Annealing effects on the microwave linewidth broadening of FeCuNbSiB ferromagnetic films
M. J. P. Alves, D. E. Gonzalez-Chavez, F. Bohn, and R. L. Sommer

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I. INTRODUCTION

Microwave magnetization dynamics in systems with reduced dimensions is a fundamental issue for current and emerging technological applications. In particular, the control of the damping in ferromagnetic thin films is crucial to the optimization of magnetic microwave and spintronic devices. In the last decades, large efforts have been devoted to the investigation of soft magnetic materials to be employed in such devices. For instance, this is the case of permalloy and CoFeB films, which have relatively low Gilbert damping parameter of $\alpha \approx 1.0 \times 10^{-3}$ (Ref. 5) and $\alpha \approx 4.0 \times 10^{-3}$ (Ref. 6), respectively. At the same time, YIG films also become a promising option with low Gilbert damping parameter, $\alpha \approx 3.0 \times 10^{-5}$, although they lack important magnetic properties, such as high saturation magnetization. In this sense, amorphous and nanocrystalline ferromagnetic alloys arise as attractive candidates for these applications, due to their soft magnetic features. In particular, amorphous FeCuNbSiB alloys, the precursor of the so-called Finemet nanocrystalline alloy, as well as the nanocrystalline phase itself present remarkable soft magnetic properties, evidenced by high magnetic permeability, high saturation magnetization, low magnetostriction constant, and low coercive fields, making possible their applications in high performance and low energy loss devices. These materials also exhibit low damping, but in addition to the Gilbert damping, one must consider the two-magnon scattering relaxation process due to the grain structure and ferromagnetic resonance (FMR) linewidth broadening due to effective field inhomogeneities. The contribution of these mechanisms can be determined by analyzing the FMR linewidth as function of frequency.

When the amorphous precursor of the Finemet nanocrystalline phase is deposited as a thin film, the stress induced by the coating process may degrade its soft magnetic properties. The influence of the thickness and stress stored with thickness on the dynamic magnetic properties of amorphous FeCuNbSiB films has already been investigated. At the same time, it has been previously observed that the best static magnetic properties are achieved with annealing temperatures below the onset of crystallization of the $\alpha$-FeSi nanogranular FeSiB, probably due to stress relief in the samples. In this paper, we report an experimental investigation of the annealing effects on microwave linewidth broadening of FeCuNbSiB ferromagnetic films. We systematically investigate the relationship between non-uniform residual stress and the microwave linewidth broadening in films with thickness of 100 nm submitted to different annealing temperatures below 400 °C. We correlate the non-uniform residual stress obtained from grazing incidence x-ray diffraction (GIXRD) measurements with the FMR linewidth due to effective field inhomogeneities measured from broadband ferromagnetic resonance absorption measurements, as well as we also estimate the Gilbert and two-magnon scattering contributions to the total ferromagnetic resonance FMR linewidth. We show that the effective field inhomogeneities constitute the main contribution to the microwave linewidth, while this contribution is related to the non-uniform residual stress in the films which is reduced by thermal annealing.

II. EXPERIMENT

For this study, we produced FeCuNbSiB ferromagnetic films with nominal composition of Fe$_{53.5}$Nb$_{5}$Cu$_{6}$Si$_{13}$B$_{9}$ and thickness of 100 nm. The films are deposited by magnetron sputtering onto glass substrates covered with a 2 nm thick Ta buffer layer. The deposition is carried with 5.2 mTorr pressure and 20 sccm Ar gas flow after a 1×10$^{-7}$ Torr base pressure is attained in the whole chamber. The Ta layer is deposited using a DC source with current of 50 mA, while the FeCuNbSiB layer is deposited using a 65 W RF power.
supply. During the deposition, the substrate moves (does not rotate) at constant speed through the plasma to improve the film uniformity. In order to induce an uniaxial magnetic anisotropy and define in-plane easy and hard magnetization axes, a constant magnetic field of 1 kOe is applied in the plane of the substrate during the film deposition. The easy axis is along the direction of the applied field. Square samples with dimensions of about 5 × 5 mm² were cut from the as-prepared films and annealed at temperatures, between 100°C and 400°C, for 60 min in a vacuum of 8 × 10⁻⁶ Torr.

The structural characterization is performed through GIXRD measurements, obtained using a Panalytical Xpert Pro diffractometer with Cu-Kα radiation, λCu-Kα = 1.54056Å. In this kind of measurement, the x-ray incidence angle with respect to the sample surface is fixed, allowing the control of the penetration depth and the reduction of the contribution from the substrate. It is well-known that the line broadening of the diffraction profile arises from instrumental apparatus, grain size, and non-uniform residual strain of the lattice parameter. The later is related to the non-uniform residual stress in the sample. This contribution is obtained from a single diffraction peak by applying the method discussed in detail in Ref. 20.

Quasi-static magnetic hysteresis loops along the easy and hard axes were acquired with a vibrating sample magnetometer, with maximum external magnetic field of ±300 Oe. The coercive field \( H_C \) is obtained directly from the measured loops, while the anisotropy field \( H_A \) is estimated following the procedures described in Ref. 21.

Finally, the dynamic magnetic behavior is investigated through broadband ferromagnetic resonance absorption measurements. We perform the experiment using a Rohde & Schwarz ZVA24 vector network analyzer, combined with a coplanar waveguide for frequencies in the range 1.0–8.0 GHz and static magnetic fields between ±800 Oe. In this case, FMR power absorption is obtained with in-plane DC magnetic applied along the easy axis of the sample. From the measurements, we extract the total FMR linewidth by fitting the absorption peaks. And from the analysis of the frequency dependence of the total FMR linewidth, we estimate the Gilbert damping, the contribution of the two-magnon scattering relaxation process and the effective field inhomogeneities contribution to the microwave linewidth broadening. Detailed information on the experimental setup and procedure used to obtain the absorbed microwave power is found in Ref. 5.

III. RESULTS AND DISCUSSIONS

In order to investigate the annealing effects on the microwave linewidth broadening of FeCuNbSiB ferromagnetic films, first of all, we characterize the structural and quasi-static magnetic properties of the samples.

Figure 1 presents the GIXRD patterns measured for FeCuNbSiB ferromagnetic films annealed at selected temperatures. The patterns for all samples present similar behavior. In particular, they exhibit a broad peak around \( 20 \approx 44.61° \), corresponding to the (110) diffraction peak of \( \alpha \)-Fe. This broad peak is due the low crystalline volume fraction of the \( \alpha \)-Fe grains with a large non-uniform strain. The considered range of annealing temperature is below the onset of the \( \alpha \)-FeSi nanograins crystallization and, for this reason, a diffraction peak associated with such \( \alpha \)-FeSi nanograins embedded in an amorphous ferromagnetic matrix of FeSiB is not observed.

Considering grazing incidence x-ray diffraction measurements, the non-uniform residual strain/stress can be obtained from a single diffraction peak. In this case, the experimental diffraction peak is fitted using a Voigt function, resulting in the separation of the Lorentzian \( \beta^L \) and Gaussian \( \beta^G \) contributions to the peak broadening, associated with the grain size \( \xi \) and non-uniform strain \( \epsilon \), respectively. In order to obtain the structural contributions to the broadening \( \beta^{L,G}_S \), the fitting parameters \( \beta^{L,G}_S \) are corrected by the instrumental broadening \( \beta^{L,G}_I \), obtained from the measurement of a LaB₆ standard powder sample, through the expression

\[
\beta^L_S = \beta^L_I - \beta^L_I' \\
\beta^G_S = \sqrt{(\beta^G_I)^2 - (\beta^L_I')^2}.
\]

(1)

Thus, the grain size \( \xi \) and non-uniform strain \( \epsilon \) are given by

\[
\xi = \frac{\lambda_{Cu-Kα}}{\beta^L_S \cos \theta},
\]

(2)

\[
\epsilon = \frac{\beta^G_S}{4 \tan \theta}.
\]

(3)

where \( \theta \) is the angle of the diffraction peak.

Figure 2 summarizes the structural parameters obtained from the GIXRD patterns following the aforementioned procedure. The as-prepared sample presents grain size \( \xi \) of 5.0 nm and non-uniform strain \( \epsilon \) of 5.6 × 10⁻². With the annealing, the grain size presents initially an increase up to 6.6 nm for \( T_A = 150°C \), followed by a decrease to 2.8 nm for \( T_A = 350°C \), and a further increase to 3.4 nm for \( T_A = 400°C \). The decrease in grain size with the increase in the annealing temperature is associated with an increase in the number of small \( \alpha \)-Fe grains, reducing the mean grain size.
size. Interestingly, the annealing at $T_A = 200^\circ C$ induces an additional non-uniform strain, owed to the difference between the thermal expansion coefficients of the film and the substrate. For higher temperatures, the thermal expansion has smaller influence, and we observe the relief of the residual stress. The smallest values of the grain size and non-uniform strain are found for $T_A = 350^\circ C$, this is the same annealing temperature for which the minimum coercive and anisotropy fields are observed, as we show below.

Figure 3 shows the quasi-static magnetization curves obtained with external magnetic field applied along the in-plane easy and hard axes. The magnetization curves corroborate the uniaxial in-plane magnetic anisotropy for all samples. Despite the similar shape of the magnetization curves irrespective on the annealing, an evolution of the coercive $H_C$ and anisotropy $H_A$ fields with the annealing temperature is observed. Figure 4 shows the dependence of $H_C$ and $H_A$ with the annealing temperature. In this case, the initial increase in the $H_C$ can be attributed to the formation of pinning centers for the domain walls due to the increase in the number of $\alpha$-Fe crystallization nuclei and stress centers in the bulk of the sample due to the difference between the thermal expansion coefficients of the film and the substrate. For higher temperatures, both $H_C$ and $H_A$ have a reduction with the annealing temperature, a fact related to the relief of the residual stress in the films. The smallest $H_C$ and $H_A$ values are verified for $T_A = 350^\circ C$, annealing temperature in which the minimum non-uniform stress is observed. Thus, our results directly relate the improvement of the soft magnetic properties with the relief of the non-uniform residual stress.

Figure 5 shows the broadband ferromagnetic resonance absorption spectra for the as-prepared sample. The color scale denotes the absorption amplitude from blue (minimum) to red (maximum). The amplitude maxima on the branches correspond to the resonant modes which define the dispersion relation ($f$ vs $H$). All samples present similar behavior.
with single resonant modes. The dispersion relations were well fitted using the Kittel equation for frequencies up to 6.5 GHz

$$f_r = \frac{\gamma}{2\pi} \sqrt{H_{\text{FMR}}(H_{\text{FMR}} + 4\pi M_{\text{eff}})}, \quad (4)$$

using $|\gamma|/2\pi = 2.8$ MHz/Oe to obtain the $4\pi M_{\text{eff}}$ listed in the Table II.

For each frequency, we extract the FMR absorption profile as a function of the external magnetic field, and obtain the experimental field linewidth $\Delta H_{\text{exp}}$ by fitting the FMR peak using a Voigt function.

The experimental ferromagnetic resonance linewidth for the studied films is a result of the contribution of three main processes: the intrinsic Gilbert damping $\Delta H_G$, the two-magnon scattering processes $\Delta H_{\text{TMS}}$, and a linewidth broadening due to effective field inhomogeneities $\Delta H_{\text{inh}}$. In our case, as the linewidth $\Delta H_{\text{inh}}$ is comparable to the linewidth $\Delta H_L = \Delta H_{\text{TMS}} + \Delta H_G$, the total linewidth is given by$^{1,2}$

$$\Delta H_{\text{Tot}} = \left(\Delta H_{\text{inh}}\right)^2 + 0.909\Delta H_{\text{inh}}\Delta H_L + 0.462(\Delta H_L)^2 \quad \Delta H_{\text{inh}} + 0.462\Delta H_L \quad (5).$$

From the Landau-Lifshitz-Gilbert equation, we find that the Gilbert linewidth $\Delta H_G$ is directly proportional to the FMR frequency $f_r$, and it can be expressed by

$$\Delta H_G = \frac{4\pi\alpha f}{|\gamma|}, \quad (6)$$

where $|\gamma|$ is the gyromagnetic factor and $\alpha$ is the Gilbert damping parameter.

The two-magnon scattering relaxation process is related to the short wavelength spatial fluctuations of the effective field.$^{24}$ This leads to a coupling between FMR uniform mode and spin wave modes with the same angular frequency $\omega$. To take it into account, we consider the grain-to-grain two-magnon scattering model.$^{2,24}$ This relaxation mechanism presents a nonlinear broadening of FMR linewidth $\Delta H_{\text{TMS}}$ as a function of frequency, given by$^2$

$$\Delta H_{\text{TMS}} = \frac{|\gamma|H_s^2}{P_A(\omega)} \int A_{\mathbf{k}} C_\mathbf{k}(\xi) \delta(\omega - \omega_\mathbf{k}) d^2k, \quad (7)$$

where $H_s$ is an anisotropy field which is different from the effective anisotropy field obtained from hysteresis curves,$^2$ $P_A(\omega) = \partial\delta f_{\text{FMR}}/\partial(|\gamma|H)$ relates frequency and field swept linewidths, $C_\mathbf{k}(\xi)$ is the Fourier transform of the grain-to-grain non-uniform effective field correlation function, $A_{\mathbf{k}}$ is the spin wave ellipticity function, and the delta function $\delta(\omega - \omega_\mathbf{k})$ is employed to select the spin wave mode with angular frequency $\omega_\mathbf{k}$ equals to the FMR uniform mode $\omega$. For the $\Delta H_{\text{TMS}}$ calculation, the integration in Eq. (7) must be performed over the in-plane spin wave vector space $\mathbf{k}$ and, here, this procedure is performed numerically using the full dispersion relation $\omega_\mathbf{k}$ for thin films reported in Ref. 24.

The last contribution to the ferromagnetic resonance linewidth, the linewidth broadening due to effective field inhomogeneities $\Delta H_{\text{inh}}$, is independent on the frequency.$^{23}$ This linewidth broadening is related to long-range inhomogeneities of the effective field, in contrast to $\Delta H_{\text{TMS}}$. The effective field inhomogeneities may result from non-uniform residual strain through magnetoelastic coupling, non-uniform distributions of grains, crystallographic defects, and non-uniform RF excitation fields. The contribution $\Delta H_{\text{inh}}$ arises from the superposition of a collection of resonance fields observed in different parts of the sample,$^{23}$ while the contribution from the non-uniform RF is constant and estimated as below 5 Oe for our setup and sample thickness. This value was obtained from measurements performed with high quality samples (not shown here).

Figure 6 shows the experimental $\Delta H_{\text{exp}}$ and calculated $\Delta H_{\text{Tot}}$ ferromagnetic resonance linewidth as function of the frequency measured for the FeCuNbSiB ferromagnetic films after annealing, together with the calculated contributions from the intrinsic Gilbert damping $\Delta H_G$, the two-magnon scattering $\Delta H_{\text{TMS}}$, and the effective field inhomogeneities $\Delta H_{\text{inh}}$. The quantitative agreement between calculated results and experiment is good for all samples. In particular, taking into account the aforementioned three contributions to the ferromagnetic resonance linewidth, we are able to reproduce the frequency dependence of the experimental linewidth up to 6.5 GHz. For higher frequencies, not addressed here, other contributions, e.g., grain-boundary two-magnon scattering$^{1,25}$ must be considered. The calculated results come from Eq. (5) combined with Eqs. (6) and (7). For the calculation of $\Delta H_{\text{TMS}}$, we used the grain size $\xi$ obtained from the RX analysis, $4\pi M_{\text{eff}}$ obtained from the dispersion relation fit of the experimental data, while the solid red line is the calculated curve. The purple, green, and blue solid lines represent, respectively, the calculated contributions of intrinsic Gilbert damping $\Delta H_G$, the two-magnon scattering $\Delta H_{\text{TMS}}$, and effective field inhomogeneities $\Delta H_{\text{inh}}$. 

![FIG. 6. Measured and calculated ferromagnetic resonance linewidth $\Delta H_{\text{tot}}$ for the annealed FeCuNbSiB ferromagnetic films. $\Delta H_{\text{tot}}$ was obtained by Eq. (5) due to large contribution of linewidth $\Delta H_{\text{inh}}$. The symbols represent the experimental data, while the solid red line is the calculated curve. The purple, green, and blue solid lines represent, respectively, the calculated contributions of intrinsic Gilbert damping $\Delta H_G$, the two-magnon scattering $\Delta H_{\text{TMS}}$, and effective field inhomogeneities $\Delta H_{\text{inh}}$.](image)
to Eq. (4), while the exchange constant $A$ and $H_a$ were chosen to reproduce the observed features in $\Delta H_{\text{Exp}}$. These parameters are summarized in Tables I and II.

As seen from Figure 6, $\Delta H_G$ is larger than $\Delta H_{\text{TMS}}$ for all samples in the studied frequency range. The Gilbert damping constant reaches the minimum of $\alpha = (3.2 \pm 0.3) \times 10^{-3}$ after annealing at $T_A = 250^\circ C$, which is probably related to a homogenization of the grain distribution which in turn reduces the structural disorder. It must be noticed that $\alpha$ in our samples is slightly smaller than in traditional materials as NiFe and CoFeB. From Figure 6, it can also be seen that the dominant contribution to $\Delta H_{\text{Tot}}$ in all samples is the linewidth due to effective field inhomogeneities $\Delta H_{\text{inh}}$.

Table II summarizes the numerical values of the physical parameters obtained from the analysis of the GIXRD patterns and from the analysis of the ferromagnetic resonance linewidth $\Delta H_{\text{Tot}}$.

Figure 7 shows the calculated contribution of the effective field inhomogeneities $\Delta H_{\text{inh}}$ to the ferromagnetic resonance linewidth for the FeCuNbSiB ferromagnetic films annealed at distinct temperatures. With the annealing procedure, $\Delta H_{\text{inh}}$ decreases from 38 Oe for the as prepared sample to 28 Oe at 350 $^\circ C$.

As previously discussed, the smallest values of the grain size and non-uniform strain are found for $T_A = 350^\circ C$, and this fact is reflected in the improved magnetic properties of this annealed film. At the same time, the smallest $\Delta H_{\text{inh}}$ values are also obtained around this range of temperature, revealing a direct correlation between the $\Delta H_{\text{inh}}$ values with the non-uniform strain/stress of the film. So, the relief of the non-uniform residual stress in the film reduces the linewidth due to effective field inhomogeneities.

### Table I. Parameters used in the calculation of $\Delta H_{\text{TMS}}$.

<table>
<thead>
<tr>
<th>$T_A$ ($^\circ C$)</th>
<th>$A$ ($\times 10^{-6}$ erg/cm$^2$)</th>
<th>$4\pi m_{\text{eff}}$ (kG)</th>
<th>$H_a$ (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>As prepared</td>
<td>0.29</td>
<td>8.35</td>
<td>29</td>
</tr>
<tr>
<td>100</td>
<td>0.34</td>
<td>7.71</td>
<td>46</td>
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<tr>
<td>150</td>
<td>0.35</td>
<td>7.91</td>
<td>33</td>
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<tr>
<td>200</td>
<td>0.34</td>
<td>8.64</td>
<td>38</td>
</tr>
<tr>
<td>250</td>
<td>0.27</td>
<td>8.58</td>
<td>33</td>
</tr>
<tr>
<td>350</td>
<td>0.35</td>
<td>8.79</td>
<td>54</td>
</tr>
<tr>
<td>400</td>
<td>0.34</td>
<td>8.48</td>
<td>48</td>
</tr>
<tr>
<td>Maximum error</td>
<td>$\ldots$</td>
<td>$\pm 0.05$</td>
<td>$\ldots$</td>
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</table>

### Table II. Summary of the obtained physical parameters.

<table>
<thead>
<tr>
<th>$T_A$ ($^\circ C$)</th>
<th>$\xi$ (nm)</th>
<th>$e$ $\times 10^{-2}$</th>
<th>$\Delta H_{\text{inh}}$ (Oe)</th>
<th>$H_{\text{eff}}$ $\times 10^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>As prepared</td>
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<td>5.6</td>
<td>38.6</td>
<td>3.5</td>
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<tr>
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</tr>
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</tr>
<tr>
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<td>32.6</td>
<td>5.0</td>
</tr>
<tr>
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<td>5.3</td>
<td>33.2</td>
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</tr>
<tr>
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<td>28.4</td>
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</tr>
<tr>
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<td>4.6</td>
<td>28.5</td>
<td>6.1</td>
</tr>
<tr>
<td>Maximum error</td>
<td>$\ldots$</td>
<td>$\pm 0.5$</td>
<td>$\pm 1.5$</td>
<td>$\pm 0.3$</td>
</tr>
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</table>

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