The lifetime of the magnetic excitations in the adiabatic approach is infinitely long. Edwards and Muniz<sup>33</sup> proved that the adiabatic approximation gives quantitatively reliable results only to order of  $q^2$  in the dispersion relation. Rigorously, one can use it to calculate the exchange stiffness, but if one tries to extend the adiabatic approach to the short-wavelength region, the method may break down. Mills and co-workers<sup>34-36</sup> extended the discussion to ultrathin films and made careful comparisons between the dispersion curves calculated with dynamical theory and the adiabatic approach at very short wavelengths.

Relativistic effects can also have an important impact on the damping of the spin-wave excitations allowing transitions which are forbidden in the spin-conserving nonrelativistic description. These transitions for the low-energy excitations can contribute to the Gilbert damping. The investigation of the relativistic effects in magnetic excitations beyond the adiabatic approximation is an important issue; this, however, extends the scope of the present study.

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