Ab Initio Calculation of the Gilbert Damping Parameter via the Linear Response Formalism

H. Ebert, S. Mankovsky, and D. Ködderitzsch
University of Munich, Department of Chemistry, Butenandtstrasse 5-13, D-81377 Munich, Germany

P. J. Kelly
Faculty of Science and Technology and MESA$^+$ Institute for Nanotechnology, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands

(Received 1 March 2011; published 2 August 2011)

A Kubo-Greenwood-like equation for the Gilbert damping parameter α is presented that is based on the linear response formalism. Its implementation using the fully relativistic Korringa-Kohn-Rostoker band structure method in combination with coherent potential approximation alloy theory allows it to be applied to a wide range of situations. This is demonstrated with results obtained for the bcc alloy system $Fe_{1-x}Co_x$ as well as for a series of alloys of Permalloy with $5d$ transition metals. To account for the thermal displacements of atoms as a scattering mechanism, an alloy-analogy model is introduced. The corresponding calculations for Ni correctly describe the rapid change of α when small amounts of substitutional Cu are introduced.

DOI: 10.1103/PhysRevLett.107.066603 PACS numbers: 72.25.Rb, 71.20.Be, 71.70.Ej, 75.78.−n

The magnetization dynamics that is relevant for the performance of any type of magnetic device is in general governed by damping. In most cases the magnetization dynamics can be modeled successfully by means of the Landau-Lifshitz-Gilbert (LLG) equation [1] that accounts for damping in a phenomenological way. The possibility to calculate the corresponding damping parameter from first principles would open the perspective of optimizing materials for devices and has therefore motivated extensive theoretical work in the past. This led among others to Kambersky's breathing Fermi surface (BFS) [2] and torque-correlation models (TCM) [3], that in principle provide a firm basis for numerical investigations based on electronic structure calculations [4,5]. The spin-orbit coupling that is seen as a key factor in transferring energy from the magnetization to the electronic degrees of freedom is explicitly included in these models. Most *ab initio* results have been obtained for the BFS model though the torque-correlation model makes fewer approximations [4,6]. In particular, it in principle describes the physical processes responsible for Gilbert damping over a wide range of temperatures as well as chemical (alloy) disorder. However, in practice, like many other models it depends on a relaxation time parameter τ that describes the rate of transfer due to the various types of possible scattering mechanisms. This weak point could be removed recently by Brataas *et al.* [7] who described the Gilbert damping by means of scattering theory. This development supplied the formal basis for the first parameter-free investigations on disordered alloys for which the dominant scattering mechanism is potential scattering caused by chemical disorder [8] or temperature induced structure disorder [9].

As pointed out by Brataas *et al.* [7], their approach is completely equivalent to a formulation in terms of the linear response or Kubo formalism. The latter route is taken in this communication that presents a Kubo-Greenwood-like expression for the Gilbert damping parameter. Application of the scheme to disordered alloys demonstrates that this approach is indeed fully equivalent to the scattering theory formulation of Brataas *et al.* [7]. In addition a scheme is introduced to deal with the temperature dependence of the Gilbert damping parameter.

Following Brataas *et al.* [7], the starting point of our scheme is the Landau-Lifshitz-Gilbert (LLG) equation for the time derivative of the magnetization $\vec{M}$:

$$\frac{1}{\gamma}\frac{d\vec{M}}{d\tau} = -\vec{M}\times\vec{H}_{\text{eff}} + \vec{M}\times\left[\frac{\tilde{G}(\vec{M})}{\gamma^2 M_s^2}\frac{d\vec{M}}{d\tau}\right], \qquad (1)$$

where M_s is the saturation magnetization, γ the gyromagnetic ratio, and $\tilde{G}$ the Gilbert damping tensor. Accordingly, the time derivative of the magnetic energy is given by

$$\dot{E}_{\text{mag}} = \vec{H}_{\text{eff}}\cdot\frac{d\vec{M}}{d\tau} = \frac{1}{\gamma^2}\dot{\vec{m}}[\tilde{G}(\vec{m})\dot{\vec{m}}] \qquad (2)$$

in terms of the normalized magnetization $\vec{m} = \vec{M}/M_s$. On the other hand, the energy dissipation of the electronic system $\dot{E}_{\text{dis}} = \langle\frac{d\hat{H}}{d\tau}\rangle$ is determined by the underlying Hamiltonian $\hat{H}(\tau)$. Expanding the normalized magnetization $\vec{m}(\tau)$, that determines the time dependence of $\hat{H}(\tau)$ about its equilibrium value, $\vec{m}(\tau) = \vec{m}_0 + \vec{u}(\tau)$, one has

$$\hat{H} = \hat{H}_0(\vec{m}_0) + \sum_\mu \vec{u}_\mu \frac{\partial}{\partial\vec{u}_\mu}\hat{H}(\vec{m}_0). \qquad (3)$$

Using the linear response formalism, $\dot{E}_{\text{dis}}$ can be written as [7]

 © 2011 American Physical Society

$$\dot{E}_{\mathrm{dis}} = -\pi\hbar \sum_{ii'} \sum_{\mu\nu} \dot{u}_\mu \dot{u}_\nu \left\langle \psi_i \left| \frac{\partial \hat{H}}{\partial u_\mu} \right| \psi_{i'} \right\rangle \left\langle \psi_{i'} \left| \frac{\partial \hat{H}}{\partial u_\nu} \right| \psi_i \right\rangle \times \delta(E_F - E_i)\delta(E_F - E_{i'}), \quad (4)$$

where E_F is the Fermi energy and the sums run over all eigenstates α of the system. Identifying $\dot{E}_{\mathrm{mag}} = \dot{E}_{\mathrm{dis}}$, one gets an explicit expression for the Gilbert damping tensor $\tilde{G}$ or equivalently for the damping parameter $\alpha = \tilde{G}/(\gamma M_s)$ [7]. In full analogy to electric transport [10], the sum over eigenstates $|\psi_i\rangle$ may be expressed in terms of the retarded single-particle Green's function $\mathrm{Im}G^+(E_F) = -\pi \sum_i |\psi_i\rangle\langle\psi_i| \delta(E_F - E_i)$. This leads for the parameter α to a Kubo-Greenwood-like equation

$$\alpha_{\mu\nu} = -\frac{\hbar\gamma}{\pi M_s} \mathrm{Trace} \left\langle \frac{\partial \hat{H}}{\partial u_\mu} \mathrm{Im}G^+(E_F) \frac{\partial \hat{H}}{\partial u_\nu} \mathrm{Im}G^+(E_F) \right\rangle_c \quad (5)$$

with $\langle \ldots \rangle_c$ indicating a configurational average in case of a disordered system (see below). Identifying $T_\mu = \partial \hat{H}/\partial u_\mu$ with the component of the magnetic torque operator $\hat{\vec{T}}$ along the direction $\vec{n}$, such that $\hat{I}_{\vec{n}} = \partial \hat{H}/\partial \vec{u}(\vec{n} \times \vec{u}) = \partial \hat{H}/\partial u_\mu (\vec{n} \times \vec{u})_\mu$ this expression obviously gives the parameter α in terms of a torque-torque correlation function. However, in contrast to the conventional TCM the electronic structure is not represented in terms of Bloch states but using the retarded electronic Green's function giving the present approach much more flexibility.

The most reliable way to account for spin-orbit coupling as the source of Gilbert damping is to evaluate Eq. (5) using a fully relativistic Hamiltonian within the framework of local spin density formalism (LSDA) [11]:

$$\hat{H} = c\vec{\alpha}\,\vec{p} + \beta mc^2 + V(\vec{r}) + \beta\vec{\sigma}\,\vec{m}\,B(\vec{r}). \quad (6)$$

Here α_i and β are the standard Dirac matrices and $\vec{p}$ is the relativistic momentum operator [12]. The functions V and B are the spin-averaged and spin-dependent parts, respectively, of the LSDA potential. Equation (6) implies for the T_μ operator occurring in Eq. (5) the expression

$$T_\mu = \frac{\partial}{\partial u_\mu} \hat{H} = \beta B \sigma_\mu. \quad (7)$$

The Green's function G^+ in Eq. (5) can be obtained in a very efficient way by using the spin-polarized relativistic version of multiple scattering theory [11] that allows us to treat magnetic solids:

$$G^+(\vec{r}_n, \vec{r}'_m, E) = \sum_{\Lambda\Lambda'} Z^n_\Lambda(\vec{r}_n, E) \tau^{nm}_{\Lambda\Lambda'}(E) Z^{m\times}_{\Lambda'}(\vec{r}'_m, E) - \sum_{\Lambda} Z^n_\Lambda(\vec{r}_<, E) J^{n\times}_{\Lambda'}(\vec{r}_>, E) \delta_{nm}. \quad (8)$$

Here coordinates $\vec{r}_n$ referred to the center of cell n have been used with $|\vec{r}_<| = \min(|\vec{r}_n|, |\vec{r}'_n|)$ and $|\vec{r}_>| = \max(|\vec{r}_n|, |\vec{r}'_n|)$. The four-component wave functions $Z^n_\Lambda(\vec{r}, E)$ ($J^n_\Lambda(\vec{r}, E)$) are regular (irregular) solutions to the single-site Dirac equation for site n and $\tau^{nm}_{\Lambda\Lambda'}(E)$ is the so-called scattering path operator that transfers an electronic wave coming in at site m into a wave going out from site n with all possible intermediate scattering events accounted for coherently.

Using matrix notation, this leads to the following expression for the damping parameter:

$$\alpha_{\mu\mu} = \frac{g}{\pi\mu_{\mathrm{tot}}} \sum_n \mathrm{Trace} \langle \underline{T}^{0\mu} \underline{\tilde{\tau}}^{0n} \underline{T}^{n\mu} \underline{\tilde{\tau}}^{n0} \rangle_c \quad (9)$$

with the g factor $2(1 + \mu_{\mathrm{orb}}/\mu_{\mathrm{spin}})$ in terms of the spin and orbital moments, μ_{spin} and μ_{orb}, respectively, the total magnetic moment $\mu_{\mathrm{tot}} = \mu_{\mathrm{spin}} + \mu_{\mathrm{orb}}$, and $\tilde{\tau}^{0n}_{\Lambda\Lambda'} = \frac{1}{2i}(\tau^{0n}_{\Lambda\Lambda'} - \tau^{0n}_{\Lambda'\Lambda})$ and with the energy argument E_F omitted. The matrix elements of the torque operator, $\underline{T}^{n\mu}$, are identical to those occurring in the context of exchange coupling [13] and can be expressed in terms of the spin-dependent part B of the electronic potential with matrix elements:

$$T^{n\mu}_{\Lambda'\Lambda} = \int d^3r Z^{n\times}_{\Lambda'}(\vec{r}) [\beta\sigma_\mu B_{xc}(\vec{r})] Z^n_\Lambda(\vec{r}). \quad (10)$$

As indicated above, the expressions in Eqs. (5)–(10) can be applied straightforwardly to disordered alloys. In this case the brackets $\langle \ldots \rangle_c$ indicate the necessary configurational average. This can be done by describing in a first step the underlying electronic structure (for $T = 0$ K) on the basis of the coherent potential approximation (CPA) alloy theory. In the next step the configurational average in Eq. (5) is taken following the scheme worked out by Butler [10] when dealing with the electrical conductivity at $T = 0$ K or residual resistivity, respectively, of disordered alloys. This implies, in particular, that so-called vertex corrections of the type $\langle T_\mu \mathrm{Im}G^+ T_\nu \mathrm{Im}G^+ \rangle_c - \langle T_\mu \mathrm{Im}G^+ \rangle_c \langle T_\nu \mathrm{Im}G^+ \rangle_c$ that account for scattering-in processes in the language of the Boltzmann transport formalism are properly accounted for.

Thermal vibrations as a source of electron scattering can in principle be accounted for by a generalization of Eqs. (5)–(10) to finite temperatures and by including the electron-phonon self-energy $\Sigma_{\text{el-ph}}$ when calculating the Green's function G^+. Here we restrict ourselves to elastic scattering processes by using a quasistatic representation of the thermal displacements of the atoms from their equilibrium positions. We introduce an alloy-analogy model to average over a discrete set of displacements that is chosen to reproduce the thermal root mean square average displacement $\sqrt{\langle u^2 \rangle_T}$ for a given temperature T. This was chosen according to $\langle u^2 \rangle_T = \frac{1}{4} \frac{3h^2}{\pi^2 mk\Theta_D} [\frac{\Phi(\Theta_D/T)}{\Theta_D/T} + \frac{1}{4}]$ with $\Phi(\Theta_D/T)$ the Debye function, h the Planck constant, k the Boltzmann constant, and Θ_D the Debye temperature [14]. Ignoring the zero temperature term $1/4$ and assuming a frozen potential for the atoms, the situation can be dealt

with in full analogy to the treatment of disordered alloys described above.

The approach described above has been applied to the ferromagnetic 3*d*-transition metal alloy systems bcc $Fe_{1-x}Co_x$, fcc $Fe_{1-x}Ni_x$, and fcc $Co_{1-x}Ni_x$. Figure 1 shows as an example results for bcc $Fe_{1-x}Co_x$ for $x \leq 0.7$. The calculated damping parameter $\alpha(x)$ for $T = 0$ K is found to be in very good agreement with the results based on the scattering theory approach [8] demonstrating numerically the equivalence of the two approaches. An indispensable requirement to achieving this agreement is to include the vertex corrections mentioned above. In fact, ignoring them leads in some cases to completely unphysical results. To check the reliability of the standard CPA, that implies a single-site approximation when performing the configurational average, we performed calculations on the basis of the nonlocal CPA [15]. Using a four-atom cluster led to practically the same results as the CPA except for the very dilute case. As found before for fcc $Fe_{1-x}Ni_x$ [8] the theoretical results for α reproduce the concentration dependence of the experimental data quite well but are found to be too low (see below). As suggested by Eq. (9) the variation of $\alpha(x)$ with concentration x may reflect to some extent the variation of the average magnetic moment of the alloy, μ_{tot}. Because the moments and spin-orbit coupling strength do not differ very much for Fe and Co, the variation of $\alpha(x)$ should be determined in the concentrated regime primarily by the electronic structure at the Fermi energy E_F. As Fig. 1 shows, there is indeed a close correlation with the density of states $n(E_F)$ that may be seen as a measure for the number of available relaxation channels.

While the scattering and linear response approach are completely equivalent when dealing with bulk alloys the latter allows us to perform the necessary configuration averaging in a much more efficient way. This allows us to study with moderate effort the influence of varying the alloy composition on the damping parameter α.

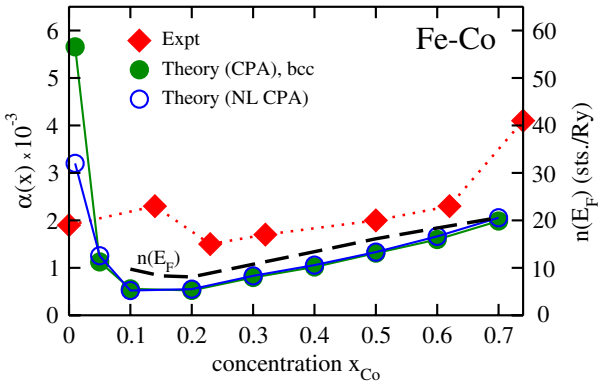

FIG. 1 (color online). Gilbert damping parameter for bcc $Fe_{1-x}Co_x$ as a function of Co concentration: full circles—the present results within CPA; empty circles—within nonlocal CPA (NL CPA); and full diamonds—experimental data by Oogane [20].

Corresponding work has been done, in particular, using Permalloy as a starting material and adding transition metals (TM) [16] or rare earth metals [17]. If we use the present scheme to study the effect of substituting Fe and Ni atoms in Permalloy with a 5*d* TM, we find an increase of α nearly linear with the 5*d* TM content, just as in experiment [16]. The total damping for 10% 5*d* TM content shown in Fig. 2 (top) varies roughly parabolically over the 5*d* TM series. In contrast to the $Fe_{1-x}Co_x$ alloys considered above, there is now an *S*-like variation of the moments μ_{spin}^{5d} over the series (Fig. 2, bottom), characteristic of 5*d* impurities in the pure hosts Fe and Ni [18,19]. In spite of this behavior of μ_{spin}^{5d} the variation of $\alpha(x)$ seems again to be correlated with the density of states $n^{5d}(E_F)$ (Fig. 2 bottom). Again the trend of the experimental data is well reproduced by the calculated values that are, however, somewhat too low.

One possible reason for the discrepancy between the theoretical and experimental results shown in Figs. 1 and 2 might be the neglect of the influence of finite temperatures. This can be included as indicated above to account for the thermal displacement of the atoms in a quasistate way by performing a configurational average over the displacements using the CPA. This leads even for pure systems to a scattering mechanism and this way to a finite value for α. Corresponding results for pure Ni are given in Fig. 3 that show in full accordance with experiment a rapid decrease of α with increasing temperature until a regime with a weak variation of α with T is reached. This behavior is commonly interpreted as a transition from conductivity-like to resistivitylike behavior reflecting the dominance of intra- and interband transition, respectively [4], that is related to the increase of the broadening of electron energy bands caused by the increase of scattering events with temperature. Adding even less than 1 at.% Cu to Ni strongly reduces the conductivitylike behavior at low temperatures while leaving the high-temperature behavior essentially unchanged. A further increase of the Cu content leads to the impurity-scattering processes responsible for

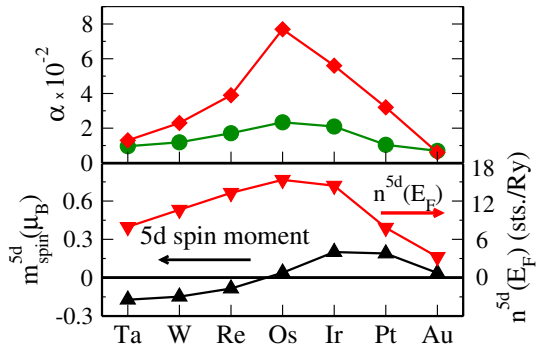

FIG. 2 (color online). Top: Gilbert damping parameter α for Py/5*d* TM systems with 10% 5*d* TM content in comparison with experiment [16]; bottom: spin magnetic moment m_{spin}^{5d} and density of states $n(E_F)$ at the Fermi energy of the 5*d* component in Py/5*d* TM systems with 10% 5*d* TM content.

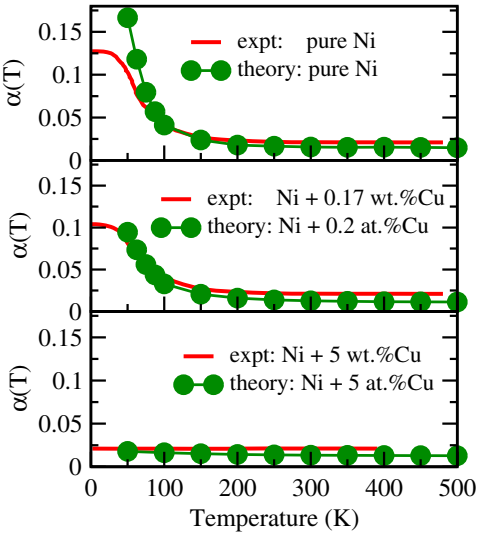

FIG. 3 (color online). Temperature variation of Gilbert damping of pure Ni and Ni with Cu impurities: present theoretical results vs experiment [21].

band broadening dominating α. This effect completely suppresses the conductivitylike behavior in the low-temperature regime because of the increase of scattering events due to chemical disorder. Again this is fully in line with the experimental data, providing a straightforward explanation for their peculiar variation with temperature and composition.

From the results obtained for Ni one may conclude that thermal lattice displacements are only partly responsible for the finding that the damping parameters obtained for Py doped with the 5*d* TM series, and $Fe_{1-x}Co_x$ are somewhat low compared with experiment. This indicates that additional relaxation mechanisms like magnon scattering contribute. Again, these can be included at least in a quasistatic way by adopting the point of view of a disordered local moment picture. This implies scattering due to random temperature-dependent fluctuations of the spin moments that can also be dealt with using the CPA.

In summary, a formulation for the Gilbert damping parameter α in terms of a torque-torque-correlation function was derived that led to a Kubo-Greenwood-like equation. The scheme was implemented using the fully relativistic Korringa-Kohn- Rostoker band structure method in combination with the CPA alloy theory. This allows us to account for various types of scattering mechanisms in a parameter-free way. Corresponding applications to disordered transition metal alloys led to very good agreement with results based on the scattering theory approach of Brataas *et al.* demonstrating the equivalence of both approaches. The flexibility and numerical efficiency of the present scheme was demonstrated by a study on a series of Permalloy-5*d* TM systems. To investigate the influence of finite temperatures on α, a so-called alloy-analogy model was introduced that deals with the thermal displacement of atoms in a quasistatic manner. Applications to pure Ni gave results in very good agreement with experiment and, in particular, reproduced the dramatic change of α when Cu is added to Ni.

The authors would like to thank the DFG for financial support within the SFB 689 "Spinphänomene in reduzierten Dimensionen" and within project Eb154/23 for financial support. P. J. K acknowledges support by EU FP7 ICT Grant No. 251759 MACALO.

[1] T. L. Gilbert, IEEE Trans. Magn. **40**, 3443 (2004).
[2] V. Kambersky, Can. J. Phys. **48**, 2906 (1970).
[3] V. Kambersky, Czech. J. Phys. **26**, 1366 (1976).
[4] K. Gilmore, Y. U. Idzerda, and M. D. Stiles, Phys. Rev. Lett. **99**, 027204 (2007).
[5] M. Fähnle and D. Steiauf, Phys. Rev. B **73**, 184427 (2006).
[6] V. Kambersky, Phys. Rev. B **76**, 134416 (2007).
[7] A. Brataas, Y. Tserkovnyak, and G. E. W. Bauer, Phys. Rev. Lett. **101**, 037207 (2008).
[8] A. A. Starikov, P. J. Kelly, A. Brataas, Y. Tserkovnyak, and G. E. W. Bauer, Phys. Rev. Lett. **105**, 236601 (2010).
[9] Y. Liu, A. A. Starikov, Z. Yuan, and P. J. Kelly, Phys. Rev. B **84**, 014412 (2011).
[10] W. H. Butler, Phys. Rev. B **31**, 3260 (1985).
[11] H. Ebert, in *Electronic Structure and Physical Properties of Solids*, edited by H. Dreyssé, Lecture Notes in Physics Vol. 535 (Springer, Berlin, 2000), p. 191.
[12] M. E. Rose, *Relativistic Electron Theory* (Wiley, New York, 1961).
[13] H. Ebert and S. Mankovsky, Phys. Rev. B **79**, 045209 (2009).
[14] E. M. Gololobov, E. L. Mager, Z. V. Mezhevich, and L. K. Pan, Phys. Status Solidi (b) **119**, K139 (1983).
[15] D. Ködderitzsch, H. Ebert, D. A. Rowlands, and A. Ernst, New J. Phys. **9**, 81 (2007).
[16] J. O. Rantschler, R. D. McMichael, A. Castillo, A. J. Shapiro, W. F. Egelhoff, B. B. Maranville, D. Pulugurtha, A. P. Chen, and L. M. Connors, J. Appl. Phys. **101**, 033911 (2007).
[17] G. Woltersdorf, M. Kiessling, G. Meyer, J.-U. Thiele, and C. H. Back, Phys. Rev. Lett. **102**, 257602 (2009).
[18] B. Drittler, N. Stefanou, S. Blügel, R. Zeller, and P. H. Dederichs, Phys. Rev. B **40**, 8203 (1989).
[19] N. Stefanou, A. Oswald, R. Zeller, and P. H. Dederichs, Phys. Rev. B **35**, 6911 (1987).
[20] M. Oogane, T. Wakitani, S. Yakata, R. Yilgin, Y. Ando, A. Sakuma, and T. Miyazaki, Jpn. J. Appl. Phys. **45**, 3889 (2006).
[21] S. M. Bhagat and P. Lubitz, Phys. Rev. B **10**, 179 (1974).