We present a microscopic theory for magnetization relaxation in metallic ferromagnets of nanoscopic dimensions that is based on the dynamic spin response matrix in the presence of spin-orbit coupling. Our approach allows the calculation of the spin excitation damping rate even for perfectly crystalline systems, where existing microscopic approaches fail. We demonstrate that the relaxation properties are not completely determined by the transverse susceptibility alone, and that the damping rate has a non-negligible frequency dependence in experimentally relevant situations. Our results indicate that the standard Landau-Lifshitz-Gilbert phenomenology is not always appropriate to describe spin dynamics of metallic nanostructure in the presence of strong spin-orbit coupling.

Magnetization relaxation in metals is at the heart of spin current generation and detection processes currently under investigation, many of them candidates to play protagonist roles in innovative spintronic devices. The Landau-Lifshitz-Gilbert (LLG) equation is widely used to describe the spin dynamic properties of magnetic materials [1,2]. It includes an important system-dependent parameter, called the Gilbert damping constant, usually denoted by $\alpha_G$, that regulates the relaxation of the magnetization towards stability, after it is driven out of equilibrium. Recently, a lot of effort has been put into the determination of this damping rate [2-8], which characterizes the pumping and absorption of pure spin currents in nanostructures that are of great interest in the field of spintronic. In most of them spin-orbit interaction is significant, and responsible for a desirable interplay between charge spin and angular momentum excitations.

There is a general agreement between practitioners in the field that a proper microscopic theory of magnetization relaxation in metals requires a good description of the electronic structure of the system including spin-orbit coupling [3-8]. The conventional approach is to combine a realistic electronic structure with some kind of adiabatic approximation to derive expressions that can be directly related to the Landau-Lifshitz-Gilbert phenomenology. This strategy has been employed by Kamberský [3] and many others since [4-8]. This conventional approach has important limitations. It neglects the coupling between transverse spin, longitudinal spin and charge excitations (which is an important consequence of the spin-orbit coupling), and incorrectly predicts the divergence of the damping parameter for a perfectly crystalline system. Actually, for ferromagnets that display rotation symmetry in spin space, the Goldstone theorem ensures that any experiment which measures the total transverse magnetic moment of the sample will produce a resonant response with zero linewidth [9]. In the presence of spin-orbit interaction, however, this symmetry is explicitly broken, and the resonant spectrum acquires a finite linewidth [10].

We put forward a more fundamental microscopic approach to the calculation of the spin dynamics damping rate that takes fully into account the effects of SOC on the spectrum of spin excitations of itinerant systems. Namely, we consider the coupling of transverse spin excitations to longitudinal spin and charge excitations, induced by the spin-orbit interaction. We calculate the FMR spectrum at finite frequencies and arbitrary anisotropy values, without employing any adiabatic approximation. We will show that those ingredients are essential to correctly describe the magnetization relaxation in very clean metallic ferromagnets of nanoscopic dimensions, and that the Landau-Lifshitz-Gilbert phenomenology fails to capture essential features of the magnetization dynamics in those systems.

This letter is organized as follows: we will present briefly our formalism, discuss its main features and present numerical results for two model systems that illustrate common but qualitatively different situations.

**General Formalism** - The spectrum of spin excitations of a ferromagnet can be obtained from the spectral density associated with the transverse spin susceptibility

$$
\chi^+(l, l'; \Omega) = \int dt e^{i\Omega t} \langle\langle S^+_l(t), S^-_{l'}(0)\rangle\rangle,
$$

where

$$
\langle\langle S^+_l(t), S^-_{l'}(0)\rangle\rangle = -i\theta(t)\langle[S^+_l(t), S^-_{l'}(0)]\rangle,
$$

and

$$
S^+_l = \sum_\mu a^\dagger_{\mu l} a_{\mu l}.
$$

The operator $a^\dagger_{\mu l}$ creates one electron in the atomic basis state $\mu$ localized at lattice site $l$ with spin $\sigma$. Although we are usually interested in $\chi^+(l, l'; \Omega)$ as defined above, its equation of motion involves the orbital-resolved susceptibility,

$$
\chi^+_{\mu\sigma l, \mu'\sigma' l'}(l, l'; t) \equiv \langle\langle a^\dagger_{\mu l}(t) a_{\mu' l'}(t), a^\dagger_{\mu' l'}(0) a_{\mu l}(0)\rangle\rangle.
$$

In the absence of spin-orbit coupling (SOC) and within the random phase approximation (RPA), the equation of

**Microscopic theory of Gilbert damping in metallic ferromagnets**

A. T. Costa and R. B. Muniz

Instituto de Física, Universidade Federal Fluminense, 24210-346 Niterói, RJ, Brazil.
motion for $\chi^{+}(l,l';t)$ is closed and $\chi^{+}(l,l';\Omega)$ can be expressed in the well-known RPA form,

$$\chi^{+}(\Omega) = [1 + U\chi^{+}_0(\Omega)]^{-1}\chi^{+}_0(\Omega)$$

where $\chi^{+}_0(\Omega)$ is the mean-field (sometimes called non-interacting, or Hartree-Fock) susceptibility. This expression is schematic and must be understood as a matrix in orbital and site indices, in real space, or a wave-vector dependent matrix in reciprocal space. The crucial point, however, is that, in the absence of spin-orbit coupling, within the RPA, the transverse spin susceptibility is uncoupled from any other susceptibility. This ceases to be true when SOC is included, as we demonstrated in ref. [10] $\chi^{+}$ becomes coupled to three other susceptibilities, namely

$$\chi^{(2)}_{\mu'\mu''\nu'}(l,l';t) \equiv \langle a_{\mu'\tau}^{\dagger}(t) a_{\mu\nu}(t), a_{\mu'\tau}^{\dagger}(0) a_{\mu''\nu'}(0) \rangle,$$

$$\chi^{(3)}_{\mu'\mu''\nu'}(l,l';t) \equiv \langle a_{\mu'\tau}^{\dagger}(t) a_{\mu\nu}(t), a_{\mu'\tau}^{\dagger}(0) a_{\mu''\nu'}(0) \rangle,$$

$$\chi^{(4)}_{\mu'\mu''\nu'}(l,l';t) \equiv \langle a_{\mu'\tau}(t) a_{\mu\nu'}(t), a_{\mu'\tau}(0) a_{\mu''\nu'}(0) \rangle.$$ 

The system of equations of motion obeyed by these four susceptibilities can be cast into a form strongly resembling the RPA result by introducing a block-vector $\tilde{\chi} \equiv (\chi^{(1)}, \chi^{(2)}, \chi^{(3)}, \chi^{(4)})^T$, with $\chi^{(1)} \equiv \chi^{+}$. With an equivalent definition for the mean-field susceptibilities $\chi^{(m)}_0$ we write

$$\tilde{\chi}(\Omega) = \tilde{\chi}_0(\Omega) - \Lambda \tilde{\chi}(\Omega),$$

where the “super-matrix” $\Lambda$ is proportional to the effective Coulomb interaction strength and involves convolutions of single particle Green functions. Explicit forms for its matrix elements are found in Ref. [10]. The numerical analysis of the susceptibilities $\chi^{(2)}$, $\chi^{(3)}$ and $\chi^{(4)}$ show that their absolute values are many orders of magnitude smaller than those of $\chi^{(1)} \equiv \chi^{+}$. It is, thus, tempting to argue that the transverse susceptibility is approximately decoupled from $\chi^{(2)}$, $\chi^{(3)}$ and $\chi^{(4)}$ and that it can be calculated via the usual RPA expression with the single particle Green functions obtained with spin-orbit coupling taken into account. This is not a good approximation in general, since the matrix elements of $\Lambda$ that couple $\chi^{(1)}$ to the other susceptibilities are far from negligible. Our numerical calculations indicate that they are essential to determine correctly the features of the FMR mode around the resonance frequency. Thus, the behaviour of $\chi^{(1)}$ in the presence of spin-orbit coupling cannot be inferred from $\chi^{(1)}_0$ in the zero-frequency limit alone, as it is usually assumed in the literature on the calculation of the Gilbert damping parameter [9].

**Numerical Results** - We start the discussion by presenting results for the Gilbert constant $\alpha_G$ for unsupported ultrathin Co films. Here we determine $\alpha_G$ from the ratio between the FMR linewidth $\Delta\Omega$ and the resonance frequency $\Omega_0$. First we turn off spin-orbit coupling to check the consistency of our approach. Even with SOC turned off we still find a finite linewidth for the FMR mode. It comes, as we will shortly demonstrate, from the small imaginary part $\eta$ that is usually added to the energy in the numerical calculations of the single particle Green functions, in order to move their poles from the real axis. We calculate $\alpha_G$ for various values of $\eta$ and extrapolate to $\eta \to 0^+$, as shown in Fig. 1. It is clear that $\lim_{\eta \to 0^+} \alpha_G = 0$. Thus, our approach correctly predicts that the Gilbert damping constant vanishes in the absence of SOC, as it should. Indeed, it is easy to show [9] that the FMR mode is a stationary state of the mean-field hamiltonian and, as such, has infinite lifetime in the limit $\eta \to 0^+$. Now we discuss the dependence of $\alpha_G$ on $\eta$ for a fixed, non-zero value of the spin-orbit coupling strength $\xi$. We used LCAO parameters appropriate for bulk Co to describe the electronic structure of all Co films we investigated. The quantitative details of the ferromagnetic ground state and excitation spectra are sensitive to the LCAO parameters used, but their qualitative behaviour is very robust to small changes in the electronic structure. Our strategy is to use the same set of LCAO parameters for all film thicknesses to avoid modifications in $\alpha_G$ coming directly from changes in the LCAO parameters. This allows us to focus on geometric effects and on the $\eta$-dependence.

Figure 2 shows the dependence of the Gilbert damping constant $\alpha_G$ on the imaginary part $\eta$ for Co films of various thicknesses. Clearly $\alpha_G$ approaches finite values as $\eta \to 0$. Cobalt has a small spin-orbit coupling constant. We would like to investigate the effect of increasing the strength of the SOC on the damping rate. Instead of artificially increasing $\xi$ in Co we consider a more realistic setting where a double layer of Co is attached to a
non-magnetic substrate with high SOC parameter, such as Pt. This system has a particularly interesting feature: the magnetization easy axis is perpendicular to the plane. However, we found that, for the LCAO parameters we employed, the magnetization in-plane is also a stable configuration, with a small magnetocrystalline anisotropy. The damping rate, however, is much larger in the 2Co/2Pt system than in the unsupported Co films. This is a nice example of how the anisotropy energy is strongly influenced by the system’s symmetry, but the damping rate is relatively insensitive to it, depending strongly on the intensity of the spin-orbit coupling. It is also an extremely convenient situation to test an assumption very frequently found in the literature on Gilbert damping, although sometimes not explicitly stated: that the FMR linewidth $\Delta \Omega$ is linearly dependent on the resonance frequency $\Omega_0$ and that $\Delta \Omega \to 0$ as $\Omega_0 \to 0$. This is not an unreasonable hypothesis, considering the weak static fields commonly used in FMR experiments and the smallness of the spin-orbit coupling constant, compared to other energy scales of a ferromagnet. Our calculations for the Co films confirm that this relationship is approximately held. In this case, the Gilbert constant $\alpha_G$ may be extracted from the FMR spectrum by simply fitting it to a Lorentzian and is practically field-independent. However, our results for 2Co/2Pt indicate that the FMR linewidth is finite as $\Omega_0 \to 0$, leading to a significantly frequency-dependent $\alpha_G$, as shown in Fig. 3. In order to illustrate how the determination of a damping parameter is affected by the finite value of $\Delta \Omega$ as $\Omega_0 \to 0$ we extracted the linewidths from the calculated spectra for the 2Co/2Pt system by fitting Lorentzians to our calculated spectral densities. The results are shown in Fig. 3. One of its most important consequences is that, if one wishes to define a value of $\alpha_G$ for the system above, it must be defined as a function of the Zeeman field, as is illustrated in Fig. 3. In principle this poses a problem for the procedure usually employed to determine FMR spectra experimentally, since there the free variable is the Zeeman field, not the frequency of the exciting field. In Fig. 4 we illustrate this issue by plotting the FMR spectral density as a function of the Zeeman field for two fixed pumping frequencies, 24 GHz and 54 GHz. The curves have nice Lorentzian shapes, but the values for the Gilbert damping parameter $\alpha_G$ extracted from these curves depend on the pumping frequency ($\alpha_G = 0.034$ for $\Omega_0 = 0.10$ meV and $\alpha_G = 0.042$ for $\Omega_0 = 0.22$ meV). Also, they do not correspond to any of the values shown in Fig. 3, although the Zeeman field values that determine the linewidth in Fig. 4 lie within the range of Zeeman field values showed in Fig. 3. Thus, if $\alpha_G$ is defined as $\Delta \Omega/\Omega_0$, its value for a given sample depends
FIG. 4: Spectral density of the FMR mode for the 2Co/2Pt system plotted as a function of the Zeeman field $B$ at a fixed pumping frequencies: $\nu_p = 24$ GHz (squares) and $\nu_p = 54$ GHz (circles). The solid curves are Lorentzian fits to the calculated points.

FIG. 5: a) Spectral densities of the FMR mode for the 2Co/2Pt system with Stoner enhancement in Pt turned on (black line) and off (red line).

on weather the FMR spectrum is obtained in a fixed frequency or fixed Zeeman field set ups. Our results also imply that the existing expressions for the damping constant $\alpha_G$ are not valid in general, specially for very clean systems with large spin-orbit coupling materials. The conventional approaches express $\alpha_G$ as the ratio $\Delta \Omega/\Omega_0$ in the $\Omega_0 \to 0$ limit. As we have just shown, this limit does not exist in general, since $\Delta \Omega$ approaches a finite value as $\Omega_0 \to 0$.

In experimental papers [12, 13] the FMR linewidth is assumed to have a zero-frequency offset, just as we described. This is usually attributed to extrinsic broadening mechanisms, such as two-magnon scattering [14], due to the combination between inhomogeneities in the magnetic films and dipolar interactions. This is certainly the case in systems with small SOC, such as Fe films deposited on GaAs or Au [12]. However, we have shown that there can be zero-frequency offset of intrinsic origin if the SOC is large. The effect of this intrinsic offset should be easily separated from that of the two-magnon scattering mechanism, since the latter is not active when the magnetization is perpendicular to the plane of the film [14].

We would like to remark that Stoner enhancement in Pt plays a very important role in the determination of the damping rate. We had shown previously [15] that, in the absence of spin-orbit coupling, Stoner enhancement had a very mild effect on the damping rate in the Co/Pd(001) system. In the presence of SOC, however, the effect can be very large indeed. Both magnetocrystalline anisotropy and damping rate are significantly different in the enhanced and non-enhanced cases, as shown in Fig. 5. The Gilbert parameter is also very different in the two cases: $\alpha_G^{\text{enh}} = 0.11$, whereas $\alpha_G^{\text{non-enh}} = 0.33$. Thus, proper treatment of Stoner enhancement in substrates like Pd an Pt is essential for the correct determination of spin relaxation features.

We presented a microscopic approach to the calculation of the Gilbert damping parameter $\alpha_G$ for ultra-thin metallic magnetic films, illustrated by results for Co films and Co/Pt bilayers. Our approach is based on the evaluation of the dynamic transverse susceptibility in the presence of spin-orbit coupling, taking into account realistic electronic structures and the coupling between transverse spin, longitudinal spin and charge excitations. It predicts finite values of $\alpha_G$ in the limit of perfectly crystalline films, a regime where methods based on the torque correlation formula find a diverging Gilbert damping parameter. We showed that the coupling between transverse, longitudinal and charge excitations, due to spin-orbit coupling, is of fundamental importance for the correct determination of FMR spectra in metallic systems. We have also shown that the damping rate extracted from the FMR spectrum for fixed pumping frequency differs considerably from that extracted from the FMR spectrum for fixed Zeeman field. In this case the Gilbert damping parameter $\alpha_G$ becomes frequency dependent, in contrast to what is assumed in the standard Landau-Lifshitz-Gilbert phenomenology. Moreover, we have numerical indications that the Gilbert parameter is not well defined in the limit of vanishing resonance frequency, a fact that is very relevant to calculational schemes based on the adiabatic approximation. Incidentally, Stoner enhancement in materials like Pt and Pd also plays an important role in the determination of FMR frequencies and damping rates. These results may lead to important modifications of the interpretation of damping "constants", either calculated or inferred from...
experimental results, for systems where spin-orbit coupling is strong. We believe these issues may be crucial for the correct description of relaxation in very clean systems of nanoscopic dimensions, specially in the presence of relatively weak magnetocrystalline anisotropy.

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