Nonlocal torque operators in *ab initio* theory of the Gilbert damping in random ferromagnetic alloys

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We present an *ab initio* theory of the Gilbert damping in substitutionally disordered ferromagnetic alloys. The theory rests on introduced nonlocal torques which replace traditional local torque operators in the well-known torque-correlation formula and which can be formulated within the atomic-sphere approximation. The formalism is sketched in a simple tight-binding model and worked out in detail in the relativistic tight-binding linear muffin-tin orbital method and the coherent potential approximation (CPA). The resulting nonlocal torques are represented by nonrandom, non-site-diagonal, and spin-independent matrices, which simplifies the configuration averaging. The CPA-vertex corrections play a crucial role for the internal consistency of the theory and for its exact equivalence to other first-principles approaches based on the random local torques. This equivalence is also illustrated by the calculated Gilbert damping parameters for binary NiFe and FeCo random alloys, for pure iron with a model atomic-level disorder, and for stoichiometric FePt alloys with a varying degree of $L1_0$ atomic long-range order.

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I. INTRODUCTION

The dynamics of magnetization of bulk ferromagnets, ultrathin magnetic films, and magnetic nanoparticles represents an important property of these systems, especially in the context of high-speed magnetic devices for data storage. While a complete picture of magnetization dynamics including, e.g., excitation of magnons and their interaction with other degrees of freedom is still a challenge for the modern theory of magnetism, remarkable progress has been achieved in recent years concerning the dynamics of the total magnetic moment, which can be probed experimentally by means of the ferromagnetic resonance [1] or by the time-resolved magneto-optical Kerr effect [2]. Time evolution of the macroscopic magnetization vector $\mathbf{M}$ can be described by the well-known Landau-Lifshitz-Gilbert (LLG) equation [3,4]

$$\frac{d\mathbf{M}}{dt} = \mathbf{B}_{\rm eff} \times \mathbf{M} + \frac{\mathbf{M}}{M} \times \left(\underline{\boldsymbol{\alpha}} \cdot \frac{d\mathbf{M}}{dt} \right), \tag{1}$$

where $\mathbf{B}_{\rm eff}$ denotes an effective magnetic field (with the gyromagnetic ratio absorbed) acting on the magnetization, $M = |\mathbf{M}|$, and the quantity $\underline{\boldsymbol{\alpha}} = \{\alpha_{\mu\nu}\}$ denotes a symmetric 3×3 tensor of the dimensionless Gilbert damping parameters $(\mu, \nu = x, y, z)$. The first term in Eq. (1) defines a precession of the magnetization vector around the direction of the effective magnetic field, and the second term describes a damping of the dynamics. The LLG equation in itinerant ferromagnets is appropriate for magnetization precessions that are very slow as compared to precessions of the single-electron spin due to the exchange splitting and to frequencies of interatomic electron hoppings.

A large number of theoretical approaches to the Gilbert damping have been worked out during the last two decades; here we mention only schemes within the one-electron theory of itinerant magnets [5–20], where the most important effects of electron-electron interaction are captured by means of a local spin-dependent exchange-correlation (XC) potential. These techniques can be naturally combined with existing first-principles techniques based on the density-functional theory, which leads to parameter-free calculations of the Gilbert damping tensor of pure ferromagnetic metals, their ordered and disordered alloys, diluted magnetic semiconductors, etc. One part of these approaches is based on a static limit of the frequency-dependent spin-spin correlation function of a ferromagnet [5–8,15,16]. Other routes to the Gilbert damping employ relaxations of occupation numbers of individual Bloch electron states during quasistatic nonequilibrium processes or transition rates between different states induced by the spin-orbit (SO) interaction [9–12,14,20]. The dissipation of magnetic energy accompanying the slow magnetization dynamics, evaluated within a scattering theory or the Kubo linear response formalism, leads also to explicit expressions for the Gilbert damping tensor [13,17–19]. Most of these formulations yield relations equivalent to the so-called torque-correlation formula

$$\alpha_{\mu\nu} = -\alpha_0 \mathrm{Tr}\{T_\mu (G_+ - G_-) T_\nu (G_+ - G_-)\}, \tag{2}$$

in which the torque operators T_μ are either due to the XC or SO terms of the one-electron Hamiltonian. In Eq. (2), which has the form of the Kubo-Greenwood formula and is valid for zero temperature of electrons, the quantity α_0 is related to the system magnetization (and to fundamental constants and units used; see Sec. II B), the trace is taken over the whole Hilbert space of valence electrons, and the symbols $G_\pm = G(E_{\rm F} \pm i0)$ denote the one-particle retarded and advanced propagators (Green's functions) at the Fermi energy $E_{\rm F}$.

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Implementation of the above-mentioned theories in first-principles computational schemes proved opposite trends of the intraband and interband contributions to the Gilbert damping parameter as functions of a phenomenological quasiparticle lifetime broadening [7,11,12]. These qualitative studies have recently been put on a more solid basis by considering a particular mechanism of the lifetime broadening, namely, a frozen temperature-induced structural disorder, which represents a realistic model for treatment of temperature dependence of the Gilbert damping [21,22]. This approach explained quantitatively the low-temperature conductivity-like and high-temperature resistivity-like trends of the damping parameters of iron, cobalt, and nickel. Further improvements of the model, including static temperature-induced random orientations of local magnetic moments, have appeared recently [23].

The *ab initio* studies have also been successful in reproduction and interpretation of values and concentration trends of the Gilbert damping in random ferromagnetic alloys, such as the NiFe alloy with the fcc structure (permalloy) [17,22] and Fe-based alloys with the bcc structure (FeCo, FeV, FeSi) [19,22,24]. Other studies also addressed the effects of doping the permalloy and bcc iron by $5d$ transition-metal elements [19,20,22] and of the degree of atomic long-range order in equiconcentration FeNi and FePt alloys with the $L1_0$-type structures [20]. Recently, an application to half-metallic Co-based Heusler alloys has appeared as well [25]. The obtained results revealed correlations of the damping parameter with the density of states at the Fermi energy and with the size of magnetic moments [22,24].

In a one-particle mean-field-like description of a ferromagnet, the total spin is not conserved due to the XC field and the SO interaction. The currently employed forms of the torque operators T_μ in the torque-correlation formula (2) reflect these two sources; both the XC- and the SO-induced torques are local, and their equivalence for the theory of Gilbert damping has been discussed by several authors [15,16,26]. In the case of random alloys, this equivalence rests on a proper inclusion of vertex corrections in the configuration averaging of the damping parameters $\alpha_{\mu\nu}$ as two-particle quantities.

The purpose of the present paper is to introduce another torque operator that can be used in the torque-correlation formula (2) and to discuss its properties. This operator is due to intersite electron hopping, and it is consequently nonlocal; in contrast to the local XC- and SO-induced torques which are random in random crystalline alloys, the nonlocal torque is nonrandom, i.e., independent of the particular configuration of a random alloy, which simplifies the configuration averaging of Eq. (2). We show that a similar nonlocal effective torque appears in the fully relativistic linear muffin-tin orbital (LMTO) method in the atomic-sphere approximation (ASA) used recently for calculations of the conductivity tensor in spin-polarized random alloys [27,28]. Here we discuss theoretical aspects of the averaging in the coherent-potential approximation (CPA) [29,30] and illustrate the developed *ab initio* scheme by applications to selected binary alloys. We also compare the obtained results with those of the LMTO-supercell technique [17] and with other CPA-based techniques, the fully relativistic Korringa-Kohn-Rostoker (KKR) method [19,22] and the LMTO method with a simplified treatment of the SO interaction [20].

This paper is organized as follows. The theoretical formalism is contained in Sec. II, with a general discussion of various torque operators and results of a simple tight-binding model presented in Sec. II A. Section II B describes the derivation of the LMTO torque-correlation formula with nonlocal torques; technical details are left to Appendix A concerning linear-response calculations with varying basis sets and to Appendix B regarding the LMTO method for systems with a tilted magnetization direction. Selected formal properties of the developed theory are discussed in Sec. II C. Applications of the developed formalism can be found in Sec. III. Details of numerical implementation are listed in Sec. III A, followed by illustrating examples for systems of three different types: binary solid solutions of $3d$ transition metals in Sec. III B, pure iron with a simple model of random potential fluctuations in Sec. III C, and stoichiometric FePt alloys with a partial long-range order in Sec. III D. The main conclusions are summarized in Sec. IV.

II. THEORETICAL FORMALISM

A. Torque-correlation formula with alternative torque operators

The torque operators T_μ entering the torque-correlation formula (2) are closely related to components of the time derivative of electron spin. For spin-polarized systems described by means of an effective Schrödinger-Pauli one-electron Hamiltonian H, acting on two-component wave functions, the complete time derivative of the spin operator is given by the commutation relation $t_\mu = -i[\sigma_\mu/2, H]$, where $\hbar = 1$ is assumed and σ_μ ($\mu = x,y,z$) denote the Pauli spin matrices. Let us write the Hamiltonian as $H = H^{\rm p} + H^{\rm xc}$, where $H^{\rm p}$ includes all spin-independent terms and the SO interaction (Hamiltonian of a paramagnetic system), while $H^{\rm xc} = \mathbf{B}^{\rm xc}(\mathbf{r}) \cdot \boldsymbol{\sigma}$ denotes the XC term due to an effective magnetic field $\mathbf{B}^{\rm xc}(\mathbf{r})$. The complete time derivative (spin torque) can then be written as $t_\mu = t_\mu^{\rm so} + t_\mu^{\rm xc}$, where

$$t_\mu^{\rm so} = -i[\sigma_\mu/2, H^{\rm p}], \quad t_\mu^{\rm xc} = -i[\sigma_\mu/2, H^{\rm xc}]. \tag{3}$$

As discussed, e.g., in Ref. [15], the use of the complete torque t_μ in the torque-correlation formula (2) leads identically to zero; the correct Gilbert damping coefficients $\alpha_{\mu\nu}$ follow from Eq. (2) by using either the SO-induced torque $t_\mu^{\rm so}$ or the XC-induced torque $t_\mu^{\rm xc}$. Note that only transverse components (with respect to the easy axis of the ferromagnet) of the vectors $\mathbf{t}^{\rm so}$ and $\mathbf{t}^{\rm xc}$ are needed for the relevant part of the Gilbert damping tensor (2).

The equivalence of both torque operators (3) for the Gilbert damping can be extended. Let us consider a simple system described by a model tight-binding Hamiltonian H, written now as $H = H^{\rm loc} + H^{\rm nl}$, where the first term $H^{\rm loc}$ is a lattice sum of local atomiclike terms and the nonlocal second term $H^{\rm nl}$ includes all intersite hopping matrix elements. Let us assume that all effects of the SO interaction and XC fields are contained in the local term $H^{\rm loc}$, so that the hopping elements are spin independent and $[\sigma_\mu, H^{\rm nl}] = 0$. (Note that this assumption, often used in model studies, is satisfied only approximatively in real ferromagnets with different widths of the majority and minority spin bands.) Let us write explicitly $H^{\rm loc} = \sum_{\mathbf{R}} (H^{\rm p}_{\mathbf{R}} + H^{\rm xc}_{\mathbf{R}})$, where $\mathbf{R}$ labels the lattice sites and

where $H^{\rm p}_{\mathbf{R}}$ comprises the spin-independent part and the SO interaction of the **R**th atomic potential, while $H^{\rm xc}_{\mathbf{R}}$ is due to the local XC field of the **R**th atom. The operators $H^{\rm p}_{\mathbf{R}}$ and $H^{\rm xc}_{\mathbf{R}}$ act only in the subspace of the **R**th site; the subspaces of different sites are orthogonal to each other. The total spin operator can be written as $\sigma_\mu/2 = (1/2)\sum_{\mathbf{R}} \sigma_{\mathbf{R}\mu}$, where the local operator $\sigma_{\mathbf{R}\mu}$ is the projection of σ_μ on the **R**th subspace. Let us assume that each term $H^{\rm p}_{\mathbf{R}}$ is spherically symmetric and that $H^{\rm xc}_{\mathbf{R}} = \mathbf{B}^{\rm xc}_{\mathbf{R}} \cdot \boldsymbol{\sigma}_{\mathbf{R}}$, where the effective field $\mathbf{B}^{\rm xc}_{\mathbf{R}}$ of the **R**th atom has a constant size and direction. Let us introduce local orbital-momentum operators $L_{\mathbf{R}\mu}$ and their counterparts including the spin, $J_{\mathbf{R}\mu} = L_{\mathbf{R}\mu} + (\sigma_{\mathbf{R}\mu}/2)$, which are generators of local infinitesimal rotations with respect to the **R**th lattice site, and let us define the corresponding lattice sums $L_\mu = \sum_{\mathbf{R}} L_{\mathbf{R}\mu}$ and $J_\mu = \sum_{\mathbf{R}} J_{\mathbf{R}\mu} = L_\mu + (\sigma_\mu/2)$. Then the local terms $H^{\rm p}_{\mathbf{R}}$ and $H^{\rm xc}_{\mathbf{R}}$ satisfy, respectively, commutation rules $[J_{\mathbf{R}\mu}, H^{\rm p}_{\mathbf{R}}] = 0$ and $[L_{\mathbf{R}\mu}, H^{\rm xc}_{\mathbf{R}}] = 0$. By using the above assumptions and definitions, the XC-induced spin torque (3) due to the XC term $H^{\rm xc} = \sum_{\mathbf{R}} H^{\rm xc}_{\mathbf{R}}$ can be reformulated as

$$t^{\rm xc}_\mu = -i\sum_{\mathbf{R}} \left[\sigma_{\mathbf{R}\mu}/2, H^{\rm xc}_{\mathbf{R}}\right] = -i\sum_{\mathbf{R}} \left[J_{\mathbf{R}\mu}, H^{\rm xc}_{\mathbf{R}}\right]$$
$$= -i\sum_{\mathbf{R}} \left[J_{\mathbf{R}\mu}, H^{\rm p}_{\mathbf{R}} + H^{\rm xc}_{\mathbf{R}}\right] = -i[J_\mu, H^{\rm loc}] \equiv t^{\rm loc}_\mu. \quad (4)$$

The last commutator defines a local torque operator $t^{\rm loc}_\mu$ due to the local part of the Hamiltonian $H^{\rm loc}$ and the operator J_μ, in contrast to the spin operator $\sigma_\mu/2$ in Eq. (3). Let us define the complementary nonlocal torque $t^{\rm nl}_\mu$ due to the nonlocal part of the Hamiltonian $H^{\rm nl}$, namely,

$$t^{\rm nl}_\mu = -i[J_\mu, H^{\rm nl}] = -i[L_\mu, H^{\rm nl}], \quad (5)$$

and let us employ the fact that the complete time derivative of the operator J_μ, i.e., the torque $\tilde{t}_\mu = -i[J_\mu, H] = t^{\rm loc}_\mu + t^{\rm nl}_\mu$, leads identically to zero when used in Eq. (2). This fact implies that the Gilbert damping parameters can be also obtained from the torque-correlation formula with the nonlocal torques $t^{\rm nl}_\mu$. These torques are equivalent to the original spin-dependent local XC- or SO-induced torques; however, the derived nonlocal torques are spin independent, so that commutation rules $[t^{\rm nl}_\mu, \sigma_\nu] = 0$ are satisfied.

In order to see the effect of different forms of the torque operators, Eqs. (3) and (5), we have studied a tight-binding model of p orbitals on a simple cubic lattice with the ground-state magnetization along the z axis. The local (atomiclike) terms of the Hamiltonian are specified by the XC term $b\sigma_{\mathbf{R}z}$ and the SO term $\xi \mathbf{L}_{\mathbf{R}} \cdot \boldsymbol{\sigma}_{\mathbf{R}}$, which are added to a random spin-independent p level at energy $\epsilon_0 + D_{\mathbf{R}}$, where ϵ_0 denotes the nonrandom center of the p band, while the random parts $D_{\mathbf{R}}$ satisfy configuration averages $\langle D_{\mathbf{R}}\rangle = 0$ and $\langle D_{\mathbf{R}'} D_{\mathbf{R}}\rangle = \gamma\delta_{\mathbf{R}'\mathbf{R}}$ with the disorder strength γ. The spin-independent nonlocal (hopping) part of the Hamiltonian has been confined to nonrandom nearest-neighbor hoppings parametrized by two quantities, W_1 ($pp\sigma$ hopping) and W_1' ($pp\pi$ hopping) (see, e.g., Ref. [31]). The particular values have been set to $b = 0.3$, $\xi = 0.2$, $E_{\rm F} - \epsilon_0 = 0.1$, $\gamma = 0.05$, $W_1 = 0.3$, and $W_1' = -0.1$ (the hoppings were chosen such that the band edges for $\epsilon_0 = b = \xi = \gamma = 0$ are ± 1). The configuration average of the propagators $\langle G_\pm\rangle = \bar{G}_\pm$ and of the torque correlation (2) was performed in the self-consistent Born approximation (SCBA) including the vertex corrections. Since all three torques, Eqs. (3) and (5), are nonrandom operators in our model, the only relevant component of the Gilbert damping tensor, namely, $\alpha_{xx} = \alpha_{yy} = \alpha$, could be unambiguously decomposed into the coherent part $\alpha^{\rm coh}$ and the incoherent part $\alpha^{\rm vc}$ due to the vertex corrections.

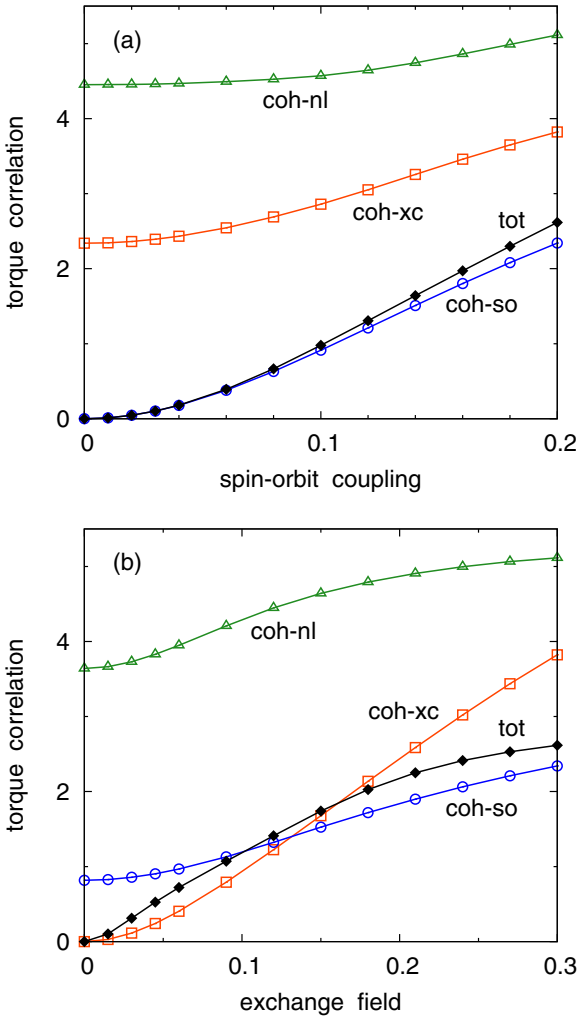

FIG. 1. (Color online) The torque correlation α/α_0, Eq. (2), in a tight-binding p-orbital model treated in the SCBA as a function (a) of the spin-orbit coupling ξ and (b) of the exchange field b. The diamonds display the total torque correlation (tot), and the open symbols denote the coherent contributions $\alpha^{\rm coh}/\alpha_0$ calculated with the SO-induced torque (coh-so); the XC-induced torque (coh-xc), Eq. (3); and the nonlocal torque (coh-nl), Eq. (5).

The results are summarized in Fig. 1, which displays the torque correlation α/α_0 as a function of the SO coupling ξ [Fig. 1(a)] and the XC field b [Fig. 1(b)]. The total value $\alpha = \alpha^{\rm coh} + \alpha^{\rm vc}$ is identical for all three forms of the torque operator, in contrast to the coherent parts $\alpha^{\rm coh}$, which exhibit markedly different values and trends when compared to each other and to the total α. This result is in line with conclusions drawn by the authors of Refs. [15,16,26], proving the importance of the vertex corrections for obtaining the same Gilbert damping parameters from the SO- and XC-induced

torques. The only exception seems to be the case of the SO splitting being much weaker than the exchange splitting, where the vertex corrections for the SO-induced torque can be safely neglected [see Fig. 1(a)]. This situation, encountered in $3d$ transition metals and their alloys, has been treated with the SO-induced torque on an *ab initio* level with neglected vertex corrections in Refs. [11,12]. On the other hand, the use of the XC-induced torque calls for a proper evaluation of the vertex corrections; their neglect leads to quantitatively and physically incorrect results, as documented by recent first-principles studies [19,22]. The vertex corrections are also indispensable for the nonlocal torque, in particular for correct vanishing of the total torque correlation both in the nonrelativistic limit [$\xi \to 0$, Fig. 1(a)] and in the nonmagnetic limit [$b \to 0$, Fig. 1(b)].

Finally, let us discuss briefly the general equivalence of the SO- and XC-induced spin torques, Eq. (3), in the fully relativistic four-component Dirac formalism [32,33]. The Kohn-Sham-Dirac Hamiltonian can be written as $H = H^{\rm p} + H^{\rm xc}$, where $H^{\rm p} = c\boldsymbol{\alpha}\cdot\mathbf{p} + mc^2\beta + V(\mathbf{r})$ and $H^{\rm xc} = \mathbf{B}^{\rm xc}(\mathbf{r})\cdot\beta\boldsymbol{\Sigma}$, where c is the speed of light, m denotes the electron mass, $\mathbf{p} = \{p_\mu\}$ refers to the momentum operator, $V(\mathbf{r})$ is the spin-independent part of the effective potential, and $\boldsymbol{\alpha} = \{\alpha_\mu\}$, β, and $\boldsymbol{\Sigma} = \{\Sigma_\mu\}$ are the well-known 4×4 matrices of the Dirac theory [34,35]. Then the XC-induced torque is $\mathbf{t}^{\rm xc} = \mathbf{B}^{\rm xc}(\mathbf{r})\times\beta\boldsymbol{\Sigma}$, which is currently used in the KKR theory of the Gilbert damping [19,22]. The SO-induced torque is $\mathbf{t}^{\rm so} = \mathbf{p}\times c\boldsymbol{\alpha}$; that is, it is given directly by the relativistic momentum ($\mathbf{p}$) and velocity ($c\boldsymbol{\alpha}$) operators. One can see that the torque $\mathbf{t}^{\rm so}$ is local but independent of the particular system studied. A comparison of both alternatives, concerning the total damping parameters as well as their coherent and incoherent parts, would be desirable; however, this task is beyond the scope of the present study.

B. Effective torques in the LMTO method

In our *ab initio* approach to the Gilbert damping, we employ the torque-correlation formula (2) with torques derived from the XC field [15,19,22]. The torque operators are constructed by considering infinitesimal deviations of the direction of the XC field of the ferromagnet from its equilibrium orientation, taken as a reference state. These deviations result from rotations by small angles around axes perpendicular to the equilibrium direction of the XC field; components of the torque operator are then given as derivatives of the one-particle Hamiltonian with respect to the rotation angles [36].

For practical evaluation of Eq. (2) in an *ab initio* technique (such as the LMTO method), one has to consider a matrix representation of all operators in a suitable orthonormal basis. The most efficient techniques of the electronic structure theory typically require basis vectors tailored to the system studied; in the present context, this leads naturally to basis sets depending on the angular variables needed to define the torque operators. Evaluation of the torque correlation using angle-dependent bases is discussed in Appendix A, where we prove that Eq. (2) can be calculated solely from the matrix elements of the Hamiltonian and their angular derivatives [see Eq. (A7)], whereas the angular dependence of the basis vectors does not contribute directly to the final result.

The relativistic LMTO-ASA Hamiltonian matrix for the reference system in the orthogonal LMTO representation is given by [37–39]

$$H = C + (\sqrt{\Delta})^{+} S(1-\gamma S)^{-1}\sqrt{\Delta}, \tag{6}$$

where C, $\sqrt{\Delta}$, and γ denote site-diagonal matrices of the standard LMTO potential parameters and S is the matrix of canonical structure constants. The change of the Hamiltonian matrix H due to a uniform rotation of the XC field is treated in Appendix B; it is summarized for finite rotations in Eq. (B7) and for angular derivatives of H in Eqs. (B8) and (B9). The resolvent $G(z) = (z-H)^{-1}$ of the LMTO Hamiltonian (6) for complex energies z can be expressed using the auxiliary resolvent $g(z) = [P(z)-S]^{-1}$, which represents an LMTO counterpart of the scattering-path operator matrix of the KKR method [32,33]. The symbol $P(z)$ denotes the site-diagonal matrix of potential functions; their analytic dependence on z and on the potential parameters can be found elsewhere [27,37]. The relation between both resolvents leads to the formula [28]

$$G_{+} - G_{-} = F(g_{+} - g_{-})F^{+}, \tag{7}$$

where the same abbreviation $F = (\sqrt{\Delta})^{-1}(1-\gamma S)$ as in Eq. (B8) was used and $g_{\pm} = g(E_{\rm F} \pm i0)$.

The torque-correlation formula (2) in the LMTO-ASA method follows directly from relations (A7), (B8), (B9), and (7). The components of the Gilbert damping tensor $\{\alpha_{\mu\nu}\}$ in the LLG equation (1) can be obtained from a basic tensor $\{\tilde{\alpha}_{\mu\nu}\}$ given by

$$\tilde{\alpha}_{\mu\nu} = -\alpha_0\,{\rm Tr}\{\tau_\mu(g_{+}-g_{-})\tau_\nu(g_{+}-g_{-})\}, \tag{8}$$

where the quantities

$$\tau_\mu = -i[\mathcal{J}^\mu, S] = -i[\mathcal{L}^\mu, S] \tag{9}$$

define components of an effective torque in the LMTO-ASA method. The site-diagonal matrices $\mathcal{J}^\mu$ and $\mathcal{L}^\mu$ ($\mu = x,y,z$) are Cartesian components of the total and orbital angular momentum operators, respectively; see text discussion around Eqs. (B8) and (B9). The trace in (8) extends over all orbitals of the crystalline solid, and the prefactor can be written as $\alpha_0 = (2\pi M_{\rm spin})^{-1}$, where $M_{\rm spin}$ denotes the spin magnetic moment of the whole crystal in units of the Bohr magneton $\mu_{\rm B}$ [15,19,22].

Let us discuss properties of the effective torque (9). Its form is obviously identical to the nonlocal torque (5). The matrix τ_μ is not site diagonal, but for a random substitutional alloy on a nonrandom lattice, it is nonrandom (independent of the alloy configuration). Moreover, it is given by a commutator of the site-diagonal nonrandom matrix $\mathcal{J}^\mu$ (or $\mathcal{L}^\mu$) and the LMTO structure-constant matrix S. These properties point to a close analogy between the effective torque and the effective velocities in the LMTO conductivity tensor based on the concept of intersite electron hopping [27,28,40]. Let us mention that existing *ab initio* approaches employ random torques, either the XC-induced torque in the KKR method [19,22] or the SO-induced torque in the LMTO method [20]. Another interesting property of the effective torque τ_μ (9) is its spin independence, which follows from the spin independence of the matrices $\mathcal{L}^\mu$ and S.

The explicit relation between the symmetric tensors $\{\alpha_{\mu\nu}\}$ and $\{\tilde{\alpha}_{\mu\nu}\}$ can easily be formulated for the ground-state magnetization along the z axis; then it is given simply by $\alpha_{xx} = \tilde{\alpha}_{yy}, \alpha_{yy} = \tilde{\alpha}_{xx}$, and $\alpha_{xy} = -\tilde{\alpha}_{xy}$. These relations reflect the fact that an infinitesimal deviation towards the x axis results from an infinitesimal rotation of the magnetization vector around the y axis and vice versa. Note that the other components of the Gilbert damping tensor ($\alpha_{\mu z}$ for $\mu = x,y,z$) are not relevant for the dynamics of small deviations of the magnetization direction described by the LLG equation (1). For the ground-state magnetization pointing along a general unit vector $\mathbf{m} = (m_x,m_y,m_z)$, one has to employ the Levi-Civita symbol $\epsilon_{\mu\nu\lambda}$ in order to get the Gilbert damping tensor $\underline{\boldsymbol{\alpha}}$ as

$$\alpha_{\mu\nu} = \sum_{\mu'\nu'} \eta_{\mu\mu'}\eta_{\nu\nu'}\tilde{\alpha}_{\mu'\nu'}, \tag{10}$$

where $\eta_{\mu\nu} = \sum_\lambda \epsilon_{\mu\nu\lambda}m_\lambda$. The resulting tensor (10) satisfies the condition $\underline{\boldsymbol{\alpha}} \cdot \mathbf{m} = 0$ appropriate for the dynamics of small transverse deviations of magnetization.

The application to random alloys requires configuration averaging of $\tilde{\alpha}_{\mu\nu}$ (8). Since the effective torques τ_μ are nonrandom, one can write a unique decomposition of the average into the coherent and incoherent parts, $\tilde{\alpha}_{\mu\nu} = \tilde{\alpha}^{\rm coh}_{\mu\nu} + \tilde{\alpha}^{\rm vc}_{\mu\nu}$, where the coherent part is expressed by means of the averaged auxiliary resolvents $\bar{g}_\pm = \langle g_\pm \rangle$ as

$$\tilde{\alpha}^{\rm coh}_{\mu\nu} = -\alpha_0 {\rm Tr}\{\tau_\mu(\bar{g}_+ - \bar{g}_-)\tau_\nu(\bar{g}_+ - \bar{g}_-)\} \tag{11}$$

and the incoherent part (vertex corrections) is given as a sum of four terms, namely,

$$\tilde{\alpha}^{\rm vc}_{\mu\nu} = -\alpha_0 \sum_{p=\pm}\sum_{q=\pm} {\rm sgn}(pq){\rm Tr}\langle \tau_\mu g_p \tau_\nu g_q \rangle_{\rm vc}. \tag{12}$$

In this work, the configuration averaging has been done in the CPA. Details concerning the averaged resolvents can be found, e.g., in Ref. [39], and the construction of the vertex corrections for transport properties was described in the Appendix to Ref. [30].

C. Properties of the LMTO torque-correlation formula

The damping tensor (8) has been formulated in the canonical LMTO representation. In the numerical implementation, the well-known transformation to a tight-binding (TB) LMTO representation [41,42] is advantageous. The TB-LMTO representation is specified by a diagonal matrix β of spin-independent screening constants ($\beta_{\mathbf{R}'\ell'm's',\mathbf{R}\ell ms} = \delta_{\mathbf{R}'\mathbf{R}}\delta_{\ell'\ell}\delta_{m'm}\delta_{s's}\beta_{\mathbf{R}\ell}$ in a nonrelativistic basis), and the transformation of all quantities between both LMTO representations has been discussed in the literature for pure crystals [42] and for random alloys [28,39,43]. The same techniques can be used in the present case together with an obvious commutation rule $[\mathcal{J}^\mu,\beta] = [\mathcal{L}^\mu,\beta] = 0$. Consequently, the conclusions drawn are the same as for the conductivity tensor [28]: the total damping tensor (8) and its coherent (11) and incoherent (12) parts in the CPA are invariant with respect to the choice of the LMTO representation.

It should be mentioned that the central result, namely, relations (8) and (9), is not limited to the LMTO theory, and it can be translated into the KKR theory as well, similar to the conductivity tensor in the formalism of intersite hopping [40]. The LMTO structure-constant matrix S and the auxiliary Green's function $g(z)$ will then be replaced, respectively, by the KKR structure-constant matrix and by the scattering-path operator [32,33]. Note, however, that the total ($\mathcal{J}^\mu$) and orbital ($\mathcal{L}^\mu$) angular momentum operators in the effective torques (9) will be represented by the same matrices as in the LMTO theory.

Let us mention for completeness that the present LMTO-ASA theory allows one to introduce effective local (but random) torques as well. This is based on the fact that only the Fermi-level propagators $g_\pm$ defined by the structure constant matrix S and by the potential functions at the Fermi energy, $P = P(E_{\rm F})$, enter the zero-temperature expression for the damping tensor $\tilde{\alpha}_{\mu\nu}$ (8). Since the equation of motion $(P - S)g_\pm = 1$ implies immediately $S(g_+ - g_-) = P(g_+ - g_-)$ and, similarly, $(g_+ - g_-)S = (g_+ - g_-)P$, one can obviously replace the nonlocal torques τ_μ (9) in the torque-correlation formula (8) by their local counterparts

$$\tau^{\rm xc}_\mu = i[P,\mathcal{J}^\mu], \quad \tau^{\rm so}_\mu = i[P,\mathcal{L}^\mu]. \tag{13}$$

These effective torques are represented by random, site-diagonal matrices; $\tau^{\rm xc}_\mu$ and $\tau^{\rm so}_\mu$ correspond, respectively, to the XC-induced torque used in the KKR method [22] and to the SO-induced torque used in the LMTO method with a simplified treatment of the SO interaction [20]. In the case of random alloys treated in the CPA, the randomness of the local torques (13) calls for the approach developed by Butler [44] for the averaging of the torque-correlation coefficient (8). One can prove that the resulting damping parameters $\tilde{\alpha}_{\mu\nu}$ obtained in the CPA with the local and nonlocal torques are fully equivalent to each other; this equivalence rests heavily on a proper inclusion of the vertex corrections [45], and it leads to further important consequences. First, the Gilbert damping tensor vanishes exactly for zero SO interaction, which follows from the use of the SO-induced torque $\tau^{\rm so}_\mu$ and from the obvious commutation rule $[P,\mathcal{L}^\mu] = 0$ that is valid for the spherically symmetric potential functions (in the absence of SO interaction). This result is in agreement with the numerical study of the toy model in Sec. II A [see Fig. 1(a) for $\xi = 0$]. On an *ab initio* level, this property has been obtained numerically both in the KKR method [22] and in the LMTO method [26]. Second, the XC- and SO-induced local torques (13) within the CPA are exactly equivalent as well, as has been indicated in a recent numerical study for a random bcc $Fe_{50}Co_{50}$ alloy [26]. In summary, the nonlocal torques (9) and both local torques (13) can be used as equivalent alternatives in the torque-correlation formula (8) provided that the vertex corrections are included consistently with the CPA averaging of the single-particle propagators.

III. ILLUSTRATING EXAMPLES

A. Implementation and numerical details

The numerical implementation of the described theory and the calculations have been done with tools similar to those in our recent studies of ground-state [46] and transport [27,28,47] properties. The ground-state magnetization was taken along the z axis, and the self-consistent XC potentials were obtained in the local-spin-density approximation

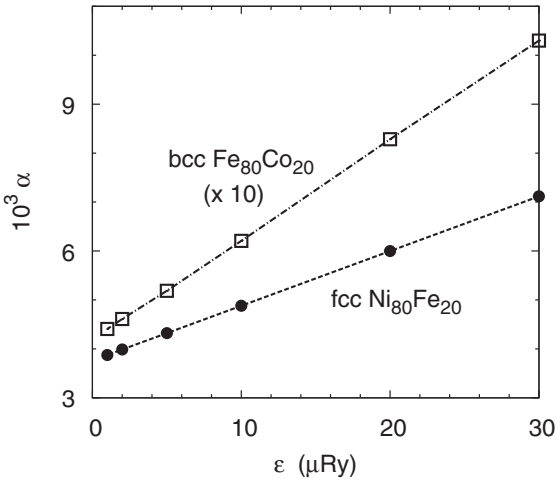

FIG. 2. The Gilbert damping parameters α of random fcc $Ni_{80}Fe_{20}$ (circles) and bcc $Fe_{80}Co_{20}$ (squares) alloys as functions of the imaginary part of energy ε. The values of α for the $Fe_{80}Co_{20}$ alloy are magnified by a factor of 10.

(LSDA) with parametrization according to Ref. [48]. The valence basis comprised s-, p-, and d-type orbitals, and the energy arguments for the propagators $\bar{g}_{\pm}$ and the CPA-vertex corrections were obtained by adding a tiny imaginary part $\pm\varepsilon$ to the real Fermi energy. We have found that the dependence of the Gilbert damping parameter on ε is quite smooth and that the value of $\varepsilon = 10^{-6}$ Ry is sufficient for the studied systems; see Fig. 2 for an illustration. Similar smooth dependences have also been obtained for other investigated alloys, such as permalloy doped by $5d$ elements, Heusler alloys, and stoichiometric FePt alloys with a partial atomic long-range order. In all studied cases, the number N of $\mathbf{k}$ vectors needed for reliable averaging over the Brillouin zone (BZ) was properly checked; as a rule, $N \sim 10^8$ in the full BZ was sufficient for most systems, but for diluted alloys (a few percent of impurities), $N \sim 10^9$ had to be taken.

B. Binary fcc and bcc solid solutions

The developed theory has been applied to random binary alloys of $3d$ transition elements Fe, Co, and Ni, namely, to the fcc NiFe and bcc FeCo alloys. The most important results, including a comparison to other existing *ab initio* techniques, are summarized in Fig. 3. One can see the good agreement of the calculated concentration trends of the Gilbert damping parameter $\alpha = \alpha_{xx} = \alpha_{yy}$ with the results of an LMTO-supercell approach [17] and of the KKR-CPA method [22]. The decrease of α with increasing Fe content in the concentrated NiFe alloys can be related to the increasing alloy magnetization [17] and to the decreasing strength of the SO interaction [20], whereas the behavior in the dilute limit can be explained by intraband scattering due to Fe impurities [11,12,14]. In the case of the FeCo system, the minimum of α around 20% Co, which is also observed in room-temperature experiments [49,50], is related primarily to a similar concentration trend of the density of states at the Fermi energy [22], although the maximum of the magnetization at roughly the same alloy composition [51] might partly contribute as well.

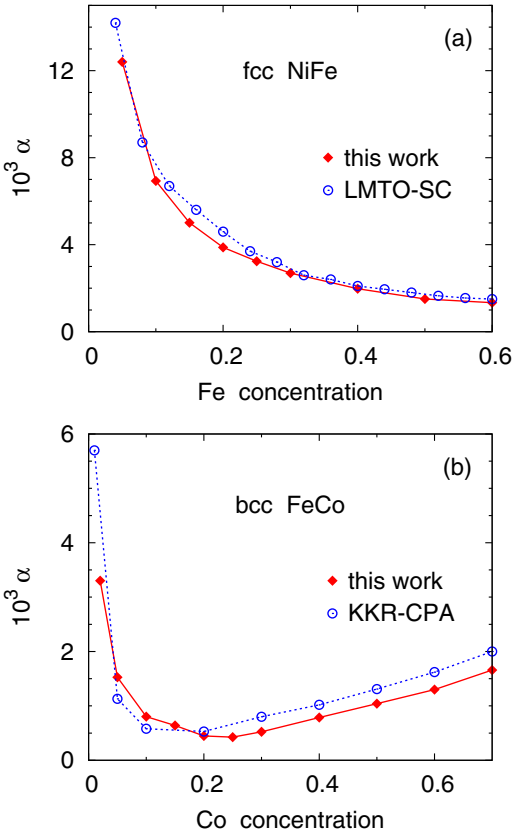

FIG. 3. (Color online) The calculated concentration dependences of the Gilbert damping parameter α for (a) random fcc NiFe and (b) bcc FeCo alloys. The results of this work are marked by the diamonds, whereas the circles depict the results of other approaches: the LMTO supercell (LMTO-SC) technique [17] and the KKR-CPA method [22].

A more detailed comparison of all *ab initio* results is presented in Table I for the fcc $Ni_{80}Fe_{20}$ random alloy (permalloy). The differences in the values of α from the different techniques can be ascribed to various theoretical

TABLE I. Comparison of the Gilbert damping parameter α for the fcc $Ni_{80}Fe_{20}$ random alloy (permalloy) calculated by the present approach and by other techniques using the CPA or supercells (SC). The last column displays the coherent part α^{coh} of the total damping parameter according to Eq. (11). The experimental value corresponds to room temperature.

Method	α	α^{coh}
This work, $\varepsilon = 10^{-5}$ Ry	4.9×10^{-3}	1.76
This work, $\varepsilon = 10^{-6}$ Ry	3.9×10^{-3}	1.76
KKR-CPA[a]	4.2×10^{-3}	
LMTO-CPA[b]	3.5×10^{-3}	
LMTO-SC[c]	4.6×10^{-3}	
Experiment[d]	8×10^{-3}	

[a]Reference [22].
[b]Reference [20].
[c]Reference [17].
[d]Reference [49].

features and numerical details employed, such as the simplified treatment of the SO interaction in Ref. [20] instead of the fully relativistic description or the use of supercells in Ref. [17] instead of the CPA. Taking into account that calculated residual resistivities for this alloy span a wide interval between 2 $\mu\Omega$ cm (see Refs. [27,52]) and 3.5 $\mu\Omega$ cm (see Ref. [17]), one can consider the scatter of the calculated values of α in Table I as having little importance. The theoretical values of α are smaller systematically than the measured values, typically by a factor of 2. This discrepancy might be partly due to the effects of finite temperatures as well as due to additional structural defects of real samples.

A closer look at the theoretical results reveals that the total damping parameters α are appreciably smaller than the magnitudes of their coherent and vertex parts (see Table I for the case of permalloy). This is in agreement with the results of the model study in Sec. II A; similar conclusions about the importance of the vertex corrections have been done with the XC-induced torques in other CPA-based studies [19,22,26]. The present results prove that this unpleasant feature of the nonlocal torques does not represent a serious obstacle in obtaining reliable values of the Gilbert damping parameter in random alloys. We note that the vertex corrections can be negligible in approaches employing the SO-induced torques, at least for systems with SO splittings much weaker than the XC splittings [12], such as the binary ferromagnetic alloys of 3*d* transition metals [26] (see also Sec. II A).

C. Pure iron with a model disorder

As mentioned in Sec. I, the Gilbert damping of pure ferromagnetic metals exhibits nontrivial temperature dependences, which have been reproduced by means of *ab initio* techniques with various levels of sophistication [11,12,21,23]. In this study, we have simulated the effect of finite temperatures by introducing static fluctuations of the one-particle potential. The adopted model of atomic-level disorder assumes that random spin-independent shifts $\pm\delta$, constant inside each atomic sphere and occurring with probabilities of 50% for both signs, are added to the nonrandom self-consistent potential obtained at zero temperature. The Fermi energy is kept frozen, equal to its self-consistent zero-temperature value. This model can be easily treated in the CPA; the resulting Gilbert damping parameter α of pure bcc Fe as a function of the potential shift δ is plotted in Fig. 4.

The calculated dependence $\alpha(\delta)$ is nonmonotonic, with a minimum at $\delta \approx 30$ mRy. This trend is in qualitative agreement with trends reported previously by other authors, who employed phenomenological models of the electron lifetime [11,12] as well as models for phonons and magnons [21,23]. The origin of the nonmonotonic dependence $\alpha(\delta)$ has been identified on the basis of the band structure of the ferromagnetic system as an interplay between the intraband contributions to α, dominating for small values of δ, and the interband contributions, dominating for large values of δ [7,11,12]. Since the present CPA-based approach does not use any bands, we cannot perform a similar analysis.

The obtained minimum value of the Gilbert damping, $\alpha_{\min} \approx 10^{-3}$ (Fig. 4), agrees reasonably well with the values obtained by the authors of Refs. [11,12,21,23]. This agreement

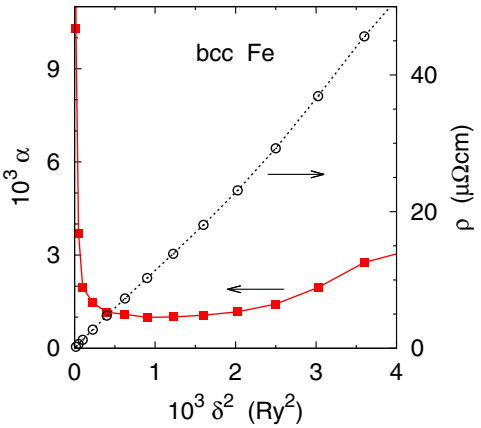

FIG. 4. (Color online) The calculated Gilbert damping parameter α (squares) and the residual resistivity ρ (circles) of pure bcc iron as functions of δ^2, where δ is the strength of a model atomic-level disorder.

indicates that the atomic-level disorder employed here is equivalent to a phenomenological lifetime broadening. For a rough quantitative estimation of the temperature effect, one can employ the calculated resistivity ρ of the model, which increases essentially linearly with δ^2 (see Fig. 4). Since the metallic resistivity due to phonons increases linearly with the temperature T (for temperatures not much smaller than the Debye temperature), one can assume a proportionality between δ^2 and T. The resistivity of bcc iron at the Curie temperature $T_C = 1044$ K due to lattice vibrations can be estimated around 35 $\mu\Omega$ cm [23,53], which sets an approximate temperature scale to the data plotted in Fig. 4. However, a more accurate description of the temperature dependence of the Gilbert damping parameter cannot be obtained, mainly due to the neglected true atomic displacements and the noncollinearity of magnetic moments (magnons) [23].

D. FePt alloys with a partial long-range order

Since important ferromagnetic materials include ordered alloys, we address here the Gilbert damping in stoichiometric FePt alloys with $L1_0$ atomic long-range order (LRO). Their transport properties [47] and the damping parameter [20] have recently been studied by means of the TB-LMTO method depending on a varying degree of the LRO. These fcc-based systems contain two sublattices with respective occupations $Fe_{1-y}Pt_y$ and $Pt_{1-y}Fe_y$, where y $(0 \leqslant y \leqslant 0.5)$ denotes the concentration of antisite atoms. The LRO parameter S $(0 \leqslant S \leqslant 1)$ is then defined as $S = 1 - 2y$, so that $S = 0$ corresponds to the random fcc alloy and $S = 1$ corresponds to the perfectly ordered $L1_0$ structure.

The resulting Gilbert damping parameter is displayed in Fig. 5 as a function of S. The obtained trend with a broad maximum at $S = 0$ and a minimum around $S = 0.9$ agrees very well with the previous result [20]. The values of α in Fig. 5 are about 10% higher than those in Ref. [20], which can be ascribed to the fully relativistic treatment in the present study in contrast to a simplified treatment of the SO interaction in Ref. [20]. The Gilbert damping in the FePt alloys is an order of magnitude stronger than in the alloys of 3*d* elements

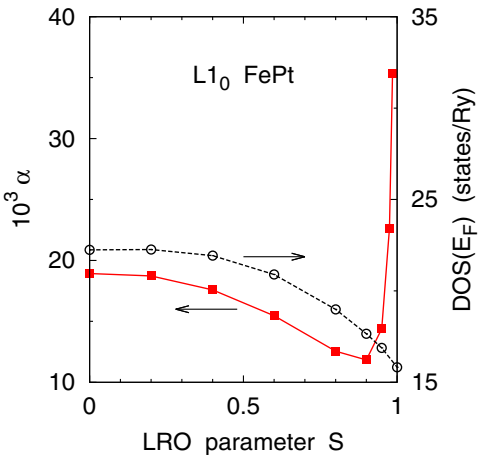

FIG. 5. (Color online) The calculated Gilbert damping parameter α (squares) and the total DOS (per formula unit) at the Fermi energy (circles) of stoichiometric $L1_0$ FePt alloys as functions of the LRO parameter S.

(Sec. III B) owing to the stronger SO interaction of Pt atoms. The origin of the slow decrease of α with increasing S (for $0 \leqslant S \leqslant 0.9$) can be explained by the decreasing total density of states (DOS) at the Fermi energy (see Fig. 5), which represents an analogy to a similar correlation observed, e.g., for bcc FeCo alloys [22].

All calculated values of α shown in Fig. 5, corresponding to $0 \leqslant S \leqslant 0.985$, are appreciably smaller than the measured one, which amounts to $\alpha \approx 0.06$ reported for a thin $L1_0$ FePt epitaxial film [54]. The high measured value of α might be thus explained by the present calculations by assuming a very small concentration of antisites in the prepared films, which does not seem too realistic. Another potential source of the discrepancy lies in the thin-film geometry used in the experiment. Moreover, the divergence of α in the limit of $S \to 1$ (Fig. 5) illustrates a general shortcoming of approaches based on the torque-correlation formula (2) since the zero-temperature Gilbert damping parameter of a pure ferromagnet should remain finite. A correct treatment of this case, including the dilute limit of random alloys (Fig. 3), must take into account the full interacting susceptibility in the presence of SO interaction [15,55]. Pilot *ab initio* studies in this direction have recently appeared for nonrandom systems [56,57]; however, their extension to disordered systems goes far beyond the scope of this work.

IV. CONCLUSIONS

We have introduced nonlocal torques as an alternative to the usual local torque operators entering the torque-correlation formula for the Gilbert damping tensor. Within the relativistic TB-LMTO-ASA method, this idea leads to effective nonlocal torques as non-site-diagonal and spin-independent matrices. For substitutionally disordered alloys, the nonlocal torques are nonrandom, which allows one to develop an internally consistent theory in the CPA. The CPA-vertex corrections proved indispensable for an exact equivalence of the nonlocal nonrandom torques with their local random counterparts. The concept of the nonlocal torques is not limited to the LMTO method, and its formulation both in a semiempirical TB theory and in the KKR theory is straightforward.

The numerical implementation and the results for binary solid solutions show that the total Gilbert damping parameters from the nonlocal torques are much smaller than the magnitudes of the coherent parts and of the vertex corrections. Nevertheless, the total damping parameters for the studied NiFe, FeCo, and FePt alloys compare quantitatively very well with results of other *ab initio* techniques [17,20,22], which indicates a fair numerical stability of the developed theory.

The performed numerical study of the Gilbert damping in pure bcc iron as a function of an atomic-level disorder yields a nonmonotonic dependence in qualitative agreement with the trends consisting of the conductivity-like and resistivity-like regions, obtained from a phenomenological quasiparticle lifetime broadening [7,11,12] or from the temperature-induced frozen phonons [21,22] and magnons [23]. Future studies should clarify the applicability of the introduced nonlocal torques to a full quantitative description of the finite-temperature behavior as well as to other torque-related phenomena, such as the spin-orbit torques due to applied electric fields [58,59].

ACKNOWLEDGMENT

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APPENDIX A: TORQUE CORRELATION FORMULA IN A MATRIX REPRESENTATION

In this Appendix, the evaluation of the Kubo-Greenwood expression for the torque-correlation formula (2) is discussed in the case of the XC-induced torque operators using matrix representations of all operators in an orthonormal basis that varies due to the varying direction of the XC field. All operators are denoted by a hat in order to distinguish them from matrices representing these operators in the chosen basis. Let us consider a one-particle Hamiltonian $\hat{H} = \hat{H}(\theta_1,\theta_2)$ depending on two real variables θ_j, $j = 1,2$, and let us denote $\hat{T}^{(j)}(\theta_1,\theta_2) = \partial\hat{H}(\theta_1,\theta_2)/\partial\theta_j$. In our case, the variables θ_j play the role of rotation angles, and the operators $\hat{T}^{(j)}$ are the corresponding torques. Let us denote the resolvents of $\hat{H}(\theta_1,\theta_2)$ at the Fermi energy as $\hat{G}_\pm(\theta_1,\theta_2)$, and let us consider a special linear response coefficient (arguments θ_1 and θ_2 are omitted here and below for brevity)

$$\begin{aligned} c &= \mathrm{Tr}\{\hat{T}^{(1)}(\hat{G}_+ - \hat{G}_-)\hat{T}^{(2)}(\hat{G}_+ - \hat{G}_-)\} \\ &= \mathrm{Tr}\{(\partial\hat{H}/\partial\theta_1)(\hat{G}_+ - \hat{G}_-)(\partial\hat{H}/\partial\theta_2)(\hat{G}_+ - \hat{G}_-)\}. \end{aligned} \quad \text{(A1)}$$

This torque-correlation coefficient equals the Gilbert damping parameter (2) with the prefactor $(-\alpha_0)$ suppressed. For its evaluation, we introduce an orthonormal basis $|\chi_m(\theta_1,\theta_2)\rangle$ and represent all operators in this basis. This leads to matrices $H(\theta_1,\theta_2) = \{H_{mn}(\theta_1,\theta_2)\}$, $G_\pm(\theta_1,\theta_2) = \{(G_\pm)_{mn}(\theta_1,\theta_2)\}$, and $T^{(j)}(\theta_1,\theta_2) = \{T^{(j)}_{mn}(\theta_1,\theta_2)\}$, where

$$\begin{aligned} H_{mn} &= \langle\chi_m|\hat{H}|\chi_n\rangle, \quad (G_\pm)_{mn} = \langle\chi_m|\hat{G}_\pm|\chi_n\rangle, \\ T^{(j)}_{mn} &= \langle\chi_m|\hat{T}^{(j)}|\chi_n\rangle = \langle\chi_m|\partial\hat{H}/\partial\theta_j|\chi_n\rangle, \end{aligned} \quad \text{(A2)}$$

and, consequently, to the response coefficient (A1) expressed by using the matrices (A2) as

$$c = \mathrm{Tr}\{T^{(1)}(G_+ - G_-)T^{(2)}(G_+ - G_-)\}. \tag{A3}$$

However, in the evaluation of the last expression, attention has to be paid to the difference between the matrix $T^{(j)}(\theta_1,\theta_2)$ and the partial derivative of the matrix $H(\theta_1,\theta_2)$ with respect to θ_j. This difference follows from the identity $\hat{H} = \sum_{mn} |\chi_m\rangle H_{mn}\langle\chi_n|$, which yields

$$T^{(j)}_{mn} = \partial H_{mn}/\partial\theta_j + \sum_k \langle\chi_m|\partial\chi_k/\partial\theta_j\rangle H_{kn} + \sum_k H_{mk}\langle\partial\chi_k/\partial\theta_j|\chi_n\rangle, \tag{A4}$$

where we employed the orthogonality relations $\langle\chi_m(\theta_1,\theta_2)|\chi_n(\theta_1,\theta_2)\rangle = \delta_{mn}$. Their partial derivatives yield

$$\langle\chi_m|\partial\chi_n/\partial\theta_j\rangle = -\langle\partial\chi_m/\partial\theta_j|\chi_n\rangle \equiv Q^{(j)}_{mn}, \tag{A5}$$

where we introduced elements of matrices $Q^{(j)} = \{Q^{(j)}_{mn}\}$ for $j = 1,2$. Note that the matrices $Q^{(j)}(\theta_1,\theta_2)$ reflect explicitly the dependence of the basis vectors $|\chi_m(\theta_1,\theta_2)\rangle$ on θ_1 and θ_2. The relation (A4) between the matrices $T^{(j)}$ and $\partial H/\partial\theta_j$ can now be rewritten compactly as

$$T^{(j)} = \partial H/\partial\theta_j + [Q^{(j)}, H]. \tag{A6}$$

Since the last term has the form of a commutator with the Hamiltonian matrix H, the use of Eq. (A6) in formula (A3) leads to the final matrix expression for the torque correlation,

$$c = \mathrm{Tr}\{(\partial H/\partial\theta_1)(G_+ - G_-)(\partial H/\partial\theta_2)(G_+ - G_-)\}. \tag{A7}$$

The equivalence of Eqs. (A3) and (A7) rests on the rules $[Q^{(j)}, H] = [E_\mathrm{F} - H, Q^{(j)}]$ and $(E_\mathrm{F} - H)(G_+ - G_-) = (G_+ - G_-)(E_\mathrm{F} - H) = 0$ and on the cyclic invariance of the trace. It is also required that the matrices $Q^{(j)}$ are compatible with periodic boundary conditions used in calculations of extended systems, which is obviously the case for angular variables θ_j related to the global changes (uniform rotations) of the magnetization direction.

The obtained result means that the original response coefficient (A1) involving the torques as angular derivatives of the Hamiltonian can be expressed solely by using matrix elements of the Hamiltonian in an angle-dependent basis; the angular dependence of the basis vectors does not enter explicitly the final torque-correlation formula (A7).

APPENDIX B: LMTO HAMILTONIAN OF A FERROMAGNET WITH A TILTED MAGNETIC FIELD

Here we sketch a derivation of the fully relativistic LMTO Hamiltonian matrix for a ferromagnet with the XC-field direction tilted from a reference direction along an easy axis. The derivation rests on the form of the Kohn-Sham-Dirac Hamiltonian in the LMTO-ASA method [37–39]. The symbols with a superscript 0 refer to the reference system; the symbols without this superscript refer to the system with the tilted XC field. The operators (Hamiltonians, rotation operators) are denoted by symbols with a hat. The spin-dependent parts of the ASA potentials due to the XC fields are rigidly rotated, while the spin-independent parts are unchanged, in full analogy to the approach employed in the relativistic KKR method [19,22].

The ASA Hamiltonians of both systems are given by lattice sums $\hat{H}^0 = \sum_{\mathbf{R}} \hat{H}^0_{\mathbf{R}}$ and $\hat{H} = \sum_{\mathbf{R}} \hat{H}_{\mathbf{R}}$, where the individual site contributions are coupled mutually by $\hat{H}_{\mathbf{R}} = \hat{U}_{\mathbf{R}}\hat{H}^0_{\mathbf{R}}\hat{U}^+_{\mathbf{R}}$, where $\hat{U}_{\mathbf{R}}$ denotes the unitary operator of a rotation (in the orbital and spin space) around the **R**th lattice site, which brings the local XC field from its reference direction into the tilted one. Let $|\phi^0_{\mathbf{R}\Lambda}\rangle$ and $|\dot{\phi}^0_{\mathbf{R}\Lambda}\rangle$ denote, respectively, the ϕ and $\dot{\phi}$ orbitals of the reference Hamiltonian $\hat{H}^0_{\mathbf{R}}$; then

$$|\phi_{\mathbf{R}\Lambda}\rangle = \hat{U}_{\mathbf{R}}\big|\phi^0_{\mathbf{R}\Lambda}\big\rangle, \quad |\dot{\phi}_{\mathbf{R}\Lambda}\rangle = \hat{U}_{\mathbf{R}}\big|\dot{\phi}^0_{\mathbf{R}\Lambda}\big\rangle \tag{B1}$$

define the ϕ and $\dot{\phi}$ orbitals of the Hamiltonian $\hat{H}_{\mathbf{R}}$. The orbital index Λ labels all linearly independent solutions (regular at the origin) of the spin-polarized relativistic single-site problem; the detailed structure of Λ can be found elsewhere [37–39]. Let us introduce further the well-known empty-space solutions $|K^{\infty,0}_{\mathbf{R}N}\rangle$ (extending over the whole real space), $|K^{\mathrm{int},0}_{\mathbf{R}N}\rangle$ (extending over the interstitial region), and $|K^0_{\mathbf{R}N}\rangle$ and $|J^0_{\mathbf{R}N}\rangle$ (both truncated outside the **R**th sphere), needed for the definition of the LMTOs of the reference system [41,42,60]. Their index N, which defines the spin-spherical harmonics of the large component of each solution, can be taken either in the nonrelativistic (ℓms) form or in its relativistic ($\kappa\mu$) counterpart. We define further

$$|Z_{\mathbf{R}N}\rangle = \hat{U}_{\mathbf{R}}\big|Z^0_{\mathbf{R}N}\big\rangle \quad \text{for} \quad Z = K^\infty, K, J. \tag{B2}$$

Isotropy of the empty space guarantees relations (for $Z = K^\infty$, K, J)

$$|Z_{\mathbf{R}N}\rangle = \sum_{N'} \big|Z^0_{\mathbf{R}N'}\big\rangle U_{N'N}, \quad \big|Z^0_{\mathbf{R}N}\big\rangle = \sum_{N'} |Z_{\mathbf{R}N'}\rangle U^+_{N'N}, \tag{B3}$$

where $U = \{U_{N'N}\}$ denotes a unitary matrix representing the rotation in the space of spin-spherical harmonics and where $U^+_{N'N} \equiv (U^+)_{N'N} = (U_{NN'})^* = (U^{-1})_{N'N}$; the matrix U is the same for all lattice sites **R** since we consider only uniform rotations of the XC-field direction inside the ferromagnet. The expansion theorem for the envelope orbital $|K^{\infty,0}_{\mathbf{R}N}\rangle$ is

$$\big|K^{\infty,0}_{\mathbf{R}N}\big\rangle = \big|K^{\mathrm{int},0}_{\mathbf{R}N}\big\rangle + \big|K^0_{\mathbf{R}N}\big\rangle - \sum_{\mathbf{R}'N'} \big|J^0_{\mathbf{R}'N'}\big\rangle S^0_{\mathbf{R}'N',\mathbf{R}N}, \tag{B4}$$

where $S^0_{\mathbf{R}'N',\mathbf{R}N}$ denote elements of the canonical structure-constant matrix (with vanishing on-site elements, $S^0_{\mathbf{R}N',\mathbf{R}N} = 0$) of the reference system. The use of relations (B3) in the expansion (B4) together with an abbreviation

$$\big|K^{\mathrm{int}}_{\mathbf{R}N}\big\rangle = \sum_{N'} \big|K^{\mathrm{int},0}_{\mathbf{R}N'}\big\rangle U_{N'N} \tag{B5}$$

yields the expansion of the envelope orbital $|K^\infty_{\mathbf{R}N}\rangle$ as

$$\big|K^\infty_{\mathbf{R}N}\big\rangle = \big|K^{\mathrm{int}}_{\mathbf{R}N}\big\rangle + |K_{\mathbf{R}N}\rangle - \sum_{\mathbf{R}'N'} |J_{\mathbf{R}'N'}\rangle (U^+ S^0 U)_{\mathbf{R}'N',\mathbf{R}N}, \tag{B6}$$

where U and U^+ denote site-diagonal matrices with elements $U_{\mathbf{R}'N',\mathbf{R}N} = \delta_{\mathbf{R}'\mathbf{R}}U_{N'N}$ and $(U^+)_{\mathbf{R}'N',\mathbf{R}N} = \delta_{\mathbf{R}'\mathbf{R}}U^+_{N'N}$. Note the same form of expansions (B4) and (B6), with the orbitals $|Z^0_{\mathbf{R}N}\rangle$ replaced by the rotated orbitals $|Z_{\mathbf{R}N}\rangle$ ($Z = K^\infty, K, J$), with the interstitial parts $|K^{\mathrm{int},0}_{\mathbf{R}N}\rangle$ replaced by their linear

combinations $|K^{\rm int}_{\mathbf{R}N}\rangle$, and with the structure-constant matrix S^0 replaced by the product U^+S^0U.

The nonorthogonal LMTO $|\chi^0_{\mathbf{R}N}\rangle$ for the reference system is obtained from the expansion (B4), in which all orbitals $|K^0_{\mathbf{R}N}\rangle$ and $|J^0_{\mathbf{R}N}\rangle$ are replaced by linear combinations of $|\phi^0_{\mathbf{R}\Lambda}\rangle$ and $|\dot\phi^0_{\mathbf{R}\Lambda}\rangle$. A similar replacement of the orbitals $|K_{\mathbf{R}N}\rangle$ and $|J_{\mathbf{R}N}\rangle$ by linear combinations of $|\phi_{\mathbf{R}\Lambda}\rangle$ and $|\dot\phi_{\mathbf{R}\Lambda}\rangle$ in the expansion (B6) yields the nonorthogonal LMTO $|\chi_{\mathbf{R}N}\rangle$ for the system with the tilted XC field. The coefficients in these linear combinations, obtained from conditions of continuous matching at the sphere boundaries and leading directly to the LMTO potential parameters, are identical for both systems, which follows from the rotation relations (B1) and (B2). For these reasons, the only essential difference between both systems in the construction of the nonorthogonal and orthogonal LMTOs (and of the accompanying Hamiltonian and overlap matrices in the ASA) is due to the difference between the matrices S^0 and U^+S^0U.

As a consequence, the LMTO Hamiltonian matrix in the orthogonal LMTO representation for the system with a tilted magnetization is easily obtained from that for the reference system, Eq. (6), and it is given by

$$H = C + (\sqrt{\Delta})^+U^+SU(1-\gamma U^+SU)^{-1}\sqrt{\Delta}, \qquad \text{(B7)}$$

where C, $\sqrt{\Delta}$, and γ are site-diagonal matrices of the potential parameters of the reference system and where we suppressed the superscript 0 at the structure-constant matrix S of the reference system. Note that the dependence of H on the XC-field direction is contained only in the similarity transformation U^+SU of the original structure-constant matrix S generated by the rotation matrix U. For the rotation by an angle θ around an axis along a unit vector $\mathbf{n}$, the rotation matrix is given by $U(\theta) = \exp(-i\mathbf{n}\cdot\mathbf{J}\theta)$, where the site-diagonal matrices $\mathbf{J} \equiv (\mathcal{J}^x, \mathcal{J}^y, \mathcal{J}^z)$ with matrix elements $\mathcal{J}^\mu_{\mathbf{R}'N',\mathbf{R}N} = \delta_{\mathbf{R}'\mathbf{R}}\mathcal{J}^\mu_{N'N}$ ($\mu = x,y,z$) reduce to the usual matrices of the total (orbital plus spin) angular momentum operator. The limit of small θ yields $U(\theta) \approx 1 - i\mathbf{n}\cdot\mathbf{J}\theta$, which leads to the θ derivative of the Hamiltonian matrix (B7) at $\theta = 0$:

$$\partial H/\partial\theta = i(F^+)^{-1}[\mathbf{n}\cdot\mathbf{J}, S]F^{-1}, \qquad \text{(B8)}$$

where we abbreviated $F = (\sqrt{\Delta})^{-1}(1-\gamma S)$ and $F^+ = (1 - S\gamma)[(\sqrt{\Delta})^+]^{-1}$. Since the structure-constant matrix S is spin independent, the total angular momentum operator $\mathbf{J}$ in (B8) can be replaced by its orbital momentum counterpart $\mathbf{L} \equiv (\mathcal{L}^x, \mathcal{L}^y, \mathcal{L}^z)$, so that

$$\partial H/\partial\theta = i(F^+)^{-1}[\mathbf{n}\cdot\mathbf{L}, S]F^{-1}. \qquad \text{(B9)}$$

Relations (B8) and (B9) are used to derive the LMTO-ASA torque-correlation formula (8).

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