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## Ferromagnetic relaxation in NiFe/Si(001) alloy thin films

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### ABSTRACT

In this paper, we present a study on the ferromagnetic relaxation in thin films. For this, a series of NiFe alloyed thin films were sputtered onto Si (001) wafers by dc magnetron sputtering, and then characterized by inplane ferromagnetic resonance (FMR). The FMR linewidth ( $\Delta H$ ) is studied as a function of the in-plane angle,  $\varphi_{H}$ , film thickness, *t*, and temperature, *T*. We show that the mechanisms responsible for the magnetization relaxation in NiFe thin films, involve angular dispersions of the uniaxial anisotropy,  $\Delta \varphi_{u}$ , and Gilbert damping, *G*. Both,  $\Delta \varphi_{u}$  and *G*, follow the 1/t law expected for interface phenomena. As function of temperature, the ferromagnetic linewidth decreases as *T* increases, in accordance with the theory of thermal activated electron-lattice scattering processes. © 2013 Trade Science Inc. - INDIA

#### **INTRODUCTION**

Ferromagnetic thin films and metallic multilayers have been the subject of intensive work during the last decades<sup>[1-3]</sup>. The fundamental magnetic, electronic and optical properties of these structures are quite different from their bulk counterparts, and over the last years, it has been shown that these properties are greatly influenced by the presence of the interfaces. On the other hand, the magnetic parameters of thin films are highly affected by growth conditions and sample treatment, purity of the alloys, substrate temperature, film thickness, roughness, etc. In general, the influence of the magnetic anisotropies on the physical properties of thin films has become one of the most important subjects in

### KEYWORDS

Magnetic thin films; Ferromagnetic linewidth; Ferromagnetic relaxation; Gilbert damping; Spin waves.

condensed matter physics, both from theoretical and experimental point of view. Experimentally, ferromagnetic resonance (FMR) has proven to be one of the ideal techniques to study magnetic anisotropies and magnetization dynamics in solid thin films<sup>[4]</sup>. This is because the resonance field and linewidth values are very sensitive to local fields, surface anisotropies, magnetic interactions between different elements at the interfaces, magnetic and structural inhomogeneities in the sample, and intrinsic properties of the material. Theoretically, phenomenological approaches and other microscopic models based on quantum-mechanical calculations have proven to be successful to confirm most experimental results on thin films and magnetic multilayered systems<sup>[5-8]</sup>. Nevertheless, the laws governing the relaxation mechanisms and

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the different types of magnetic interactions in metallic thin films still a challenge for theoretical and experimental physicists.

NiFe thin films and multilayers have been extensively studied for a long time by several experimental techniques, due to their good physical properties for applications in magnetic recording and microelectronic devices. Depending on the relative concentration of nickel and iron and on the film thickness, the magnetic properties of these materials can be controlled and improved. This makes NiFe alloys of importance in the design of thin film structures for hybrid magnetic device applications such as magnetic field sensors and magnetic random access memories (MRAM)<sup>[9,10]</sup>. For example, as the magnetic-datastorage industry approaches the Gbit/s transfer rate, the understanding of the magnetization dynamics of the soft magnetic elements used in recording heads becomes essential. In this sense, NiFe-based structures arise as the prototype for both experimental and theoretical research on this subject. Recently, some authors have devoted their attention in studying the ferromagnetic relaxation on NiFe thin films and the effect of temperature on the anisotropy fields and damping constants<sup>[11-13]</sup>. These works give important information on the damping mechanisms and explain the FMR relaxation in thin films from both microscopic and macroscopic point of view.

In this paper, we report on the FM relaxation in NiFe alloy films, grown onto Si (001) substrates by dc magnetron sputtering. The FMR linewidth is measured as function of the in-plane angle and film thickness, and is described by a phenomenological model that considers the line broadening as a combined effect of the intrinsic damping with the angular dispersion of the in-plane uniaxial axis. The temperature behavior of the FMR field and linewidth is also presented and discussed.

Our paper is organized as follows. In Sec. 2 we provide a brief outline of the sample preparation and describe the details of the FMR experiments. Sec. 3 is devoted to the phenomenological model used to interpret the FMR data, followed by their analysis in Sec. 4. Finally, the main results of this work are summarized in Sec. 5. NiFe films were grown by dc magnetron sputtering onto commercial electronic grade Si(001) wafers, with film thickness within the range of 63 Å to 147 Å. Magnetrons assure a continuous magnetic field of the order of 10 Oe during all the growth process. Before deposition, the substrates were cleaned in ultrasound baths of acetone and ethanol for 10 min, and then dried in nitrogen gas flow. The base pressure of the system prior deposition was  $2.0 \times 10^{-7}$  Torr. The films were deposited in a  $3.4 \times 10^{-3}$  Torr argon atmosphere in the sputter-up configuration, with the substrate at a distance of 9 cm from the target.

The FMR measurements were performed in a Xband VARIAN spectrometer, employing a home made cylindrical cavity with Q factor of the order of 2500. A homogenous dc magnetic field is applied in the plane of the samples in between the poles of an electromagnet and is modulated with a weak sinusoidal field provided by parallel Helmholtz coils operating at 100 Hz. The electromagnet is mounted onto a 0-360 degrees rotating base that allowed us to obtain the FMR spectra with respect to the direction of the applied field.

All spectra were taken at 9.35 GHz with the magnetic field applied in the plane of the sample, and in the temperature range 70 K < T < 300 K. The temperature was calibrated using a carbon-glass thermometer. All spectra correspond to Lorentzian line-shapes. The FMR linewidth was obtained by taking the peak-to-peak width of the experimental spectra resonance lines.

#### PHENOMENOLOGICAL MODEL

To describe the relaxation of the magnetization in a thin film let us first consider the coordinate system in Figure 1. In this frame of reference the total magnetic free energy appropriate for a non-oriented uniaxial film is given by,

### $E = -HM\sin\theta\cos(\varphi - \varphi_{\rm H}) + K_{\rm u}\sin^2\theta\sin^2(\varphi - \varphi_{\rm u}) +$ $2\pi M_{\rm eff}^2\cos^2\theta$ (1)

where  $\theta$  and  $\varphi$  are the polar and azimuthal angles of the magnetization vector, respectively; *M* is the bulk saturation magnetization; *H* is the applied magnetic field;  $K_u$  is the uniaxial anisotropy constant;  $\varphi_H$  and  $\varphi_u$  are the

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azimuthal angle of the applied field and the angle of the anisotropy axis with respect to the *x* direction, respectively;  $M_{eff}$  is an effective magnetization defined as  $4\pi M_{eff} = 4\pi M - 2K_n/M$ , where  $K_n$  is the out-of-plane uniaxial anisotropy constant.



Figure 1 : Coordinate system used to describe the orientation of *M* and *H* in the plane of the film.

For the FMR linewidth we used an expression that takes into account the contributions from the angular dispersions of the anisotropy axis and the intrinsic conduction mechanism<sup>[13]</sup>,

$$\Delta \mathbf{H} = \frac{2}{\sqrt{3}} \frac{\mathbf{G}}{\mathbf{M}^2 |\partial \omega / \partial \mathbf{H}|} \left[ \frac{\partial^2 \mathbf{E}}{\partial \theta^2} + \frac{1}{\sin^2 \theta} \frac{\partial^2 \mathbf{E}}{\partial \varphi^2} \right]_{\boldsymbol{\phi}_0, \boldsymbol{\theta}_0} + \left| \frac{\partial \mathbf{H}_{\mathbf{R}}}{\partial \varphi_{\mathbf{u}}} \right| \Delta \varphi_{\mathbf{u}} + \Delta \mathbf{H}(\mathbf{0})$$
(2)

where G is the so-called Gilbert parameter,  $H_u = 2K_u/M$ ,  $\theta_0$  and  $\varphi_0$  are the equilibrium positions of the magnetization,  $\omega(H)$  is the resonance frequency given by the resonance condition  $(\omega/\gamma)^2 = (1/M^2 \sin^2 \theta) [E_{\theta\theta} E_{\phi\phi} - (E_{\theta\phi})^2]_{\theta_0,\phi_0}$ ,  $\gamma/2\pi$  (=2.94 GHz/kOe) is the gyromagnetic ratio for an electron with g=2.08.  $|\partial \omega/\partial H|_0$  is the derivative of the resonance frequency with respect to the applied field calculated at the equilibrium position,  $H_R$ , is the resonance field for an uniaxial thin film magnetized in-plane expressed as  $H_R = [(\omega/\gamma)^2 (4\pi M_{eff})^{-1} - H_u \cos 2(\phi_0 - \phi_u)]$  [1/cos( $\phi_0 - \phi_H$ )]<sup>[14]</sup>. In our experimental condition the

out-of-plane equilibrium angle is  $\theta_0 = \pi/2$ . The first term represents the homogeneous broadening due to magnetic damping arising from spin-orbit and electron-lattice scattering. The second term is introduced to take into account the contribution from local inhomogeneities and microstructure defects, which are the main source of the anisotropy dispersion,  $\Delta \varphi_u$ . Here,  $\Delta H(0)$ is the frequency independent contribution to the linewidth related to extrinsic mechanisms such as two magnon scattering<sup>[14]</sup>. Calculating the corresponding derivatives in (2) with the condition  $4\pi M_{eff} >> H_u$ , the following approximate expression for  $\Delta H$  is obtained

$$\Delta \mathbf{H} \cong \frac{2}{\sqrt{3}} \frac{G}{\gamma^2 \mathbf{M}} \frac{\omega}{\left|\cos(\varphi_0 - \varphi_{\mathbf{H}})\right|} + 2\mathbf{H}_{\mathbf{u}} \frac{\sin 2(\varphi_0 - \varphi_{\mathbf{u}})}{\cos(\varphi_0 - \varphi_{\mathbf{H}})} \Delta \varphi_{\mathbf{u}} + \Delta \mathbf{H}(\mathbf{0})$$
(3)

Equation (3) together with the conditions  $\partial E/\partial \theta = \partial E/\partial \phi = 0$  is solved numerically to obtain the linewidth as function of the in-plane angle, given the in-plane equilibrium position of the magnetization for every value of the applied field. The theoretical values of  $\Delta H$  are then compared to the peak-to-peak FMR linewidth.

#### **RESULTS AND DISCUSSION**

Ferromagnetic resonance experiments were performed in the range of temperature from 70 K to 300 K. To obtain the linewidth,  $\Delta H_{pp}$ , the FMR spectra were first adjusted to Lorentzian lineshapes, and then the peak-to-peak linewidth was taken. As the temperature is lowered from room temperature, the signal decreases in intensity and broadens in all films.

The dependence of  $\Delta H_{pp}$  as a function of the inplane angle is shown in Figure 2 for a representative sample. It is seen that  $\Delta H$  exhibit the same symmetry as the resonance field (see right panel), with an increase in the amplitude and a pronounced shift when the temperature is lowered from 300 K to 70 K. The continuous curves are numerical fits of the experimental data to equation (2). The values of the uniaxial field used in these fits were obtained directly form the resonance field curves. For the saturation magnetization,  $4\pi M$ , we have used the value ~12.0 kG reported for NiFe thin films grown by magnetron sputtering<sup>[15]</sup>. The values of the Gilbert damping parameter, G, and the angular dis-

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persions,  $\Delta \varphi_u$ , are estimated and plotted in Figure 3. It is seen that the angular dispersions are *T*-independent, decreasing with thickness following approximately the 1/t law. The *G*-values obtained for these samples are of the same order of those reported in sputtered Py thin films<sup>[16]</sup>, also varying as 1/t within the range from 0.02 GHz to 0.06 GHz, at  $T \cong 70 \text{ K}$ . However, at  $T \cong 300 \text{ K}$ , *G* decreases with thickness to a constant value close to ~0.0020 GHz, in all samples. These results are indicative that at low temperatures, the magnetization relaxes



Figure 2 : Angular dependence of  $\Delta H$  at two different temperatures, for a tipical sample. For comparison, the in-plane symmetry of the resonance field,  $H_R$ , is shown in the right panel.

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Figure 3 : Thickness dependence of the angular dispersions and Gilbert damping parameter, at room temperature and liquid nitrogen temperature. The solid curves correspond to the 1/t law.

through a mixture of intrinsic conduction mechanisms and extrinsic relaxation, while the microstructure defects and other magnetic inhomogeneities dominate the magnetic relaxation at higher temperatures.

The temperature dependence of the FMR linewidth measured at  $\varphi_{H} = \pi/2$ , is shown in Figure 4. As temperature is lowered from room temperature  $\Delta H_{pp}$  is essentially constant, until certain critical temperature below which the signal broadens substantially. A similar behavior has been observed in Fe thin films<sup>[17]</sup> and was attributed to an increase of the magnetic anisotropies at low temperatures. This might be true in the general case, where the increase in linewidth observed at lower temperatures can be understood as the mixture of homogeneous broadening due to the intrinsic damping, and inhomogeneous broadening caused by the temperature-dependent anisotropies. However, according to Eq. (3), close to  $\varphi_{H} = \pi/2$ , the linewidth is homogeneous and is given by

$$\Delta H_{hom} \cong \frac{2}{\sqrt{3}} \frac{\omega}{\gamma^2} \frac{G(T)}{M}$$
(4)

To provide a complete picture of the homogeneous damping in NiFe thin films, the temperature behavior of the damping parameter must be known. A plausible explanation for this behavior is a thermally activated mechanism, such as electron-lattice scattering. Theoretically, this kind of process is characterized by the correlation  $G \sim \tau_c / (1 + \omega^2 \tau_c^{-2})^{[18]}$ , with  $\tau_c = \tau_0 exp(/K_B T)$ 

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being the correlation time for the thermally activated process,  $\tau_0$  is the characteristic relaxation time,  $\varepsilon$  is an energy barrier, and  $\omega$ =9.35 GHz is the resonance frequency. Within these considerations, the temperature dependence of the homogeneous part of the FMR linewidth can then be described by the phenomenological expression

$$\Delta H_{\text{hom}} \cong \frac{2}{\sqrt{3}} \left( \frac{\omega \tau_0}{\gamma^2 M} \right) \frac{A e^{\varepsilon/K_B T}}{\left[ 1 + (\omega \tau_0)^2 e^{2\varepsilon/K_B T} \right]} + \Delta H_{\infty}$$
(5)

where A is a phenomenological constant, and  $\Delta H$  the high-temperature linewidth. This equation is used as a fitting function to the experimental data. Reasonable agreement is obtained with for energy barrier,  $\varepsilon$ , from 0.29 eV to 0.38 eV as the film thickness decreases, and a characteristic relaxation time,  $\tau_0$ , of the order ~100 ps. This is interesting since this value is of the same order of the reversed time needed for magneto-optical applications. The values obtained for  $\varepsilon$  are in the energy range where spin-wave propagation process occurs in FM thin films and related compounds<sup>[19, 20]</sup> so the energy barrier could be associated to the energy gap of surface low-lying spin-waves.



Figure 4 : Temperature dependence of  $\Delta H$  measured with the magnetic field applied in the vicinity of  $\varphi_{H} = \pi/2$ . The solid curves are theoretical fits with the model described in the text.

#### SUMMARY

FMR technique was employed to study the relaxation of the magnetization in NiFe alloyed thin films. The experimental FMR linewidth displays uniaxial inplane symmetry consistent with a phenomenological framework in which, the magnetization relaxation is governed by processes that involve anisotropy dispersions induced by local inhomogeneities, and Gilbert damping. The values of G and  $\Delta \varphi_u$  followed the 1/t, typical of interface effects.

The temperature dependence of the FMR linewidth was also explored. We observed that as T increases up to room temperature, the homogeneous broadening,  $\Delta H_{hom}$ , decreases until a saturation value. This behavior is consistent with a relaxation mechanism due to electron-lattice scattering driven by thermal fluctuations. Although our analysis is phenomenological, this picture overlooks the connection between electron-lattice scattering and spin-wave activation as the origin of the magnetization relaxation in ferromagnetic thin films. From a practical point of view, our results should give an insight for understanding the dynamical response of the magnetization, required for the design of magnetic and hybrid devices at the nanoscale limit.

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