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Large Surface Effect on Magnetization Damping in Thin $Ni_{81}Fe_{19}$ Films Caused by Electron Scattering at Surfaces

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Abstract

There is a notable increase in magnetization damping in thin Permalloy ($Ni_{81}Fe_{19}$, "Py") films as their thickness is reduced, inversely proportional to their thickness. This indicates an effective magnetization relaxation mechanism at the film surface. The damping effect depends on the morphology of the surface and the type of protective layer used, and is much larger in structures of Pt/Py/Pt than in Cu/Py/Cu, Nb/Py/Nb or SiO_2/Py/PR, where PR is photoresist. There is a strong correlation between the magnetization damping and the electrical resistivity as a function of film thickness in the SiO_2/Py/PR samples. These findings suggest that electron scattering at the surface, modulated by the spin-orbit interaction, is the cause of the increased magnetization damping.

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It is extremely important to understand the mechanisms of magnetization damping in magnetic materials. Particularly since the overall damping from these mechanisms can change dramatically on a size scale on the order of present day magnetic thin films, as is shown in this Letter. This affects the performance of small magnetoelectronic devices, whose magnetic relaxation clearly can not be approximated by bulk values. It has been shown that future magnetoelectronic devices require *enhanced* damping to reduce magnetization oscillations after reversal of magnetization direction [1, 2].

Thickness dependence of ferromagnetic resonance (FMR) in thin ferromagnetic films has been studied before. Heinrich *et al.* studied ultrathin films of Fe capped with a layer of Au [3]. Platow *et al.* presented results from samples of Fe, Ni, Co and Gd all measured in situ [4]. Their films were covered with layers of either Cu or W. Azevedo *et al.* studied films of $Ni_{50}Fe_{50}$[5]. All these studies found an increase in FMR-linewidth as the film thickness was decreased. Stoecklein *et al.* and Speriosu *et al.* showed that exchange coupling of ferromagnetic films to antiferromagnetic films has a strong effect on magnetization damping [6, 7]. Large values of α (compared to thick films, or "bulk") have been measured by various experimental methods in exchange coupled layers and magnetoelectronic devices [2, 8].

In this Letter we present results of thickness dependence of the magnetization damping in thin Permalloy ($Ni_{81}Fe_{19}$, "Py") films over a wide range in thickness. Specifically, we studied the effect of interfaces on magnetization damping by sandwiching the films between thin layers of various transition metals. Our results prove that there is a large contribution from the interfaces to the magnetization damping, that depends heavily on the type and morphology of the interfaces. This contribution can overwhelm the bulk damping at film thickness up to 250 Å. Most importantly, we found that the magnetization damping and electrical resistivity in one of our sample series are strongly correlated. This provides direct evidence of a connection between magnetization damping and ordinary electron scattering. Our results suggest that electron scattering at the film surface, affected by the interfacial spin-orbit interaction, is the cause of the increased magnetization damping in these films.

The films were deposited by dc-magnetron sputtering in a vacuum system with a base pressure of 2×10^{-8} torr. During deposition they were exposed to a uniform magnetic field of ~ 150 Oe that induces uniaxial in-plane anisotropy. X-ray results show that all the Py films are (111) textured. We studied both the effect of varying the Py thickness and of sandwiching the Py layer between different materials by making structures of Si/SiO_2/X/Py/X, where X is an 80 Å thick layer of Cu, Nb or Pt. For comparison we made a series of samples of Si/SiO_2/Py/PR, where PR is photoresist used to protect the film from oxidation. 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 coefficients of our samples we measured their high-frequency susceptibility in an FMR-experiment with swept frequency and fixed dc magnetic field [9]. Typical results are shown in Fig. 1. Permalloy is magnetically soft ($H_c \leq 4$ Oe) and modest magnetic fields suffice to completely saturate its magnetic moment in the plane of the sample. In our FMR experiments the applied dc-fields are smaller than 100 Oe. At that field the uniform mode of magnetization precession resonates at a frequency below 3 GHz. The exchange stiffness in Py causes higher order spin wave modes to appear at much higher frequencies. Our films are much thinner than the skin depth at 3 GHz so the ac-field can be considered uniform throughout the films and a quasistatic approximation relates the internal and external fields. This assumption is supported by the Lorentzian lineshape 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 $\sim 10^{-4}$.

The equation of motion of the magnetization is the well known Landau-Lifshitz equation,

$$\frac{\mathrm{d}\boldsymbol{M}}{\mathrm{d}t} = -\gamma \boldsymbol{M}\times\boldsymbol{H}_{\text{eff}} - \frac{\alpha}{M}\boldsymbol{M}\times\frac{\mathrm{d}\boldsymbol{M}}{\mathrm{d}t} \quad , \tag{1}$$

where $\boldsymbol{M}$ is magnetization and $\gamma = g\,|e|\,/2mc$ is the gyromagnetic ratio, and α is the Gilbert damping coefficient. $\boldsymbol{H}_{\text{eff}}$ is the effective magnetic field seen by the magnetization, and is expressed in terms of the free energy as $\boldsymbol{H}_{\text{eff}} = -\nabla_{\boldsymbol{M}}\mathcal{F}$. Our data can be reproduced quite well using Eq. (1) in a linearized form. The free energy includes the Zeeman energy, a demagnetization term, a uniaxial in-plane anisotropy and a uniaxial out-of-plane (surface) 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 the average anisotropy of the upper and lower surfaces, and d is the film thickness. The high-frequency 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. The gyromagnetic ratio, γ, in our films is obtained by fitting the field dependence of the resonance frequency when the applied field and the magnetization coincide with the easy axis of the sample, $\phi = 0$, $\psi = \theta = \pi/2$. We determine the perpendicular anisotropy and the Gilbert damping parameter by fitting the complex susceptibility, with K_s and α as parameters. With this approach we arrive at a self-consistent result where K_s accounts for the shift in the resonance frequency as a function of film thickness, shown in Fig. 2, according to the following equation,

$$\left(\frac{\omega_r}{\gamma}\right)^2 = \left(H\sin(\phi+\psi) + 4\pi M - \frac{2K_u}{M}\cos^2\phi + \frac{4K_s}{dM}\right) \times \left(H\sin(\phi+\psi) - \frac{2K_u}{M}\cos(2\phi)\right) , \qquad (3)$$

and where the g-value is constant for any given series of samples as a function of thickness. The g-values differ between sample series though, with values ranging from 2.03 to 2.06.

The thickness dependence of the magnetization damping in our samples is shown in Fig. 3. For thick samples the damping parameter remains constant, equal to the bulk value. However, as the film thickness decreases α increases rapidly in all our samples, the rate of increase depending on the type and morphology of interfaces [10]. In the Cu-, Nb- and PR-coated samples the damping at $d_{\mathrm{Py}} \sim 35$ Å is approximately twice the bulk value. 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 4 times the bulk value. We also found that the surface roughness induced by thinning one sample by ion milling led to greatly enhanced damping. As can be seen in Fig. 3 the effect of ion milling one side of the sample is at least equal to that of having two Py/Pt interfaces.

In order to gain better understanding of the mechanism responsible for the increased damping we measured the resistivity of the PR-protected film as function of both temperature and film thickness. In Fig. 4 we plot the room temperature and the residual (4.2 K) resistivity, $\rho(295\text{ K})$ and ρ_{res} respectively, and the resistivity ratio $\rho(295\text{ K})/\rho_{\mathrm{res}}$, as functions of inverse film thickness. A Fuchs-Sondheimer-type analysis results in a bulk resistivity of $\rho_{\mathrm{b}} = 24\ \mu\Omega$cm and mean-free-path $\lambda = 96$ Å at room temperature, with the corresponding low temperature values $\rho_{\mathrm{b,res}} = 14\ \mu\Omega$cm and $\lambda_{\mathrm{res}} = 215$ Å [11]. From the resistivity ratio it is evident that the scattering associated with the film surfaces and impurities accounts for most of the resistivity as the films get thinner. The temperature dependent contribution (phonon and magnon scattering) decreases inverse proportional to the film thickness.

Viewing the magnetization damping as a function of the resistivity at room temperature, as in the lower panel in Fig. 4, reveals a strong correlation between these two quantities. Over the entire thickness range investigated, α doubles in value at the same time as the total room temperature resistivity doubles.

The 3d ferromagnetic metals and alloys typically used in thin-film magnetic devices are characterized by small Gilbert damping coefficients in the bulk, or $\alpha \leq 10^{-2}$ [3, 4, 12, 13]. Mechanisms of viscous damping in these are of at least two types: one considers decay of the given magnon mode to other magnons [14, 15], another considers one-electron scattering [16–18]. At room temperature or above, magnetization damping caused by the latter occurs mainly via modulation, by the spin-orbit interaction, of the matrix elements for phonon-and impurity-scattering of electrons. In this case it has been shown that the magnetization damping is directly proportional to the ordinary electron scattering rate, $1/\tau$, i.e. $\alpha \sim 1/\tau$ where the proportionality constant depends on properties of the Fermi surface and on the Lande g-factor [17, 18]. Numerical comparison with our data, assuming a spherical Fermi-surface and Drude-like conductivity, resulted in a slope that is one order of magnitude smaller than in the lower panel in Fig. 4.

Despite that shortcoming of our simple estimate the correlation in Fig. 4, between α and the total room temperature resistivity ($\alpha \sim \rho \sim 1/\tau$), leads us to conclude that the increased magnetization damping as our films grow thinner is caused by electron scattering at the interfaces, modulated by the spin-orbit interaction. This is further supported by the large effect with Pt-overlayers, in which case strong interfacial spin-orbit coupling is expected.

In summary we have found that there there is a strong surface effect on the magnetization damping in thin Py films. This effect is apparent in all our samples and is enhanced considerably when the samples are coated with Pt, which has strong spin-orbit coupling, and when the surface of the films is rough. We attribute this effect to electron scattering at the film surface, modulated by spin-orbit coupling. This work was supported by National Science Foundation grant no. DMR-0071770.

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FIGURE CAPTIONS

FIG. 1 Real and imaginary part of 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, 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.

FIG. 2 Upper: The dependence of the resonance frequency on the in-plane magnetization angle in a sample of SiO_2/682Py/PR, with $H_{\rm dc} = 90$ 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. Lower: 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 $K_s = 0.3$ erg/cm^2.

FIG. 3 Damping coefficient, α, obtained by fitting the susceptibility, plotted as a function of $1/d$ and $1/d^2$. In the thickest films the damping equals the bulk value, but increases rapidly as thickness is decreased.

FIG. 4 Upper: Room temperature and 4.2 K values of resistivity plotted vs. inverse film thickness. Also shown is the ratio of the room-temperature to the residual 4.2 K resistivity. Lower: Correlation between Gilbert damping, α, and room temperature resistivity.

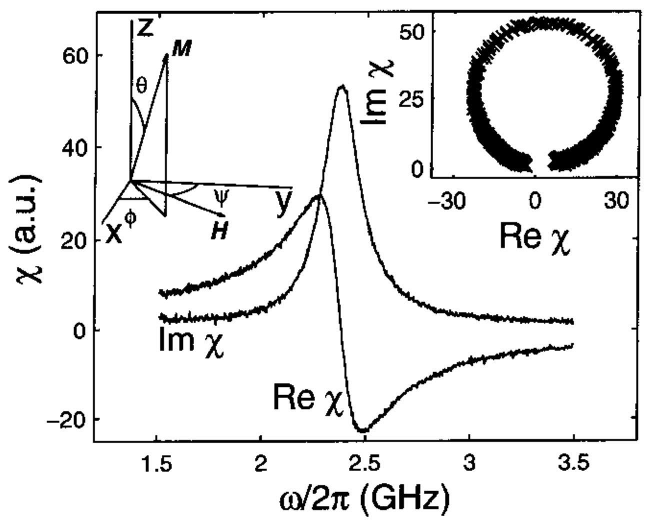

FIG. 1: Ingvarsson *et al.*, Phys. Rev. Lett.

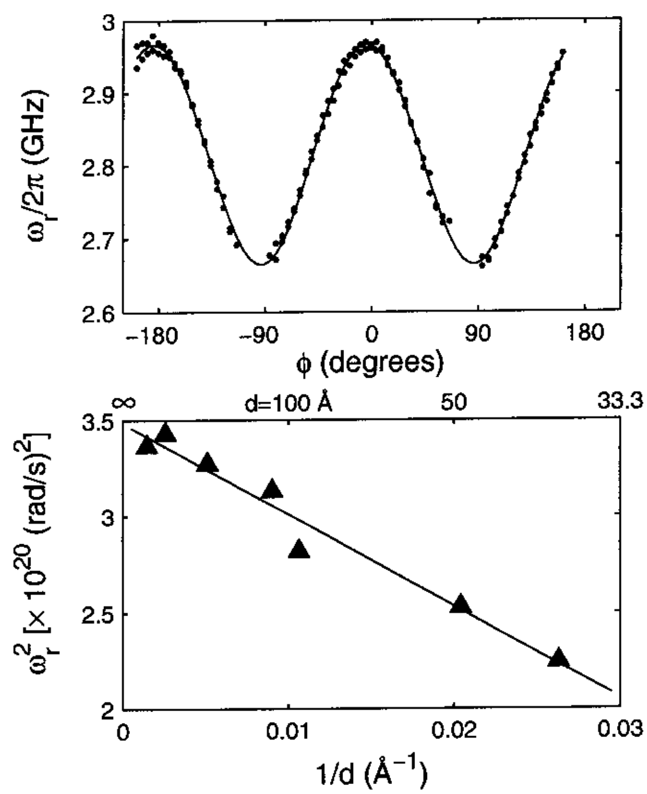

FIG. 2: Ingvarsson *et al.*, Phys. Rev. Lett.

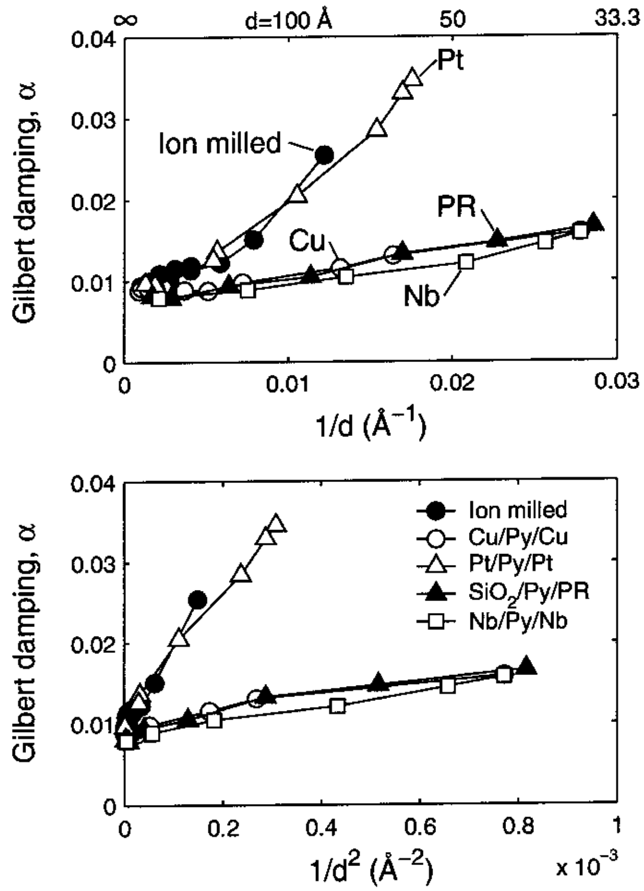

FIG. 3: Ingvarsson *et al.*, Phys. Rev. Lett.

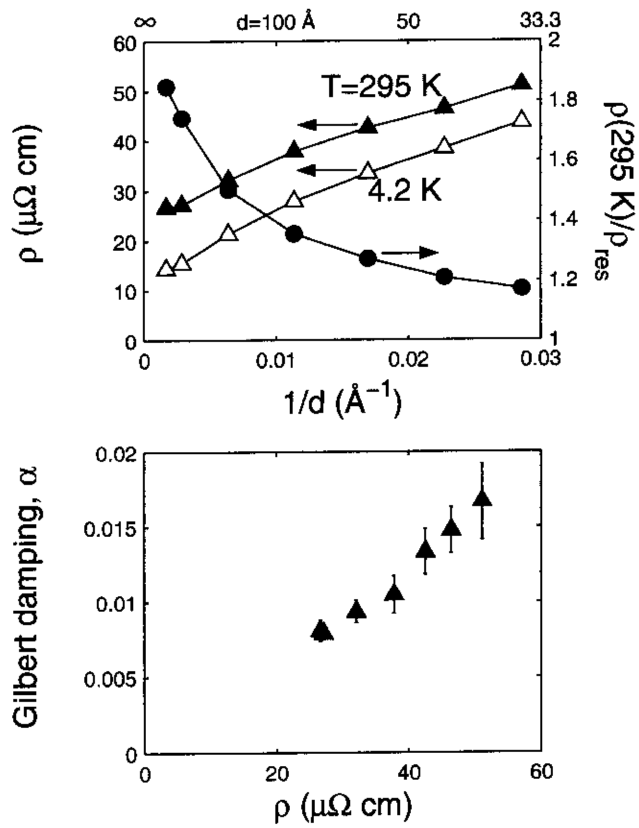

FIG. 4: Ingvarsson *et al.*, Phys. Rev. Lett.