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# Frequency dependence of spin relaxation in periodic systems

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We show that in the presence of a periodic scattering potential the spin relaxation in ultrathin ferromagnets is not a monotonous function of the frequency, as has been usually assumed taking intrinsic Gilbert and extrinsic two-magnon processes into account. The spin relaxation rate is found to substantially increase at characteristic frequencies related to the periodicity of the magnon scattering potential. We propose a theoretical model which is experimentally confirmed in $Ni_{80}Fe_{20}$ thin films by artificially introducing different scattering periodicities. As a result, the current general approach for determining spin relaxation parameters in thin films has to be reconsidered.

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Both intrinsic and extrinsic spin relaxation processes are crucial for understanding the magnetization dynamics.[1–7] While the intrinsic ones, summarized as Gilbert damping, had been known and studied for decades,[8–10] extrinsic relaxation processes have been analyzed by experimentalists and theoreticians[1,11–14] in detail only more recently.

When classifying the extrinsic processes,[15] the two-magnon scattering deserves to be paid particular attention. In thin films—probably the most often engineered system in applied magnetism today—the two-magnon scattering among the extrinsic processes is the dominating relaxation mechanism due to inevitable defects in the material. The two-magnon scattering is usually anisotropic in contrast to the Gilbert damping, which is isotropic for the majority of 3*d* metallic systems.[2,16] Besides the two-magnon scattering's anisotropy and high intensity, its sensitive dependence on the symmetry of defects in the sample made it a matter of investigations into tailoring the spin relaxation.[1,4,5,17] Also from a fundamental point of view, one must note that pure Gilbert damping is rather a theoretical construct and mostly encountered in combination with extrinsic relaxation processes in a real material. Only in simple systems such as unstructured permalloy thin films is pure Gilbert damping a valid approximation. This fact makes the separation of the intrinsic and extrinsic relaxation processes in the majority of thin-film systems necessary.

In the experiment the presence of extrinsic relaxation is identified by the frequency dependence of the linewidth ($\Delta B$) of the resonant spin precession:[2,12,18] The Gilbert damping exhibits a linear frequency dependence [Eq. (1)], whereas the two-magnon scattering follows a curved arcsin-like [Eq. (2)] behavior (Fig. 1), which was modeled by Arias and Mills.[1] Here, $\alpha$ denotes the intrinsic damping parameter, $\gamma$ the spectroscopic splitting factor, $\omega/2\pi$ the precession frequency, and $B_\perp$ the effective perpendicular field. Although nonmonotonous linewidth behavior had been considered for disordered polycrystalline systems previously,[15] the monotonous behavior according to Eq. (2) has been used universally for years. Recently, possible deviations from the arcsin behavior of the linewidth came up for discussion,[19–21] which, to the best of our knowledge, up to now had not been further investigated due to the lack of an expanded theoretical model:

$$\Delta B_G \propto \frac{\alpha}{\gamma}\omega, \tag{1}$$

$$\Delta B_{2m} \propto \arcsin\sqrt{\frac{\sqrt{\omega^2+(\gamma B_\perp/2)^2}-\gamma B_\perp/2}{\sqrt{\omega^2+(\gamma B_\perp/2)^2}+\gamma B_\perp/2}}. \tag{2}$$

In this Rapid Communication we show that the frequency dependence of the linewidth may strongly differ from the standard arcsin expression and exhibits even periodic behavior. The two-magnon process depends on the properties of the scattering field determined by the defects. We calculate it for a system with a periodic uniaxial defect matrix, as can be found in nanopatterned or self-organized systems. We compare our theory with experimental results obtained on permalloy ($Py = Ni_{80}Fe_{20}$) thin films, in which periodic defect stripes have been patterned using ion implantation.

Ion implantation, in combination with lithographically defined masks, opens the possibility to create patterned hybrid magnetic materials.[22,23] This in turn also allows to influence and tailor the magnetization *dynamics*, i.e., the magnetic damping properties, at the nanoscale.[5,24,25] A periodic stripelike pattern of the ion-beam-modified Py will serve as the defect matrix introduced above. In a first step, $1 \times 1$ mm$^2$ square shaped 30-nm-thick Py films with a 3-nm Cr capping layer were grown by molecular beam epitaxy at a base pressure of $1 \times 10^{-10}$ mbar. Due to the oxidized $SiO_2/Si(001)$ substrate the films grow to be polycrystalline. Electron beam lithography was used to fabricate 1-mm long-stripes of width $s_0$ and spacing $s_1$ (i.e., with periodicity $l = s_0 + s_1$) into a 100-nm-thick polymethyl methacrylate (PMMA) resist covering the whole sample area (see Fig. 2). After resist development the samples were implanted with $Cr^+$ ions with an energy of 5 keV and a fluence of $5 \times 10^{15}$ ions/cm$^2$. The $Cr^+$ ions either get absorbed in the PMMA resist or penetrate the Cr capping layer in between the PMMA stripes, thus reaching the Py layer as depicted in Fig. 2. TRIDYN[26] simulations of the depth profiles of the atomic concentrations after $Cr^+$ implantation (not shown) reveal that a significant Cr concentration is

 

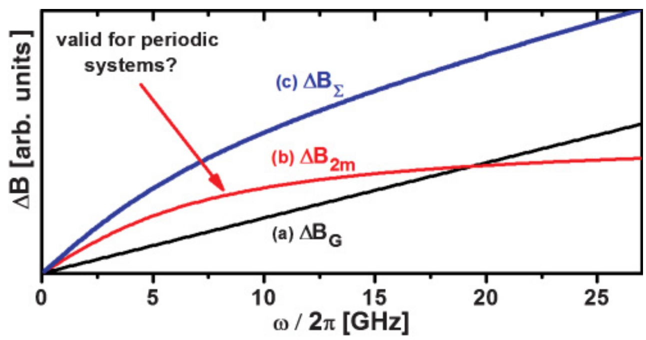


FIG. 1. (Color online) The linewidth as a function of the frequency. (a) The Gilbert contribution $\Delta B_G$ is linear. (b) The two-magnon contribution $\Delta B_{2m}$ exhibits the arcsin behavior. (c) The resulting linewidth $\Delta B_\Sigma$ is a sum of both contributions.

found in the topmost 7 nm of the sample. The concentration decreases with sample depth, starting with 50 at. % Cr at the surface. Note that the ion beam also sputters the capping layer, removing up to 1.5 nm, which is less than the thickness of the capping layer. The $Cr^+$ dopants cause local variations of the saturation magnetization,[25,27] whereas in Py no anisotropy is induced thereby. Therefore, we obtain samples in which the magnetization varies laterally, causing dipolar fields between the stripelike defects.

Magnetic anisotropy and relaxation of such samples with different periodicities were studied by ferromagnetic resonance (FMR). Using a cylindrical microwave cavity at 9.8 GHz, in-plane angular-dependent FMR measurements have been performed. At room temperature, samples with periodic defect structures, as well as unmodified samples of $Ni_{80}Fe_{20}$ thin films, reveal a very small in-plane uniaxial anisotropy field $K_2/M < 0.02$ mT and an effective perpendicular field $B_\perp = \mu_0 M_{\mathrm{eff}} = 0.81(1)$ T. The resonance fields of the samples with stripelike modifications show, in addition, lateral confinement effects for external field orientations close to the stripes' normal, which were reported in detail in Ref. 28. The in-plane angular dependence of the linewidth of these samples exhibits uniaxial behavior with maxima for field directions perpendicular to the stripes and minima for directions parallel to the stripes. This uniaxial behavior is

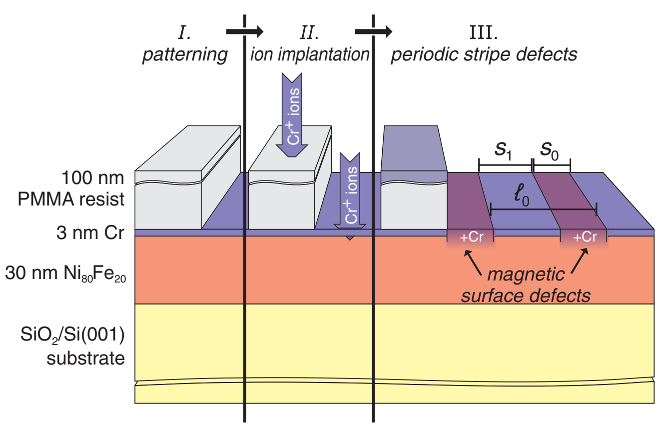


FIG. 2. (Color online) Sketch of the sample before and after ion implantation with $Cr^+$. Ions are stopped either in the resist or in the $Cr/Ni_{80}Fe_{20}$ interface region, resulting in periodic stripe defects.

explained by the two-magnon scattering induced by dipolar interactions between the stripelike defects in the sample, as described in Refs. 17 and 29. Samples with different stripe periodicities measured at 9.8 GHz showed variations of the two-magnon scattering strength perpendicular to the stripes. This observation motivated the study of the frequency dependence of the linewidth. A shorted coaxial microwave cable with a special end design was used to detect broad-band FMR.[30] In-plane measurements in the *quasicontinuous* frequency range of 2–26 GHz were performed for directions parallel and perpendicular to the stripes.

In Fig. 3(a) the experimental frequency dependence of the linewidth for the magnetic field applied parallel to the stripes is shown. The general behavior of the linewidth frequency dependence is not affected by the direction of the external magnetic field in one single sample. The convex curvature is related to the high modulation field used to increase the signal-to-noise ratio. Additional measurements (not shown here) show no isotropic two-magnon relaxation channel due to, e.g., grain-grain effects[31] in these polycrystalline samples. With the results shown in Fig. 3 we conduct comparative studies:

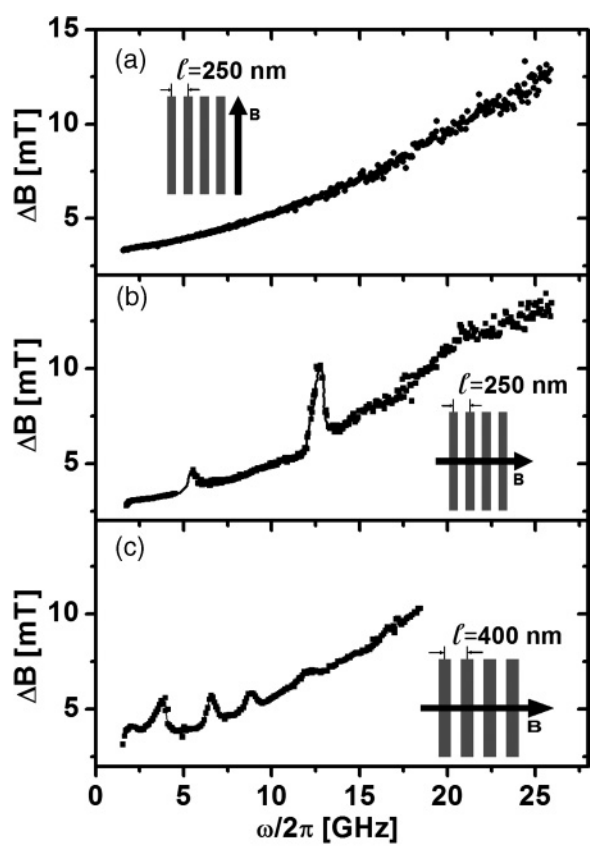


FIG. 3. Frequency dependence of the FMR linewidth of two samples structured with stripelike defects. While the general behavior is the same within one single sample for $B$ parallel to the stripes (a) and $B$ perpendicular to the stripes (b), for the latter configuration additional peaks due to the two-magnon scattering occur. Increasing the defect's periodicity $l$ from 250 to 400 nm in the second sample changes the position of the peaks and therefore the frequency dependence of the overall spin relaxation. Error bars are $\leqslant 15\%$. In (c) the frequency range is limited due to technical reasons. Solid lines are guides for the eye.

When the external magnetic field is applied perpendicular to the stripes, the behavior of the linewidth frequency dependence becomes nonmonotonous and is related to the two-magnon scattering process, which is known to be activated in such a configuration.[17,29] One finds a large peak at ∼12.7 GHz and two smaller ones at ∼5.4 and ∼21.2 GHz. These appear only for the direction perpendicular to the stripes. Yet the arcsin-like frequency dependence according to the Arias and Mills model[1] does not exhibit a nonmonotonous or even periodic behavior observed in experiment. In order to solve this problem, the mechanism of the two-magnon scattering is reconsidered in the following.

The dispersion relation of magnons parallel to the magnetization in a thin film is given by[1]

$$\omega = \gamma\left[\left(B+\mu_0 M_s\left(1-\frac{1-e^{-kd}}{kd}\right)\sin^2\phi_k+Dk^2+B_{\mathrm{MAE1}}\right)\right.$$
$$\left.\times\left(B+B_\perp-\mu_0 M_s\left(1-\frac{1-e^{-kd}}{kd}\right)+Dk^2+B_{\mathrm{MAE2}}\right)\right]^{1/2}. \tag{3}$$

$\omega$ is a function not only of the wave vector $k$, but also of the external magnetic field $B$ and sample parameters such as saturation magnetization $M_s$, effective perpendicular field $B_\perp$, spin-wave stiffness $D$, film thickness $d$, the spectroscopic splitting factor $\gamma = \mu_B g/\hbar$, and parameters $B_{\mathrm{MAE1}}$ and $B_{\mathrm{MAE2}}$, which are functions of very small anisotropy fields. Here, $\phi_k$ describes the critical angle between the magnetization and the wave vector for which the scattering can occur. As explained in detail in Ref. 1, $\phi_k$ is very small, so that $\sin\phi_k \approx 0$. In a two-magnon process a uniform magnon with $k = 0$ is scattered into a nonuniform state with the same energy and different wave vector $k_S \neq 0$, as shown in Fig. 4. Due to the local nature of this scattering process the momentum conservation can be violated.[15] In order to find the wave vector of such a final-state magnon one needs to solve the following equation:

$$\omega(k=0) = \omega(k_S). \tag{4}$$

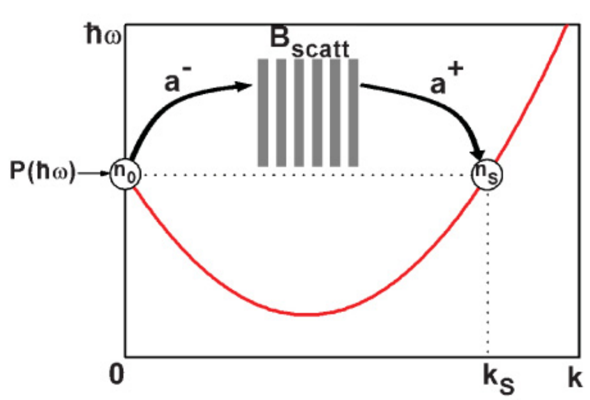


FIG. 4. (Color online) Dispersion relation of magnons in a thin film according to Eq. (3), which includes dipolar interaction, causing a minimum. In a two-magnon scattering process activated by the scattering field $B_{\mathrm{scatt}}$, a uniform magnon $n_0 = |\hbar\omega, k=0\rangle$, excited by a microwave field $P$, is annihilated ($a^-$) and a final state magnon $n_S = |\hbar\omega, k_S \neq 0\rangle$ is created ($a^+$).

By doing so, one finds that the frequency is eliminated as independent variable. However, the value of $k_S$ still depends on the external field as it is linked to the frequency by the resonance condition of the uniform precession given by Eq. (3) for $k = 0$. Thus, the wave vector of the final-state magnons $k_S = k_S(\omega)$ is a monotonously increasing function of the frequency and can be calculated using the values of the effective perpendicular field, anisotropy fields, and $g$ factor ($g = 2.11$) from the experiment and magnetization $M_S = 1.11 \times 10^6$ A/m and exchange stiffness $D = 1.847 \times 10^{-17}$ J/A of permalloy.[32] The scattering process itself is enabled by the scattering field, which couples the uniform with the final-state magnons. The coupling strength and consequently the FMR linewidth scale with the square of the Fourier transform of the scattering field for $k = k_S$,[33,34] being thus a function of frequency,

$$\Delta B_{2m}(\omega) \propto |\mathcal{F}\{B_{\mathrm{scatt}}\}[k = k_S(\omega)]|^2. \tag{5}$$

Although the exact functional form of the material's scattering field is not known, it is also periodic. Due to the periodicity $l$ of the defects, the Fourier transform of the scattering field can be assumed to have maxima for $k$ values being a multiple of the periodicity $2\pi/l$ of the stripelike defects in the reciprocal space (see Fig. 2). This requirement can be accounted for phenomenologically by multiple Gauss profiles at these values according to

$$|\mathcal{F}\{B_{\mathrm{scatt}}\}(k)|^2 \propto \sum_{n\in\mathbb{N}} \exp\left(-\frac{\left(\frac{n2\pi}{l}-k\right)^2}{2\sigma^2}\right). \tag{6}$$

The frequency dependence of the linewidth according to Eqs. (5) and (6) has been calculated numerically for different periodicities of the scattering field and is shown in Fig. 5. For

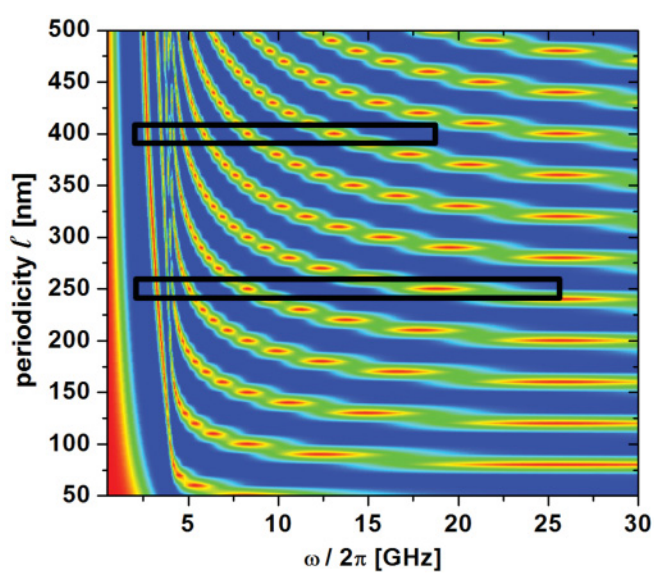


FIG. 5. (Color online) Color-coded normalized linewidth (scattering strength) according to Eqs. (5) and (6). The sections at $l = 250$ and 400 nm correspond to the frequency dependences of the linewidth shown in Figs. 3(b) and 3(c). The red (light gray) color means a larger linewidth than blue (dark gray). Elliptical structures appear due to limited numerical accuracy.

the experimental periodicities $l = 250$ and 400 nm (Fig. 3) the frequency dependence is represented by the horizontal line profile. A quantitative agreement depends very sensitively on the knowledge of the static magnetic parameters (e.g., local magnetization). The calculation confirms the experimental observation qualitatively very well. From Fig. 5 one directly recognizes which periodicity of defect structures must be patterned to enhance the spin relaxation rate in a ferromagnetic film. Based on the intrinsic relaxation of the material, additional enhancements by up to factor of 2 can be induced at chosen frequencies. The width of the peaks in Fig. 3(c) is determined not only by the angle between the line profile and the "structures" in Fig. 5, but also by the parameter $\sigma$ in Eq. (6), representing the $k$-value selectivity of the scattering field. The exact form of the material's scattering field would need to be known to calculate $\sigma$ as well as the absolute scattering rate. Our theory is based on the general scattering field approach[15,33,34] and could benefit from the Arias and Mills theory,[1] if it could be extended to a larger class of periodic defect structures.[35] Since the latter, which is beyond the scope of the present Rapid Communication, allows a transition to magnonic crystals, magnonic band gaps and the anomalous damping of the final-state magnons should also be taken into consideration.

In conclusion, we present a phenomenologic theory of the two-magnon scattering in periodically structured thin-film systems, which is confirmed by the experiment very well. Our results demonstrate that the frequency dependence of the overall spin relaxation in a large class of ferromagnetic systems is nonmonotonous and depends on the defect structure. Consequently, the usual practice of separation of intrinsic and extrinsic spin relaxation processes by means of their frequency dependence must be reconsidered. The exact structure of defects in a material, being periodic in the majority of ferromagnetic systems, needs to be ascertained first. Our findings are important for future developments, since they could explain the anomalous spin relaxation in magnonic crystals and help to tailor spin relaxation in spintronic devices by artificially inducing a defect structure to activate a desired spin relaxation channel in a specific frequency range.

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[1] R. Arias and D. L. Mills, Phys. Rev. B **60**, 7395 (1999).
[2] K. Zakeri, J. Lindner, I. Barsukov, R. Meckenstock, M. Farle, U. von Hörsten, H. Wende, W. Keune, J. Rocker, and S. S. Kalarickal *et al.*, Phys. Rev. B **76**, 104416 (2007).
[3] J. Lindner, K. Lenz, E. Kosubek, K. Baberschke, D. Spoddig, R. Meckenstock, J. Pelzl, Z. Frait, and D. L. Mills, J. Magn. Magn. Mater. **272-276**, E1653 (2004).
[4] G. Woltersdorf and B. Heinrich, Phys. Rev. B **69**, 184417 (2004).
[5] R. D. McMichael, D. J. Twisselmann, J. E. Bonevich, A. P. Chen, and W. F. Egelhoff Jr., J. Appl. Phys. **91**, 8647 (2002).
[6] M. C. Hickey and J. S. Moodera, Phys. Rev. Lett. **102**, 137601 (2009).
[7] V. Kamberský, Phys. Rev. B **76**, 134416 (2007).
[8] T. L. Gilbert, IEEE Trans. Magn. **40**, 3443 (2004).
[9] V. Kamberský, Czech. J. Phys. B **26**, 1366 (1976).
[10] C. Scheck, L. Cheng, I. Barsukov, Z. Frait, and W. E. Bailey, Phys. Rev. Lett. **98**, 117601 (2007).
[11] P. Krivosik, N. Mo, S. Kalarickal, and C. E. Patton, J. Appl. Phys. **101**, 083901 (2007).
[12] S. S. Kalarickal, P. Krivosik, M. Wu, C. E. Patton, M. L. Schneider, P. Kabos, T. J. Silva, and J. P. Nibarger, J. Appl. Phys. **99**, 093909 (2006).
[13] M. C. Weber, H. Nembach, B. Hillebrands, M. J. Carey, and J. Fassbender, J. Appl. Phys. **99**, 08J308 (2006).
[14] V. E. Demidov, O. Dzyapko, S. O. Demokritov, G. A. Melkov, and A. N. Slavin, Phys. Rev. Lett. **99**, 037205 (2007).
[15] C. W. Haas and H. B. Callen, *Magnetism* (Academic, New York, 1963), Vol. I.
[16] J. Seib, D. Steiauf, and M. Fähnle, Phys. Rev. B **79**, 092418 (2009).
[17] I. Barsukov, R. Meckenstock, J. Lindner, M. Möller, C. Hassel, O. Posth, M. Farle, and H. Wende, IEEE Trans. Magn. **46**, 2252 (2010).
[18] J. Lindner and K. Baberschke, J. Phys.: Condens. Matter **15**, S465 (2002).
[19] K. Zakeri, J. Lindner, I. Barsukov, R. Meckenstock, M. Farle, U. von Hörsten, H. Wende, W. Keune, J. Rocker, and S. S. Kalarickal *et al.*, Phys. Rev. B **80**, 059901(E) (2009).
[20] J. Lindner, I. Barsukov, C. Raeder, C. Hassel, O. Posth, R. Meckenstock, P. Landeros, and D. L. Mills, Phys. Rev. B **80**, 224421 (2009).
[21] K. Baberschke (private communication).
[22] L. Folks, R. E. Fontana, B. A. Gurney, J. R. Childress, S. Maat, J. A. Katine, J. E. E. Baglin, and A. J. Kellock, J. Phys. D **36**, 2601 (2003).
[23] J. Fassbender and J. McCord, J. Magn. Magn. Mater. **320**, 579 (2008).
[24] J. Fassbender and J. McCord, Appl. Phys. Lett. **88**, 252501 (2006).
[25] J. Fassbender, J. von Borany, A. Mücklich, K. Potzger, W. Möller, J. McCord, L. Schultz, and R. Mattheis, Phys. Rev. B **73**, 184410 (2006).
[26] W. Möller, W. Eckstein, and J. P. Biersack, Comput. Phys. Commun. **51**, 355 (1988).
[27] J. O. Rantschler, R. D. McMichael, A. Castillo, A. J. Shapiro, J. W. F. Egelhoff Jr., B. B. Maranville, D. Pulugurtha, A. P. Chen, and L. M. Connors, J. Appl. Phys. **101**, 033911 (2007).
[28] R. Meckenstock, I. Barsukov, C. Bircan, A. Remhof, D. Dietzel, and D. Spoddig, J. Appl. Phys. **99**, 08C706 (2006).
[29] I. Barsukov, P. Landeros, R. Meckenstock, J. Lindner, B. Krumme, H. Wende, D. L. Mills, and M. Farle (unpublished).
[30] N. Benatmane, S. P. Crane, F. Zavaliche, R. Ramesh, and T. W. Clinton, Appl. Phys. Lett. **96**, 082503 (2010).
[31] S. S. Kalarickal, Nan Mo, P. Krivosik, and C. E. Patton, Phys. Rev. B **79**, 094427 (2009).
[32] Z. Frait, Phys. Status Solidi B **3**, K408 (1963).
[33] B. Heinrich and J. A. C. Bland, *Ultrathin Magnetic Structures* (Springer, Berlin, 1994), Vols. I–III.
[34] M. Sparks, *Ferromagnetic-Relaxation Theory* (McGraw-Hill, New York, 1964).
[35] P. Landeros and D. L. Mills (unpublished).