Ab Initio Theory of the Gilbert Damping in Random Ferromagnetic Alloys

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Abstract We present an ab initio theory of the Gilbert damping in ferromagnetic alloys with substitutional disorder. The theory is based on nonlocal torques that are represented by nonrandom, site-off-diagonal, and spin-independent matrices, which simplifies the configuration averaging. The formalism is developed for the relativistic tight-binding linear muffin-tin orbital (TB-LMTO) method and the coherent potential approximation (CPA). 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 random local torques. The theory is illustrated by calculations for various random transition metal alloys: FeNi, FeCo, Heusler alloys, permalloy with impurities, Fe with vacancies, and stoichiometric FePt and CoPt alloys with a varying degree of L10 atomic long-range order. Results are in a reasonable agreement with other calculations and accessible experimental data.

Keywords Gilbert damping · Ferromagnetic alloys · Ab initio · Nonlocal torques

1 Introduction

The dynamics of magnetization is an important property of magnetic systems, in particular in the context of high speed magnetic devices for data storage. Although the detailed theory of magnetization dynamics including, e.g., excitation of magnons and their interaction with other degrees of freedom is still under development, remarkable progress concerning the dynamics of the total magnetic moment has been achieved during the last years.

Time evolution of the macroscopic magnetization vector \( \mathbf{M} \) can be described by the well-known Landau-Lifshitz-Gilbert (LLG) equation

\[
\frac{d\mathbf{M}}{dt} = \mathbf{B}_{\text{eff}} \times \mathbf{M} + \frac{\mathbf{M}}{M} \times \left( \alpha \cdot \frac{d\mathbf{M}}{dt} \right),
\]

(1)

where \( \mathbf{B}_{\text{eff}} \) is an effective magnetic field (with the gyromagnetic ratio absorbed) acting on the magnetization, \( M = |\mathbf{M}| \), and the quantity \( \alpha = \{\alpha_{\mu\nu}\} \) denotes a symmetric \( 3 \times 3 \) tensor of the dimensionless Gilbert damping parameters \( (\mu, \nu = x, y, z) \).

Several approaches to calculate the Gilbert damping from first principles were developed (see, e.g., references in [1]) that work well for ordered systems. Nevertheless, the situation in disordered systems is more complicated as the configuration averaging is difficult due to the randomness of the torque operators involved. We have therefore developed an alternative theory based on nonrandom torques and made basic tests of this theory for selected systems [1]. Here, we wish to present the results for other systems in order to further verify its validity and accuracy.
2 Formalism

The tensor of Gilbert damping parameters can be calculated from the torque-correlation formula

\[ \alpha_{\mu \nu} = 4\pi^2 \alpha_0 \sum_{i,j} \langle j | T_{\mu i} | i \rangle \delta(E_F - E_i) \langle i | T_{\nu j} | j \rangle \delta(E_F - E_j) \]

\[ = -\alpha_0 \text{Tr} \left[ T_{\mu i} (G_+ - G_-) T_{\nu j} (G_+ - G_-) \right], \quad (2) \]

where \( \alpha_0 = 1/(2\pi M_{\text{spin}}) \), \(|i\rangle\) are eigenstates and \( E_i \) are eigenvalues of the one-particle Hamiltonian, \( T_{\mu i} \) are torque operators, the trace is taken over the Hilbert space of valence electrons, \( G_\pm = G(E_F \pm i\alpha) \) are the one-particle retarded and advanced Green’s functions at the Fermi energy \( E_F \), and \( M_{\text{spin}} \) is the spin magnetization.

The torque operators \( T_{\mu i} \) are closely related to the time derivative of electron spin. The one-electron Hamiltonian \( H \) of a spin-polarized system can be written as \( H = H^p + H^xc \), where \( H^p \) includes all spin-independent terms and the spin-orbit (SO) interaction (a paramagnetic system) and \( H^xc = B^xc(\mathbf{r}) \cdot \mathbf{\sigma} \) denotes the exchange-correlation (XC) term due to an effective magnetic field \( B^xc(\mathbf{r}) \).

The complete time derivative of the spin operator is given by the commutator \( t_{\mu} = -i[\sigma_{\mu}/2, H] \), where \( h = 1 \) and \( \sigma_{\mu} \) denote the Pauli spin matrices. Then \( t_{\mu} = t_{\mu}^{so} + t_{\mu}^{xc} \), where

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

As discussed, e.g., in [2], the use of the complete torque \( t_{\mu} \) in the torque-correlation formula (2) leads identically to zero; the correct Gilbert damping coefficients \( \alpha_{\mu \nu} \) follow from (2) by using either the SO-induced torque \( t_{\mu}^{so} \), or the XC-induced torque \( t_{\mu}^{xc} \). The reason is that (2) contains only matrix elements of the torque evaluated on the energy shell that fulfills \( \langle i | [\sigma_{\mu}/2, H] | j \rangle = (E_j - E_i) \langle i | \sigma_{\mu} | j \rangle = 0 \) because \( E_i = E_j \). Consequently, the operators \( t_{\mu}^{so} \) and \( -t_{\mu}^{xc} \) are equivalent in \( t_{\mu}^{so} \approx -t_{\mu}^{xc} \), from the point of view of (2) even if they are not equal. They both can be used to calculate Gilbert damping parameters \( \alpha_{\mu \nu} \). They are local, depend on spin and on the type of atom so that they are suitable for ordered systems. On the other hand, in disordered systems, their dependence on the type of the atom brings a more difficult calculation of the disorder-induced vertex corrections. This problem can be avoided by introducing the nonlocal torques.

Let us consider a tight-binding Hamiltonian \( H = H^{loc} + H^{nl} \), where

\[ H^{loc} = \sum_{\mathbf{R}} \left( H^p_{\mathbf{R}} + H^xc_{\mathbf{R}} \right) \quad (4) \]

is the local atomic-like term that contains SO interaction and XC fields of all sites \( \mathbf{R} \) while \( H^{nl} \) is the nonlocal term that contains intersite hopping matrix elements and which is spin-independent (\(|{\sigma_{\mu}} | H^{nl} \rangle = 0\)). The spin-independent part \( H^p_{\mathbf{R}} \) of the local term contains on-site SO interaction and we assume that it is spherically symmetric. The local XC part \( H^xc_{\mathbf{R}} = B^xc_{\mathbf{R}} \cdot \mathbf{\sigma} \) depends on the local XC field \( B^xc_{\mathbf{R}} \) which is assumed to have a constant size and direction. \( H^p_{\mathbf{R}} \) and \( H^xc_{\mathbf{R}} \) act only on the site \( \mathbf{R} \) and the subspaces of different sites are orthogonal.

Now we consider local spin and orbital momentum operators \( \sigma_{\mu \nu} \) and \( L_{\mathbf{R} \mu} \), as well as their sum giving total momentum operator \( J_{\mathbf{R} \mu} = L_{\mathbf{R} \mu} + \sigma_{\mathbf{R} \mu}/2 \). One can find by direct calculation

\[ [J_{\mathbf{R} \mu}, H^p_{\mathbf{R}}] = 0, \quad [L_{\mathbf{R} \mu}, H_{\mathbf{R}}^{xc}] = 0. \quad (5) \]

Using (5) and introducing \( J_{\mu} = \sum_{\mathbf{R}} J_{\mathbf{R} \mu} \) and \( L_{\mu} = \sum_{\mathbf{R}} L_{\mathbf{R} \mu} \), we obtain

\[ t_{\mu}^{xc} = -i \sum_{\mathbf{R}} \left[ \sigma_{\mu \nu} | H^xc_{\mathbf{R}} \right] = -i \sum_{\mathbf{R}} \left[ J_{\mathbf{R} \mu}, H^xc_{\mathbf{R}} \right] \]

\[ = -i \sum_{\mathbf{R}} \left[ J_{\mathbf{R} \mu}, H^p_{\mathbf{R}} + H^xc_{\mathbf{R}} \right] = -i [J_{\mu}, H^{loc}] \equiv i^{loc}_{\mu}, \quad (6) \]

where \( i^{loc}_{\mu} \) is the local torques. The nonlocal torque is defined as \( t^{nl}_{\mu} = -i [J_{\mu}, H^{nl}] = -i [L_{\mu}, H^{nl}] \).

Similarly as before, operators \( i^{loc}_{\mu} \) and \( -t^{nl}_{\mu} \) are not equal, \( t^{nl}_{\mu} \neq -t^{nl}_{\mu} \), but are equivalent on the energy shell, \( t^{nl}_{\mu} \approx -t^{nl}_{\mu} \). Altogether, we have

\[ t^{so}_{\mu} \approx -t^{xc}_{\mu} \approx -t^{loc}_{\mu} = t^{nl}_{\mu}, \quad (7) \]

which means that the nonlocal torques \( t^{nl}_{\mu} \) is equivalent to all local torques. The main advantages of the nonlocal torque are that it is spin-independent and nonrandom. This considerably simplifies evaluation of the Gilbert damping for disordered systems. It turns out that the disorder-induced vertex corrections are highly important and cannot be neglected.

We made ab initio calculations within the fully relativistic tight-binding linear muffin-tin (TB-LMTO) method combined with the coherent potential approximation (CPA) and we calculated the disorder-induced vertex corrections within the CPA. The details of derivation, computational procedure, and explicit formulas can be found in [1].

3 Results

3.1 Fe\textsubscript{x}Ni\textsubscript{1−x} and Co\textsubscript{x}Fe\textsubscript{1−x} Alloys

For fcc-Fe\textsubscript{x}Ni\textsubscript{1−x} and bcc-Co\textsubscript{x}Fe\textsubscript{1−x} alloys, we have found a good agreement between our results [1] and other theoretical methods (LMTO supercell approach [3] and KKR-CPA [4]) that use a different type of torque (XC).
Table 1 Gilbert damping parameter $10^3 \alpha$ of bcc Fe$_{1-x}$Vac$_x$ with vacancies

| x   | This work | Ref. [4] |
|-----|-----------|----------|
| 0.001 | 6.59      | 8.0      |
| 0.002 | 3.49      |          |
| 0.003 | 2.45      |          |

3.2 Iron with Vacancies

As a further test, we calculated Gilbert damping for bcc Fe with vacancies (see Table 1). A comparison with results of the KKR-CPA [4] has shown a reasonable agreement. In the limit of low concentration of vacancies the Gilbert damping is increasing as expected.

3.3 Heusler Alloys

Heusler alloys are nowadays important materials with many interesting properties. The Gilbert damping in some Co-based Heusler alloys was studied experimentally [5] and theoretically [6]. There is a mismatch between the experimental and theoretical values: the calculated values are approximately ten times smaller than those measured. We made calculations for alloys with some degree of disorder to see if the disorder can be responsible for the discrepancy (Table 2). It turns out that disorder in some cases leads to increase of $\alpha$ and in some cases to decrease of $\alpha$. Nevertheless, we generally find values larger than those reported in [6], which can probably be ascribed to the fully relativistic treatment in present calculations.

Table 2 Gilbert damping parameter $\alpha$ of selected Heusler alloys. The arrows denote orientation of magnetic moments and $y = 1 - x$

| Alloy | $x$ | $10^3 \alpha$ |
|-------|-----|---------------|
| Co$_x$(Mn$_{1-y}$Al$_y$)Co$_y$(Mn$_{1-y}$Al$_y$) | 0.50 | 1.132 |
|       | 0.25 | 1.166         |
|       | 0.10 | 1.154         |
|       | 0.01 | 1.881         |
| (Co$_x$Mn$_{1-y}$)(Co$_2x$Mn$_{1-2y}$)(Co$_y$Mn$_{1-y}$)Al | 0.05 | 2.252 |
| Co$_x$(Mn$_{1-y}$Si$_y$)Co$_y$(Mn$_{1-y}$Si$_y$) | 0.50 | 3.946 |
|       | 0.05 | 4.192         |
|       | 0.01 | 4.945         |
| (Co$_x$Mn$_{1-y}$)Mn$_y$(Co$_y$Mn$_{1-y}$)Si | 0.04 | 0.4603 |
|       | 0.07 | 0.5567        |
| (Co$_x$Mn$_{1-y}$)(Co$_2x$Mn$_{1-2y}$)(Co$_y$Mn$_{1-y}$)Si | 0.04 | 1.756 |
|       | 0.005| 0.4988        |

3.4 Permalloy with Transition Metal Impurities

Permalloy (fcc-Fe$_{20}$Ni$_{80}$) is an alloy important in technical applications and here we wish to show that an addition of a third component can considerably change the Gilbert damping (see Table 3). The 5d impurities have quite a strong influence due to the strong SO coupling. Our results compare reasonably well with the theory [7, 8] and with the experiment [9].

3.5 Ordering FePt and CoPt Alloys

Finally, we address the Gilbert damping in stoichiometric CoPt and FePt alloys with L1$_0$ atomic long-range order (LRO). The transport properties [10] and the damping parameter [8] of FePt alloys have recently been studied by means of the TB-LMTO method for a varying degree of the LRO. The LRO parameter $S$ for the stoichiometric alloy AB with two sublattices which is represented as $(A_{1-w}B_w,A_{w}B_{1-w})$ is defined as $S = 1 - 2w$, where $w$ is the concentration of "wrong" atoms on each sublattice. Then $0 \leq S \leq 1$ and $S = 0$ correspond to the random fcc alloy and $S = 1$ corresponds to the perfectly ordered L1$_0$ structure.

The Gilbert damping parameter $\alpha$ is displayed in Fig. 1 as a function of $S$. The trend with a broad maximum at $S = 0$ and a minimum around $S = 0.9$ agrees for FePt system very well with [8]. Our values are about 10% higher than those in [8], which can be ascribed to the fully relativistic treatment in the present study in contrast to a simplified treatment of the SO interaction in [8]. The Gilbert damping in CoPt and FePt is an order of magnitude stronger than in the alloys of 3d elements (Section 3.1) owing to the stronger SO interaction of Pt atoms.

The dependence of $\alpha$ on the LRO parameter $S$ can be qualitatively discussed in terms of the spin magnetization $M_{spin}$ and the density of states $D(E_F)$ on the Fermi energy, because $\alpha \propto D(E_F)/M_{spin}$, see, e.g., [11]. In the systems
with constant composition the moment $M_{\text{spin}}$ varies with $S$ only slightly and the main effect comes from $D(E_F)$, cf. [1, 11].

All calculated values of $\alpha$ for FePt system are appreciably smaller than the measured ones $\alpha^{\text{exp}} \approx 0.06$ reported for a thin L1$_0$ FePt epitaxial film. The high value of $\alpha^{\text{exp}}$ might be thus explained by the present calculations either by a very small concentration of antisites in the prepared films, or by the thin-film geometry. The divergence of $\alpha$ in the limit of $S \rightarrow 1$ (Fig. 1) 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 must take into account the full interacting susceptibility in the presence of the SO interaction as well as the effect of temperature and other scattering mechanisms.

4 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 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 are necessary for an exact equivalence of the nonlocal nonrandom torques with their local random counterparts. The idea of nonlocal torques is not limited to the LMTO method and its application in the semiempirical TB theory or in the KKR theory is straightforward.

The numerical implementation and the results for binary solid solutions show that the total Gilbert damping parameters calculated from the nonlocal torques are much smaller than magnitudes of the coherent parts and of the vertex corrections [1]. Nevertheless, the total damping parameters for the studied Fe$_x$Ni$_{1-x}$, Co$_x$Fe$_{1-x}$, Heusler alloys, permalloy with impurities, Fe with vacancies, and stoichiometric FePt and CoPt alloys with a varying degree of L1$_0$ atomic long-range order compare quantitatively well with the results of other ab initio techniques, which indicates a fair numerical stability of the present theory.

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