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Relaxation dynamics of ferromagnetic FePt thin films in a broad frequency range

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Abstract
The dynamical behaviour of FePt thin films in the chemically disordered fcc phase has been studied using ferromagnetic resonance spectroscopy in a broad range of frequencies, from 1 to 100 GHz. A set of samples with thicknesses between 9 and 94 nm was investigated at room temperature in order to analyse the damping of the magnetization as a function of the excitation frequency. As expected, the increase in frequency is accompanied by larger resonance fields and linewidths. These two quantities also display an anisotropic response when the external applied field is rotated from the in-plane to the out-of-plane direction.

We have observed that in the thicker samples the linewidth is different for fields applied parallel or perpendicular to the film plane which, in principle, should not occur in homogeneous ferromagnetic films. This behaviour could be understood by the presence of a frequency-independent anisotropic inhomogeneous broadening that depends on film thickness, in addition to the Gilbert damping term (α) which gives a linear frequency dependence. Although this last term is in general isotropic, we have measured different values of α for in-plane ($\alpha_{\parallel} = 0.025(1)$) and out-of-plane ($\alpha_{\perp} = 0.021(1)$) geometries. These values of the damping constant are considerably smaller than those previously reported in chemically ordered alloys, at variance with the prediction of recent developed theoretical models.

(Some figures may appear in colour only in the online journal)

1. Introduction

FePt thin-film alloys of equiatomic composition present unique magnetic properties, such as a high coercivity, a high magnetization and a very large magnetocrystalline anisotropy, which could be larger than 5×10^7 erg cm^{-3} in the $L1_0$ chemically ordered crystalline phase. This system has been extensively studied due to its possible technological applications [1–3]. However, FePt thin films fabricated by sputtering techniques at room temperature tend to grow in a soft magnetic phase (called A1), with the atoms randomly distributed in an fcc structure. Although these films do not have an anisotropy or a coercive field as large as those observed in the ordered phase, they present some remarkable features that make them an interesting system for studying their fundamentals properties. In particular, these films show an effective anisotropy perpendicular to the film plane, which is due to the combination of magnetocrystalline anisotropy (samples grow with a strong [1 1 1] texture, which is an easy magnetocrystalline axis) and magnetoelastic effects (as-deposited films have a positive magnetostriction coefficient that induces an easy axis when the sample is under compressive stress) [4–7].

Spin-wave dynamics in FePt alloys have also attracted considerable attention lately [8–10], and a correct estimation of the damping parameter α is of crucial importance for the application of this alloy to magnonic devices. Chemically disordered FePt thin films often show a critical thickness d_{cr} that defines the transition from planar to stripe-like magnetic domains. The value of this critical thickness depends on the quality factor, $Q = K_{\perp}/2\pi M_S^2$, defined as the ratio between the component of the anisotropy perpendicular to the film plane and the shape anisotropy. If the energy associated to the perpendicular anisotropy is less than the shape demagnetizing energy, that is $Q < 1$, it is then expected that the magnetization stays essentially in the film plane. However, if the film is

thicker than d_{cr}, the magnetization presents a relatively small perpendicular component that alternates in the up and down directions forming a particular magnetic configuration called stripe domains. For films with quality factors $Q \sim 0.3$, the critical thickness is in the range 20–40 nm [4, 6, 11].

There are different ways to characterize the magnetization dynamics in magnetic materials. The most frequently used is the classical Landau–Lifshitz–Gilbert equation of motion which takes into account the damping in the precession of the magnetization $\boldsymbol{M}$, in an effective field $\boldsymbol{H}_{\text{eff}}$ (that includes the contribution of the external and anisotropy fields),

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

where α is the damping parameter. A solution of the equation of motion for the uniform mode of precession can be obtained using the well-known Smit and Beljers [12] formalism, from which it is possible to deduce the dispersion relation, i.e. the functional dependence between the excitation frequency and the applied field in the case $\alpha = 0$. In the particular case of a thin film that has an in-plane anisotropy field of magnitude H_x and an effective perpendicular anisotropy normal to the film plane given by $H_z = 4\pi M - 2K_\perp/M$, it is possible to arrive to the following closed expressions for the dispersion relation:

$$\begin{aligned}\left(\frac{\omega}{\gamma}\right)^2 &= (H_\perp - H_x - H_z)(H_\perp - H_z)\\ \left(\frac{\omega}{\gamma}\right)^2 &= (H_{||\mathrm{h}} - H_x)(H_{||\mathrm{h}} + H_z)\\ \left(\frac{\omega}{\gamma}\right)^2 &= (H_{||\mathrm{e}} + H_x)(H_{||\mathrm{e}} + H_x + H_z).\end{aligned} \tag{2}$$

The above set of equations is only valid when the magnetization is aligned in the direction of the external applied field, i.e. full saturation is achieved. For a fixed excitation frequency ω the first expression gives the resonance field in the out-of-plane geometry ($H_\perp$), while the other two can be used to obtain the in-plane resonance field parallel to the easy axis ($H_{||\mathrm{e}}$) and parallel to the hard axis ($H_{||\mathrm{h}}$). Corrections in the resonance field for $\alpha \neq 0$ have not been considered due to the quadratic dependence of the Smit–Beljers formula [16] on α and the small values of the damping constant in this system. In figure 1 we have plotted the dispersion relation for the principal directions given by equations (2), and also the numerical simulations for a few intermediate angles. It can be seen that for frequencies of X band and higher ($\omega/\gamma > 3000$ Oe), resonance absorption should be observed for all orientations of the sample with respect to the external field. However, for L band ($\omega/\gamma \approx 400$ Oe), the excitation frequency is lower than the frequency gap, and the resonance condition is only fulfilled for a small angular range close to the in-plane hard axis, or to the film normal (see the inset of figure 1).

The second term in equation (1) accounts for the intrinsic relaxation that takes place in ferromagnetic resonance (FMR) experiments. Using the same Smit and Beljers formalism it is also possible to deduce a linear relationship of the peak-to-peak linewidth as a function of the microwave frequency, valid

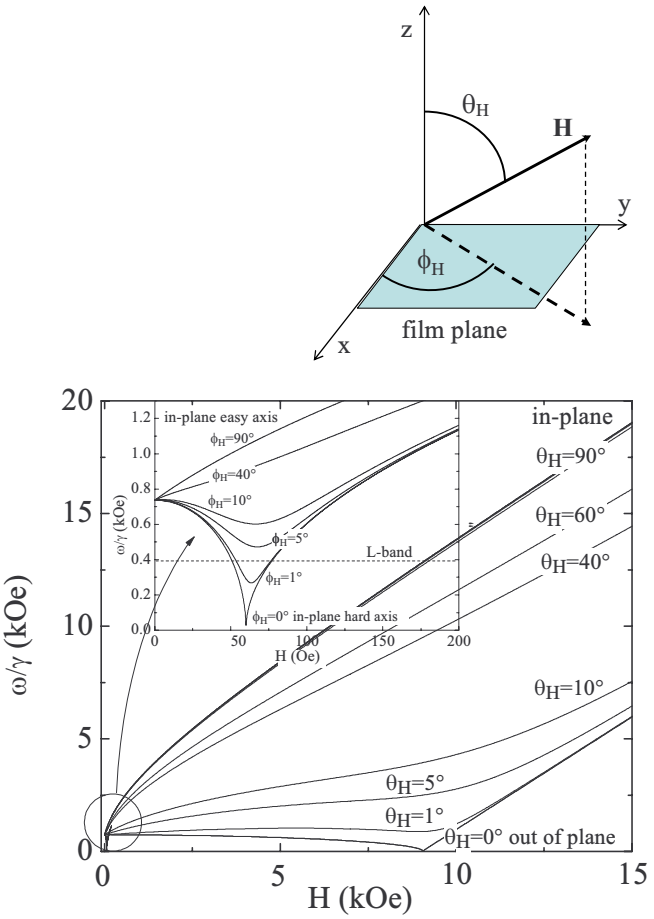

Figure 1. Dispersion relation for a homogeneous magnetic system with an in-plane easy axis of magnitude H_x and an easy plane anisotropy (or perpendicular hard axis) characterized by a field H_z, calculated for $\alpha = 0$. The above scheme shows the coordinate axes and the applied external field $\boldsymbol{H}$. Data are presented for different angles of the applied field in the range 0–$\pi/2$.
The crossing of these curves with a horizontal line gives the resonance field for a given excitation frequency. The resonance field is always minimum in the easy axis direction and maximum when $\boldsymbol{H}$ is parallel to the hard axis. For the simulations shown in the figure we have used the following parameters: $H_x = 60$ Oe, $H_z = 9000$ Oe, $g = 2.07$. The inset is a detail of the low frequency-low field region that shows the presence of an energy gap that prevents the observation of a resonance absorption for L band ($\omega/\gamma \approx 400$ Oe) when the sample is not aligned close to the hard axis.

when the magnetization vector is aligned with the external field [13–17],

$$\Delta H = \frac{2}{\sqrt{3}}\alpha\frac{\omega}{\gamma}. \tag{3}$$

In the above equation $\gamma = g\mu_{\mathrm{B}}/\hbar$, α is the Gilbert damping constant, which in general is assumed to be isotropic [18]. Using a narrow range of frequencies limited to X and K bands, we had previously reported [7] that the value of α was isotropic. However, there are several reports, both theoretical and experimental, that show that α can be anisotropic and should be considered as a tensor [19–21]. Equation (3) is often written in the form:

$$\Delta H = \Delta H_0 + \frac{2}{\sqrt{3}}\alpha\frac{\omega}{\gamma}, \tag{4}$$

where the term ΔH_0 is introduced to account for the presence of a frequency independent inhomogeneous broadening of the FMR line that describes the effect of extrinsic damping effects. This term can also be anisotropic, depending on the symmetry and distribution of the entities responsible for the line broadening.

The linear behaviour predicted by equation (4) is valid at least for in-plane and out-of-plane geometries in which the magnetization vector $\boldsymbol{M}$ is fully aligned with the external field $\boldsymbol{H}$. However, it is well known [14, 22, 23] that in many cases the linewidth deviates from a linear dependence with the excitation frequency. The two-magnon scattering mechanism is generally used to describe this non-linear extrinsic damping mechanism in, for example, ferrites and magnetic metallic films.

2. Experimental details

The films used in the FMR experiments are the same films that we have previously characterized in [6, 7]. The samples were grown on naturally oxidized Si (1 0 0) substrates at room temperature by dc magnetron sputtering from a FePt alloy target with a 50/50 atomic composition. The chamber was pumped down to a base pressure in the range of 10^{-7} Torr and the films were sputtered using 2 mTorr of Ar pressure, a power of 20 W and a target/substrate distance of 5 cm. With these conditions we obtained a sputtering rate of 0.15 nm s^{-1}. We have grown eight samples with thicknesses d = 9, 19, 28, 35, 42, 49, 56 and 94 nm.

As-made films were characterized using x-ray diffraction and EDX techniques. The photoemission spectra indicated that the Fe/Pt atomic ratio of the films was ∼45/55. The x-ray diffractograms showed that the samples tend to grow with a [1 1 1] texture normal to the film plane and that they were also subjected to an in-plane compressive stress. These two combined effects give rise to an effective magnetic anisotropy perpendicular to the film plane of magnitude $K_\perp = 1.5(4) \times 10^6$ erg cm^{-3}. FMR spectra have been acquired at room temperature using a commercial Bruker ESP300 spectrometer at frequencies ν = 1.17 GHz (L band), 9.4 GHz (X band), 24 GHz (K band) and 34 GHz (Q band). The W band (ν = 94 GHz) spectra were measured in a Bruker Elexsys E680 system. The samples were placed in the centre of the resonant cavity corresponding to each frequency, were the derivative of the absorbed power was measured using a standard field modulation and lock-in detection technique with amplitudes in the range 5–20 Oe. The sample could be rotated inside the resonator in order to measure the resonance field for different orientations. Due to the reduced sample space available in the high frequency resonators, we needed to use smaller pieces of the same sample for W band measurements.

3. Experimental results and discussion

Figure 2 illustrates the general line shape of the measured resonance spectra at different frequencies for the sample of 28 nm, with the magnetic field applied in the film plane.

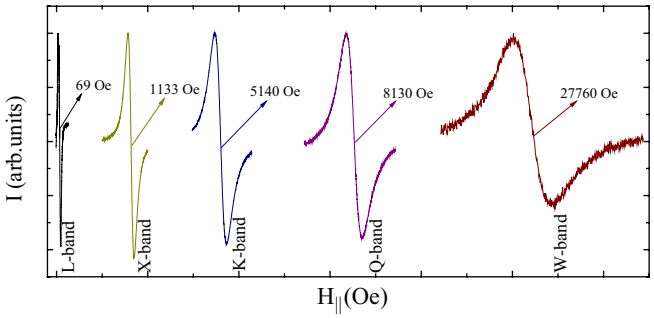

Figure 2. Typical FMR spectra for the FePt 28 nm sample measured at room temperature at the five different frequencies available. The external field was orientated parallel to the film plane and the resonance field is indicated for each frequency. The x-scale is the same for all spectra (distance between ticks is 1 kOe), but they have been horizontally shifted to fit better in a single window.

The value of the in-plane anisotropy field, $H_x \approx 60$ Oe, was estimated from in-plane angular measurements of the resonance field made at X band in the thinner films (d = 9, 19 and 28 nm). For this purpose we used the relationship $H_x \approx 2/3(H_{||h} - H_{||e})$, valid in the limit $H_x \ll \omega/\gamma$. Thicker samples ($d > d_{cr}$), in which stripe domains are generally observed, display rotatable anisotropy (the direction of the in-plane easy axis is determined by a previously applied external field of sufficient strength) and they present an essentially isotropic angular variation in FMR experiments due to this effect. The presence of an in-plane anisotropy should produce relatively large changes in the in-plane resonance absorption only at very low frequencies (L band) as can be inferred from the inset of figure 1. This is due to the particular shape of the dispersion relationship at low frequencies, which has a more complicated behaviour arising from the presence of a nonzero H_x that produces a frequency gap of magnitude $(\omega/\gamma)^2\big|_{(H=0)} = H_x(H_x + H_z)$. For the parameters used in figure 1 we can estimate $(\omega/\gamma)^2\big|_{(H=0)} \sim 750$ Oe, which is larger than the L band frequency in field units, $\omega/\gamma \approx 400$ Oe. A closer examination to the curves of figure 1 shows that when the frequency gap is larger than the microwave frequency a resonant absorption could be only observed close to a hard axis, in an angular range of a few tenths of a degree. For this reason a small misalignment is sufficient to prevent the observation of resonant absorptions at L band. The curves deduced from equations (2) should be also applied with care because they were obtained under the hypothesis of saturated magnetization, which does not necessary hold for the very low fields where the resonances are observed at L band. In our case it was impossible to find a resonance absorption at this frequency for most of the samples, especially for the thicker ones ($d > d_{cr}$). In a simple analysis this fact can be interpreted as a contradiction because these samples seem to be isotropic in the film plane. However, a possible explanation in this case is that the in-plane easy axes tend to be randomly distributed, instead of pointing in the same direction. A situation like this is equivalent to a random addition of the dispersion curves of figure 1, which is definitely not the same situation as having a film in which $H_x = 0$. As we will show later, the analysis of the linewidth is consistent with this interpretation.

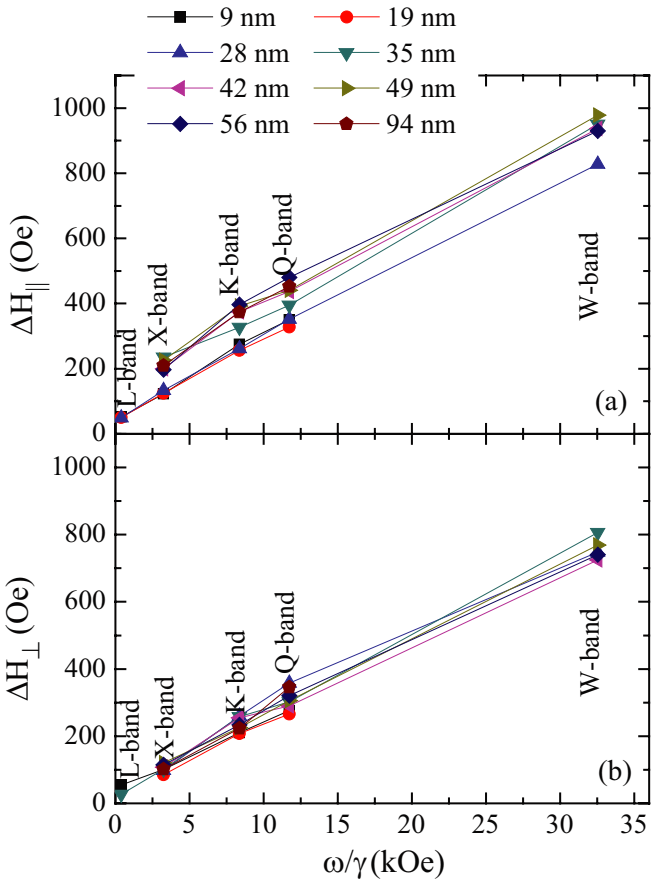

Figure 3. Linewidth as a function of the microwave frequency ω/γ (in units of magnetic field) for the different FePt samples, measured at frequencies from 1.2 to 94 GHz. Panel (*a*) shows the linewidth obtained with the external field applied parallel to the film plane, where two groups of data with the same slope but different ordinate can be distinguished. In panel (*b*) the perpendicular linewidths of all samples fall approximately on the same linear curve.

In addition to the increase of the resonance field for increasing frequencies that can be directly deduced using equations (2), it is experimentally observed in figure 2 that the linewidth is broader for larger frequencies, as predicted by equation (4). In figure 3 we show the linewidth of the different samples as a function of the microwave frequency for the in-plane (figure 3(*a*)) and the out-of-plane (figure 3(*b*)) geometries. We have found that in the two orientations the frequency dependence of the linewidth is almost linear. However, in the first case it can be observed that $\Delta H_{||}$ versus ω/γ has approximately the same slope but considerably different ordinates for the set of samples thinner and thicker than the critical thickness. When $\boldsymbol{H}$ is applied perpendicular to the film plane all the values of $\Delta H_{\perp}$ fall approximately on the same curve, although the slope is clearly smaller than that for the in-plane geometry. Linewidth data were fitted using the linear relationship of equation (4), obtaining the following values for the damping coefficient in the parallel and perpendicular configurations: $\alpha_{||} = 0.025(1)$ and $\alpha_{\perp} = 0.021(1)$. These values were obtained by averaging the slopes of ΔH versus ω/γ curves, which were found to be almost the same for the different thicknesses. This result confirms that, as expected, the intrinsic damping mechanism is independent of the film thickness. As we already mentioned, in a previous work limited to a relatively small frequency range we had reported that the value of α was isotropic.

From the new data it is evident that it is necessary to extend the frequency range in order to obtain reliable values of the damping constant. As far as we know, apart from our own data, the only reported values of the Gilbert damping constant in FePt films are for the ordered $L1_0$ phase. In the chemically ordered samples it is not possible to perform FMR experiments due to the very large magnetic anisotropy that prevents the observation of a resonant absorption [16], but an effective value of α was recently estimated from time-resolved magneto-optical Kerr effect experiments in partially ordered films [9, 10]. The reported upper limit for the effective damping constant was $\alpha \sim 0.055$ for perpendicularly magnetized films and $\alpha \sim 0.028$ for samples with in-plane magnetization. There is a recent theoretical work [24] that studied the influence of disorder in equiatomic FePt alloys, suggesting that the damping factor depends on the degree of atomic order, and tends to be larger in the case of completely disordered films. Our values are smaller than the smallest reported α values in (partially) ordered films, probably because in the cited references it was not possible to completely separate the influence of extrinsic damping effects.

As we have already shown in figure 3, it is quite clear that the values of $\alpha_{||}$ and $\alpha_{\perp}$ are different, which implies that our samples have an anisotropic damping parameter and it is necessary to consider the damping factor α as a tensor. For example, [21] discusses that the damping coefficient could be larger if the films are stressed. We know that our samples are subjected to an in-plane compressive stress which can then give rise to an increase in the damping coefficient $\alpha_{||}$ making this parameter anisotropic. Other works [19] ascribe the tensor nature of α to the coupling between the magnetic and elastic systems. In their approach, the Gilbert damping parameter is proportional to the magnetostriction coefficient λ, which in general is anisotropic, and could then give different values of α for different crystalline directions. Although our samples are not single crystals, it could happen that this effect is not averaging to zero because of the high [1 1 1] texture present in the films.

In order to check if equation (4) needs to be corrected by non-linear effects, such as two-magnon scattering, we have attempted to fit the experimental data using the same procedure as in [14]. As is well known, two-magnon scattering is maximal when the field is applied parallel to an easy direction (in this case an easy plane) and vanishes when the external field is parallel to the hard axis. Although there were some samples which appear to deviate from a linear law, the behaviour is almost linear in most cases, which is a strong indication of the very small contribution of this scattering mechanism. Unfortunately, no L band spectrum could be measured for the thicker samples in order to check if the linear behaviour of these films extends down to the lowest frequencies.

Figure 4 shows the values of the linewidth due to inhomogeneous broadening, ΔH_0, as a function of the film thickness, obtained from the ordinate of the linear fittings of figure 3, using equation (4). There is a noticeable difference of

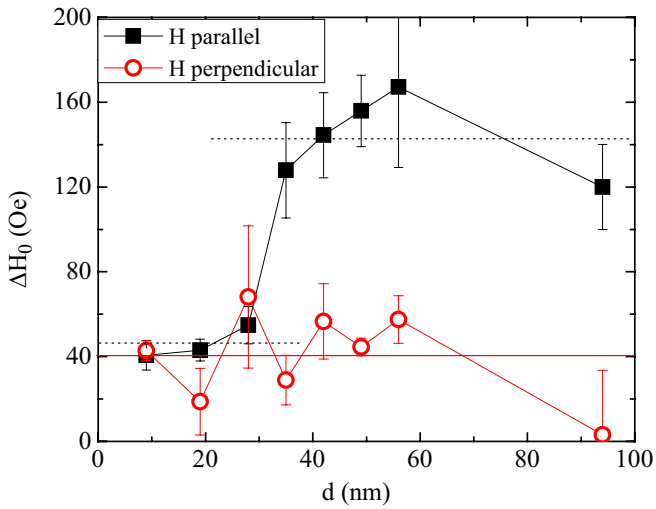

Figure 4. Linewidths due to inhomogeneous broadening obtained through the ordinate of the linear fittings of the data in figure 3. It is possible to observe a considerable increase in ΔH_0 for films thicker than d_{cr} when the external field is applied parallel to the film plane. The values obtained for the perpendicular case are approximately constant, within experimental error. The horizontal lines indicate the average values of ΔH_0 in the parallel (dashed lines) and perpendicular (continuous line) geometries.

this contribution to the linewidth between the thinner and the thicker samples when the external field is applied parallel to the film plane direction. As can be observed in figure 3(*a*), the two groups of samples follow approximately the same linear behaviour but with a considerably different ordinate. We have found the average value $\Delta H_{0||} = 46(7)$ Oe for $d < d_{cr}$ and $\Delta H_{0||} = 143(19)$ Oe for $d > d_{cr}$. When the field is applied in the perpendicular configuration we have found a single average value for this contribution, $\Delta H_{0\perp} = 40(22)$ Oe, which is of the same order as $\Delta H_{0||}$ for the thinner samples. The 94 nm sample should be considered with extra care because skin depth effects and the superposition of the uniform mode with standing spin-wave modes complicate considerably the determination of the linewidth. The explanation of the sudden increase in the inhomogeneous contribution to the linewidth for $d > d_{cr}$ seems to be related to the appearance of a structure of domains in the form of stripes in the thicker samples. We have reported in a previous paper [6] that the stripe domain structure in these films does not consist of a perfectly parallel structure of stripes, but it is more like a maze pattern with a preferential direction of alignment with many points where stripes join or bifurcate. As mentioned in [25], the pseudo uniaxial anisotropy with the easy axis aligned in the direction of the stripes originates from the presence of a small transverse effective field. If the orientation of this transverse field fluctuates in different microscopic regions of the film, it is then expected that the stripe structure loses the perfect parallelism, as we have observed in our films by using magnetic force microscopy. Following this line of reasoning, the in-plane resonance field would span in a range of values given approximately by the absolute value of this varying in-plane anisotropy field. In [6] this field was estimated to be around 400 Oe (with a dependence on film thickness) and the angular spread was around 16°. In a rough estimate this transverse field should increase the linewidth by approximately $\sin(16°) \times 400$ Oe ~ 110 Oe, which is pretty close to the value of the jump in $\Delta H_{0||}$ observed at the critical thickness.

4. Conclusions

Using a broad range of microwave excitation frequencies spreading over two decades, we were able to measure the intrinsic damping constant α and the inhomogeneous contribution ΔH_0 to the FMR linewidth in a set of disordered FePt alloy films. We have found α values considerable smaller than those reported in chemically ordered compounds; probably because in our experiment we could separate the extrinsic contributions from the intrinsic damping. Our measurements also demonstrate that the intrinsic damping is anisotropic, being minimum when the resonant absorption is measured with the field applied perpendicular to the film plane. This result was ascribed either to the presence of residual stresses or to the effect of the [1 1 1] texture of the films. We also observed a correlation between the magnetic domain structure and the inhomogeneous broadening of the linewidth: when stripe domains are present (for $d > d_{cr}$), $\Delta H_{0||}$ tends to be considerably larger than for films with planar domains. We have interpreted this result in terms of a fluctuating transverse anisotropy that arises in films which show a striped structure of magnetic domains.

Acknowledgments

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