Role of electron scattering in the magnetization relaxation of thin Ni$_{81}$Fe$_{19}$ films

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We observe a strong correlation between magnetization relaxation and electrical resistivity in thin Permalloy (Ni$_{81}$Fe$_{19}$, "Py") films. Electron scattering rates in the films were affected by varying film thickness and deposition conditions. This shows that the magnetization relaxation mechanism is analogous to "bulk" relaxation, where phonon scattering in bulk is replaced by surface and defect scattering in thin films. Another interesting finding is the increased magnetization damping with Pt layers adjacent to the Py films. This is attributed to the strong spin-orbit coupling in Pt, resulting in spin-flip scattering of electrons that enter from the Py.

INTRODUCTION

The Gilbert form of the Landau-Lifshitz equation describes the small-angle precession of magnetization in a ferromagnet:

$$\frac{d\boldsymbol{M}}{dt} = -\gamma \boldsymbol{M} \times \boldsymbol{H}_{\text{eff}} - \frac{\alpha}{M} \boldsymbol{M} \times \frac{d\boldsymbol{M}}{dt}. \tag{1}$$

Here $\boldsymbol{M}$ is the magnetization and $\gamma = g|e|/2mc$ is the gyromagnetic ratio, and α is the Gilbert damping coefficient that affects the magnitude of the viscous damping term. $\boldsymbol{H}_{\text{eff}}$ is the effective magnetic field "seen" by the magnetization, and is expressed in terms of the free energy as $\boldsymbol{H}_{\text{eff}} = -\nabla_{\boldsymbol{M}} \mathcal{F}$. The Gilbert damping coefficient α controls how rapidly the magnetization equilibrates in the absence of external stimulus. This obviously makes α a key parameter in the description of high-speed dynamics in magnetic materials. A few areas where α plays a vital role are in devices that rely on fast magnetization reversal (e.g., in giant magnetoresistive,[1] magnetic tunnel junction,[2] or other spintronic devices), current-induced magnetization reversal,[3] the generation of microwaves by spin currents,[4] etc.

By the 1970s it was shown that intrinsic magnetization relaxation in transition-metal ferromagnets could be explained by electron scattering by phonons and magnons. The former process, mediated by the spin-orbit interaction, occurs both with[5,6] and without[6–9] the accompaniment of a spin-flip. In the former case $\alpha \sim \tau^{-1}$, where τ^{-1} is the electron-scattering rate. In the latter case the angular momentum relaxes as scattered electrons repopulate the magnetization-direction-dependent Fermi volume, and $\alpha \sim \tau$ is expected. Magnon modes can relax through exchange interaction with a conduction electron, causing its spin to flip (this can be viewed simplistically as s-d exchange accompanied by a spin-flip of the s electron).[10–12] The conduction electron spin then relaxes to the lattice through the spin-orbit interaction. This also results in $\alpha \sim \tau^{-1}$ to leading order.

More recently, interesting effects were predicted for ultrathin films and for multilayers that would contribute to the effective α in Eq. (1). Arias and Mills showed that under certain conditions in ultrathin films the uniform mode (k

$=0$), excited by ferromagnetic resonance (FMR), can be scattered into $k \neq 0$ magnons by anisotropic surface defects, where k is the wave vector.[13] This two-magnon scattering theory was used by Azevedo *et al.* to explain experimental results of FMR linewidth and resonance field in NiFe films.[14] Berger predicted that transfer of electron spin angular momentum between two ferromagnetic layers, separated by a nonmagnetic layer, contributes to the magnetization relaxation (i.e., α).[15] The experimental results of Urban *et al.* confirmed an increase in the FMR linewidth with two layers of Fe separated by a nonmagnetic layer, compared to a single layer of Fe.[16]

We have studied FMR and electronic transport in NiFe films (Ni$_{81}$Fe$_{19}$, "Py" for short), in which the electronic scattering rates were affected over a wide range by (a) changing the surface scattering contribution by varying the film thickness, (b) changing the film deposition conditions, and (c) choosing different interfaces and surface treatment. We observe a strong correlation between the Gilbert damping coefficient and resistivity, i.e., $\alpha \propto \rho$ in our single-layer Py samples. Our results show that the dominant magnetization relaxation mechanism in these samples involves electron scattering, and is seemingly insensitive to whether the scattering occurs within the "bulk" of the films or at the surface. This explains why α is observed to increase with decreasing film thickness.[17] It also implies that the effective α in magnetic devices, small in at least one dimension, made with transition metals or alloys, is expected to be considerably larger than that intrinsic to the bulk material, due to an increased surface/volume ratio and to enhanced spin relaxation at interfaces.[18] Our data also appear inconsistent with the two-magnon scattering theory. Further, we observe that with nonmagnetic (NM) Pt enclosing our Py films, the magnetization relaxation is significantly enhanced, in addition to the electron scattering related mechanism above. The enhancement is attributed to spin relaxation of conduction electrons that leave the Py layer, both at the interfaces and within the NM layers.

EXPERIMENT

Our NiFe films were deposited by dc-magnetron sputtering in a vacuum system with a base pressure of 2

$\times 10^{-8}$ torr. During deposition they were (all but one series) exposed to a uniform magnetic field of ~ 150 Oe to induce uniaxial in-plane anisotropy. X-ray results show that the Py films are (111) textured. We made two series of samples of Si/SiO$_2$/Py/PR, where PR is photoresist, used to protect the film from oxidation. One of these (called *o*-Py) was grown in a uniform applied magnetic field, the other (called *d*-Py) without a deliberately applied field. Both series have a clearly defined easy axis, the direction of which in the *d*-Py was presumably defined by the Earth's field. Resistivity measurements on these samples were made using the van der Pauw method. They indicate that the *d*-Py series has more disorder than the *o*-Py ("ordered") series. We also studied the effect of adjoining Py layers with 80-Å-thick NM-metallic layers, i.e., Si/SiO$_2$/X/Py/X, where X is Cu, Nb, or Pt. The Py thickness within each sample series was varied by depositing a terraced structure on a single wafer using a movable shadow mask. The films were then lithographically patterned into arrays of discs, 1 or 2 mm in diameter. From each deposition we thus obtained a series of samples of different thickness, with minimal variations in growth conditions. We also made one sample of Si/SiO$_2$/Py, 1000 Å thick, and ion milled it several times, measuring its thickness and magnetic properties between millings.

To obtain the Gilbert damping in our samples we measured their in-plane magnetic susceptibility in an FMR experiment with swept frequency and fixed dc magnetic field. The experimental setup is essentially the same as that of Korenivski *et al.*[19] The magnetic softness of Py ($H_c \leq 4$ Oe) allows the experiments to be done with applied dc fields $H \leq 150$ Oe. Our films are thinner than the skin depth at the corresponding resonance frequencies, i.e., $\omega_r/(2\pi) \leq 3.5$ GHz for the uniform mode of spin precession. The exchange stiffness in Py causes higher-order spin wave modes to appear at much higher frequencies. The ac field is considered uniform throughout the films and a quasistatic approximation relates the internal and external fields. These assumptions are supported by the Lorentzian line shape of our resonance peaks, and holds even for the thickest samples of ~ 1000 Å. The FMR experiments were done at a small precession-cone angle, the ratio of the amplitudes of the ac- and dc-fields being $\sim 10^{-4}$. There was no detectable change in the FMR (susceptibility) when the rf power was increased by 15 dB.

The conditions above allow us to fit the susceptibility very well with a linearized form of Eq. (1). The free energy $\mathcal{F}$, includes the Zeeman energy, a demagnetization term, uniaxial in-plane anisotropy and a uniaxial out-of-plane (interface) anisotropy term. In the coordinate system shown in Fig. 1, under the assumption that the applied dc magnetic field $\boldsymbol{H}$ is in plane, $\mathcal{F}$ can be expressed as

$$\mathcal{F} = -MH \sin\theta \sin(\phi+\psi) + 2\pi M^2 \cos^2\theta$$

$$+ K_u \sin^2\theta \cos^2\phi + 2\frac{K_s}{d}\cos^2\theta, \qquad (2)$$

where K_u is an in-plane uniaxial anisotropy constant, $K_s = (K_{s1} + K_{s2})/2$ is a surface anisotropy constant representing
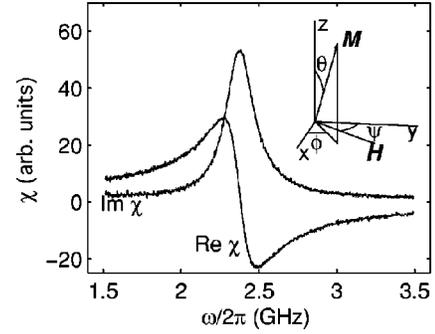
FIG. 1. Real and imaginary parts of the susceptibility as a function of frequency of a 80Nb/477Py/80Nb sandwich structure (numbers denote layer thickness in Å) structure with $H_{dc} = 60$ Oe along the easy axis of the film, $\phi = 0$ and $\theta = \psi = \pi/2$. Also shown is the coordinate system used to describe the free energy of the film lying in the *x-y* plane with the easy axis in the *x* direction.

the average anisotropy of the upper and lower surfaces, and d is the film thickness. The in-plane uniaxial anisotropy is determined by fitting the angular variation of the resonance frequency ω_r in the plane of the samples. Typical results are shown in Fig. 2, which displays a fit to $\omega_r/2\pi$ as a function of the equilibrium magnetization angle ϕ.

For the free energy given in Eq. (2) the resonance frequency is given by

$$\left(\frac{\omega_r}{\gamma}\right)^2 = \left(H\sin(\phi+\psi) + 4\pi M - \frac{2K_u}{M}\cos^2\phi + \frac{4K_s}{dM} \right)$$

$$\times \left(H\sin(\phi+\psi) - \frac{2K_u}{M}\cos(2\phi) \right). \qquad (3)$$
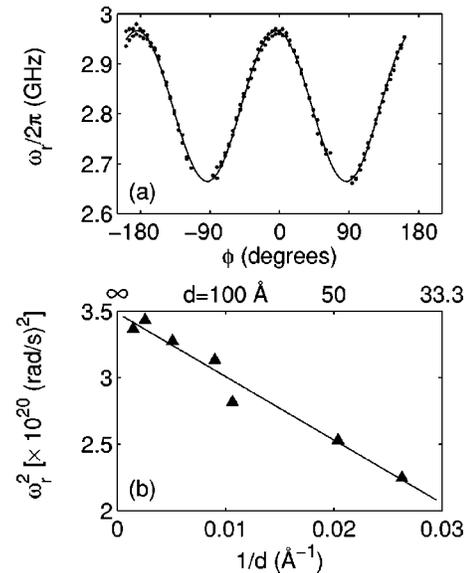
FIG. 2. (a) The dependence of the resonance frequency on the in-plane magnetization angle in a 682-Å-thick sample from the *o*-Py series, with $H_{dc} = 90$ Oe in the plane of the film. From a fit to the oscillatory angular dependence we obtain the in-plane anisotropy field, here $H_k = 10$ Oe. (b) Resonance frequency ω_r vs sample thickness d for the *o*-Py series. The shift in ω_r^2 scales as $1/d$. This is accounted for by surface anisotropy, in accord with Eq. (3). The line is a least-squares fit and corresponds to $K_s = 0.28$ erg/cm^2.
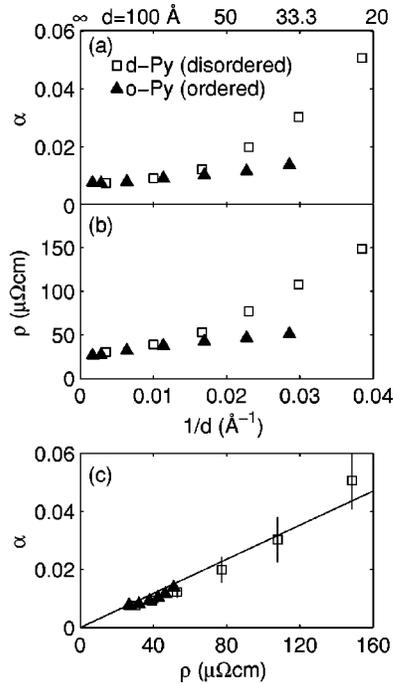
FIG. 3. (a) Gilbert damping coefficient α and (b) resistivity ρ of the d-Py (disordered) and o-Py (ordered) series as function of inverse film thickness $1/d$, at room temperature. The two series exhibit quite different thickness dependences. (c) displays the correlation between α and ρ.

With the applied field and the magnetization coincident with the easy axis of the sample, i.e., $\phi = 0$ and $\psi = \theta = \pi/2$, for each series of samples we use the leading terms in the field dependence of ω_r in Eq. (3) to obtain an initial estimate of γ. Similarly we use the thickness dependence of ω_r to obtain an initial value for K_s, as shown in Fig. 2(b). Subsequently we obtain the Gilbert damping parameter α, and refined values for K_s and γ by fitting the complex susceptibility. With this approach we arrive at a self-consistent result where a constant K_s accounts for the shift in the resonance frequency as a function of film thickness *and* where the g value is constant for any given series of samples.

Even without our careful determination of the above-mentioned values we could obtain a good estimate of α, since its effect on the resonance curve cannot be mimicked by adjusting other parameters. However, as the damping increases, the resonance peak gradually disappears, and it becomes difficult to estimate α accurately, even with our fitting procedure. This is reflected in the size of the error bars in Fig. 3(c). We emphasize that in our treatment α is the dimensionless Gilbert damping coefficient and not the frequency linewidth. Although we observe the linewidth following the same trends as α as a function of film thickness, we believe that the fitting procedure gives more reliable results than measuring the linewidth. Also, α represents the total effective damping, including both intrinsic and extrinsic damping, such as surface damping.

RESULTS AND DISCUSSION

The Gilbert damping coefficients α, for the two PR-coated series (o-Py and d-Py) are displayed in Fig. 3(a), as functions of the inverse film thickness $1/d$. Values for the thickest films approach the bulk damping, but as the thickness decreases α increases dramatically. In the o-Py the damping doubles from the thickest film to the thinnest. The effect is much more pronounced in the d-Py, where the thinnest film has roughly six times the bulk damping value. The room-temperature resistivity $\rho(d)$ of these two series is shown in Fig. 3(b). The changes in ρ reflect quite accurately the corresponding changes in α, i.e., $\Delta\rho/\rho_0 \simeq \Delta\alpha/\alpha_0$, where the subscript 0 refers to thick film values. This suggests the existence of a simple relationship between α and ρ, which is addressed below. The reason for plotting these data as function of $1/d$ is, that if one naively assumes that α can be separated into independent bulk and surface contributions a $1/d$ dependence is expected as the surface/volume ratio changes. This is analogous to the assumption made for electron scattering in the Fuchs-Sondheimer theory of surface scattering.[20]

However, neither the o-Py nor d-Py series exhibit a perfectly linear relationship between α, or ρ, and $1/d$, although the deviation of the o-Py samples from a straight line appears small. Neglecting the small deviation, a Fuchs-Sondheimer-type analysis of the o-Py resistivity results in a bulk resistivity at room temperature of $\rho_b = 24$ $\mu\Omega$ cm and mean free path $\lambda = 96$ Å (the corresponding low temperature, or residual, values are $\rho_{b,\mathrm{res}} = 14$ $\mu\Omega$ cm and $\lambda_{\mathrm{res}} = 215$ Å). It is clear from the significant departure of ρ, in the d-Py series, from the linear $1/d$ dependence, that the simple assumptions of the Fuchs-Sondheimer model do not hold there. Additional scattering mechanisms such as impurity and grain boundary scattering should be taken into account.[20] In any case, it is evident from the small resistivity ratios in both series, $\rho_{295\mathrm{K}}/\rho_{\mathrm{res}}$ ranging from 1.85 for the thickest films to 1.1 for the thinnest films, that scattering associated with film surfaces and defects (including grain boundaries) accounts for the major share of the total resistivity in all of these films. The strongest T-dependent contribution to $\rho(T)$ at low T is proportional to T^2, which is attributed to electron-electron and electron-magnon scattering.[21]

When the data in Figs. 3(a) and 3(b) are plotted together, as in Fig. 3(c), they fall on a single curve. Note that the thickness of the films in the overlap region is quite different for the two series. For instance, the thinnest o-Py film in Fig. 3(c) is 35 Å thick, and it corresponds roughly to a d-Py film of thickness 65 Å. At least to leading order, a simple proportionality, describable by *one* constant, of experimental α to ρ as functions of d is apparent in Fig. 3(c). However, an examination of the data in Figs. 3(a) and 3(b) reveals that even *two* parameter fits of α, or $\rho = a + bd^{-n}$ to the data would not be fully satisfying. These remarks suggest immediately that a significant contribution to viscous damping in very thin ferromagnetic metals is connected with the electron scattering, giving rise to resistivity in a general way not depending on whether the electron scattering is by phonons, defects, or surface irregularities.

Theoretical estimates by Kambersky[5] are particularly helpful in distinguishing a low-temperature damping term proportional to the electron-scattering time τ from a high-temperature term proportional to τ^{-1}, when electron scatter-

ing by phonons is prevalent. Electron scattering in our samples is dominated by surface and defect scattering. It is mainly caused by fixed electrostatic potentials associated with compositional interdiffusion and structural irregularities. It is therefore more appropriate to use the results of Heinrich *et al.*[10] to account for our data. They considered the effect of *s*-electron spin relaxation on *s-d* exchange and magnetization relaxation:

By definition, the equation $\alpha = \lambda/\gamma M$ relates the dimensionless Gilbert damping coefficient to the Landau-Lifshitz parameter λ. A combination of Eq. (21) in Ref. 10 and the assumption that the spin-relaxation rate $\tau_s^{-1} \sim \zeta \tau^{-1}$, where ζ is a constant, and τ^{-1} is the ordinary electron scattering rate (characteristic of electrical resistivity, i.e., it includes both scattering events that are accompanied by a spin-flip and those that are not), yields the estimate

$$\alpha \simeq \frac{\zeta \gamma m^* k_F}{2\pi^2 M \tau}. \qquad (4)$$

Here γ is the free-electron gyromagnetic ratio, m^* the effective mass of the *s* electron, and k_F the Fermi wave vector. We have neglected a term $H/(4\pi M) \sim 10^{-2}$ compared to unity. We now eliminate the scattering rate with the Drude conductivity formula $\sigma = \rho^{-1} = ne^2\tau/m^*$. Further, we crudely estimate $M \simeq \mu_B n$ with $\mu_B = \hbar\gamma/2$ the Bohr magneton and n the atomic density, to obtain

$$\frac{\alpha}{\zeta \rho} \simeq \frac{e^2 k_F}{\pi^2 \hbar} \simeq 3.7 \times 10^5 \,(\Omega\mathrm{m})^{-1}, \qquad (5)$$

estimated for a 3d metal or alloy.

The solid line in Fig. 3(c), obtained by a linear least-squares fit constrained to go through the origin, has a slope of 3×10^4 $(\Omega\mathrm{m})$. To satisfy Eq. (5) we must then have $\zeta \sim 10^{-1}$, i.e., $\tau_s^{-1} \sim 10^{-1}\tau^{-1}$. For comparison we observe that the diffuse scattering of an electron is represented by a random walk process, where the spin diffusion length $l_{\mathrm{sf}} = \lambda(\tau_s/\tau)^{1/2}$, λ is the mean free path of the electron, and τ_s/τ is the number of spin-preserving scattering events before the spin is flipped. With published values for Py of $l_{\mathrm{sf}} = 55$ Å,[22] and mean free paths $\lambda_\downarrow \lesssim 6$ Å and $\lambda_\uparrow = 46$ Å,[23] we estimate the ratio $(\tau_s/\tau)^{-1}$ to be 0.01 and 0.7 for the down- and up-spin bands, respectively. Our result lies well within that range.

Alternative to the electron scattering mechanism, one may also consider that of two-magnon scattering.[13] Since both mechanisms involve interfacial effects, they both predict a increased damping with decreasing film thickness. But, to account for the plot in Fig. 3(c), one must show how two-magnon scattering predicts an *equal damping* in two films having *unequal thicknesses* yet exhibiting *equal resistivities* because of a compensating difference in their preparation. The surface roughness of our samples, determined by atomic force microscopy, was the same 6 ± 1 Å$_{\mathrm{rms}}$, both within *and* between the two series. Furthermore, the measured surface anisotropies of the *o*-Py and *d*-Py samples are constant and identical, i.e., $K_s = 0.28$ erg/cm^2 in both cases. It therefore appears improbable that (a) the increase in α with decreasing
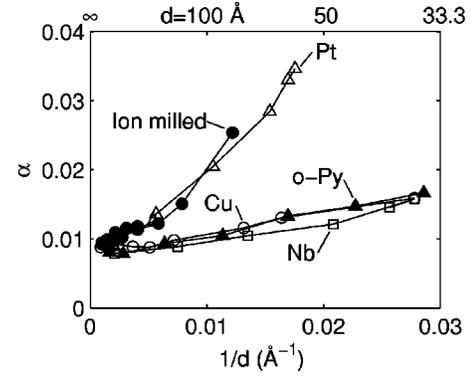
FIG. 4. Gilbert damping coefficient α, of Py adjoined with Cu, Nb, and Pt. As a reference we plot the *o*-Py series. Also shown is a Py film that was ion milled from one side.

film thickness is caused by variations in the surface anisotropy, and (b) the similarity with the changes in α and ρ is simply a coincidence. Also the theory predicts a field dependence of α, i.e., a decrease as the applied field is reduced since the two-magnon contribution vanishes in the limit of zero field.[13] We observe a constant α in the field range 150 down to 30 Oe, at which point α starts to *increase* due to incomplete saturation of the films.

The effect of adjoining Py layers with different NM metallic layers on the magnetization damping is shown in Fig. 4, with the *o*-Py series as a reference. For thick samples the damping parameter remains constant, equal to the bulk value. However, as the film thickness decreases α increases rapidly, the rate of increase depending on the type of interface. In the Cu, Nb, and *o*-Py series the damping at $d_{\mathrm{Py}} \sim 35$ Å is approximately twice the bulk value in those samples. It is apparent that the effect on α of adjoining the Py films with Cu or Nb is the same as protecting the Py with photoresist, i.e., the fact that these thin *metallic* films meet with Py is insignificant in these cases. In contrast a very pronounced effect was observed with Pt-coated surfaces. Assuming a linear dependence of α on $1/d$, the slope of the Pt-coated material is more than 4.5 times the slope for the other overlayers, and the α value at $d_{\mathrm{Py}} = 57$ Å is almost four times the bulk value. This effect was observed independently by Mizukami *et al.*[24] We also found that successive thinning of a Py-film by ion milling led to greatly enhanced damping. As can be seen in Fig. 4, the effect of ion milling one side of the sample is at least equivalent to that of having two Py/Pt interfaces.

The great increase in α in the ion-milled sample most likely arises from the increased electron scattering at the surface due to surface damage caused by the ion-milling process. Again we observe a constant α as a function of applied field, arguing against two-magnon scattering effects similar to those observed by LeCraw *et al.*[25] in single-crystal garnets. The Cu-coated samples had a surface roughness of 25 ± 3 Å$_{\mathrm{rms}}$. The Pt-coated samples were a factor of 3–4 smoother (7 ± 2 Å$_{\mathrm{rms}}$). Despite having much rougher interfaces than both the *o*-Py and the Pt-coated samples, the Cu-coated samples did not show an enhanced damping over the uncoated *o*-Py sample and much less damping than the Pt-

coated sample, as can be seen in Fig. 4. These observations confirm that the increased α in the Pt-coated series is not caused by surface roughness effects, and point toward either an interface scattering effect or to some intrinsic property of the capping layers, or both. Berger[26] predicted, for nonmagnetic layers in contact with a magnetic layer, a contribution to α from exchange coupling between localized magnetic spins and conduction electrons accompanied by spin-flip scattering both at interfaces *and* from interaction with phonons in the nonmagnetic layers through the spin-orbit interaction. The latter would imply that layers with strong spin-orbit interaction, such as Pt, would provide a more effective damping than, e.g., Cu, in qualitative agreement with our results. Recently, a paper by Tserkovnyak *et al.*[27] caught our attention. They presented a model of this same system based on the idea that the magnetization precession in the FM layer drives a spin current into the NM layer, where any spin imbalance is assumed to relax. Thus the same applies in their case, that Pt should provide a large enhancement due to its strong spin-orbit coupling and effective spin relaxation.

We did not study the resistivity of the trilayer films coated with Cu, Nb and Pt, as the situation is obviously much more complicated than in the single-layer case. The resistivity of the metallic capping layers is in all our cases lower than that of Py. Separating the resistive contributions of capping layers, Py, and interfaces is very difficult, making a comparison between α and ρ in this case less meaningful. Nonetheless, these results with adjoining nonmagnetic layers reinforce the above conclusion about the primacy of electron scattering mechanism in the viscous damping of very thin ferromagnetic films.

The effectiveness of ion milling in increasing α compared with the lack thereof in the Cu-coated samples suggests that, although the Cu-coated samples are rougher than the very smooth PR- and Pt-coated samples, the roughness is insignificant as far as magnetization damping is concerned. One may expect a much more complex surface after ion milling, caused by a mixture of redeposition of Py, surface oxidation and structural defects, none of which are present in the as-deposited films.

In conclusion we have confirmed that magnetization relaxation in ultrathin Py films is governed by processes that involve ordinary electron scattering. This is analogous to bulk relaxation except that the electron scattering in ultrathin films is increasingly caused by surfaces and defects as the films become thinner, whereas it is caused to a greater extent by phonons in bulk materials. We have also observed an increased damping in trilayers of Pt/Py/Pt which we attribute to strong spin-orbit coupling in the Pt layers. From a practical viewpoint our results highlight the important connection between electron scattering and magnetization damping. That should prove important when designing magnetic devices with a desired "optimal" dynamic response.

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