

Modification of magnetic properties and structure of Kr⁺ ion-irradiated CrPt₃ films for planar bit patterned media

T. Kato,^{1,a)} S. Iwata,¹ Y. Yamauchi,² and S. Tsunashima²

¹*Department of Quantum Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8603, Japan*

²*Department of Electrical Engineering and Computer Science, Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8603, Japan*

(Received 26 May 2009; accepted 28 July 2009; published online 8 September 2009)

30 keV Kr⁺ ions were irradiated onto L₁₂ ordered CrPt₃ (20 nm) alloy films fabricated by a magnetron sputtering deposition followed by an appropriate heat treatment, and the modification of the structure and magnetic properties of the CrPt₃ films was investigated in detail. The fabricated L₁₂ CrPt₃ (20 nm) onto a fused quartz substrate exhibited a quite large perpendicular anisotropy of 5×10^6 ergs/cc, due to the large lattice distortion of 1%. The large perpendicular anisotropy decreased with increasing the ion dose, and became almost zero at the ion dose of 2×10^{14} ions/cm² due to the loss of the magnetic order of CrPt₃. The suppression of the magnetic order of CrPt₃ by the Kr⁺ ion irradiation is attributed to the transformation of the structure from the L₁₂ phase to a disordered fcc phase, which was confirmed by x-ray diffraction analysis. Unlike the magnetization and perpendicular anisotropy, the anisotropy field of CrPt₃ does not decrease significantly with the increase in the Kr⁺ ion dose, and exhibited more than 20 kOe even at the ion dose of 1×10^{14} ions/cm². This is considered to be attributed that the lattice distortion of CrPt₃ unchanged even at the Kr⁺ ion dose of 2×10^{14} ions/cm². © 2009 American Institute of Physics. [doi:10.1063/1.3212967]

I. INTRODUCTION

Bit patterned media have attracted considerable interest as future high density magnetic recording media, since they provide a promising technology to postpone the problem of superparamagnetic limit, i.e., thermal instability of the recorded bits in the media. One of the problems for the practical use of the bit patterned media is topography of discrete magnetic bits defined by lithographical fabrication, because the rough surface of the disk disturbs stable flying of the hard disk drive (HDD) head. Ion-beam irradiation has been proposed as a new approach to pattern magnetic materials locally without etching magnetic materials, i.e., without altering the surface topography,^{1,2} and ion irradiation into Co/Pt,¹⁻⁴ Co/Pd,^{5,6} and Co/Au^{7,8} multilayers (MLs) has been reported for the modification of their perpendicular anisotropies. However, in the Co/Pt, Co/Pd, and Co/Au MLs patterned by ion irradiation, the adjacent magnetic bits are not magnetically isolated due to the exchange coupling through in-plane magnetized spacing, which will limit the ultimate density of the media.

Previously, we have reported the ion-beam patterned medium using a CrPt₃ alloy film.⁹ The CrPt₃ shows ferrimagnetism when it has an ordered L₁₂ phase, while paramagnetism when it has a disordered fcc phase, and Hellwig *et al.*¹⁰ reported that 700 keV N⁺ ion irradiation can destroy the ferrimagnetism of CrPt₃. However, such high-energy and low-mass ion irradiation is not practical for fabrication of nanopatterned media due to the deep ion penetration depth (~500 nm).¹¹ Thus we have reported the Kr⁺ or Ar⁺ ion

irradiation onto CrPt₃, and presented the read-back signals from a bit patterned CrPt₃ disk fabricated by nanoimprint patterning and ion irradiation.⁹

In this paper, variation of the structure and magnetic properties of CrPt₃ with 30 keV Kr⁺ ion irradiation is discussed in detail. In case of high-energy light ion irradiation, initial crystal structure and microstructure (grain size) are known to be maintained,¹¹ while the heavy ion irradiation may change such structures and surface roughness. The large perpendicular anisotropy of CrPt₃, which is the most important property as a recording medium, results from the lattice distortion induced in a CrPt₃ film.¹² The structural change due to the Kr⁺ ion irradiation may limit the ultimate density of the CrPt₃ bit patterned media. Thus the modification of the structure by the heavy ion irradiation (30 keV Kr⁺) and the associated change in the magnetic properties were investigated.

II. EXPERIMENTAL METHOD

L₁₂ phase CrPt₃ films were obtained by postannealing of Cr/Pt MLs. The [Cr(0.4 nm)/Pt(1.5–1.7 nm)]₁₀ MLs were prepared by alternating sputtering of Cr and Pt. We used fused quartz (SiO₂) or thermally oxidized (500 nm SiO₂) silicon as a substrate. The samples were annealed in vacuum ($<3 \times 10^{-4}$ Pa) at a temperature of 850 °C for 15 min, and then cooled down to room temperature at a rate of about 10 °C/min. 30 keV Kr⁺ ions were produced by using ion implantation system and irradiated onto L₁₂ phase CrPt₃ to suppress its magnetic order. The Kr⁺ ion was selected using mass separation electromagnet, and the ion-beam current

^{a)}Electronic mail: takeshik@nuee.nagoya-u.ac.jp.

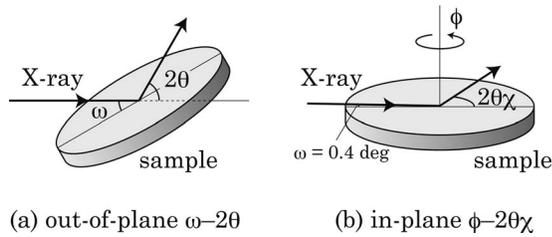


FIG. 1. Schematic drawing of the two types of x-ray scans: (a) out-of-plane ω - 2θ scan and (b) in-plane ϕ - $2\theta\chi$ scan. The x-ray incident angle ω was set to be 0.4° for the in-plane ϕ - $2\theta\chi$ scan.

density was typically set to be about $0.25 \mu\text{A}/\text{cm}^2$. The ion dose into the CrPt₃ film was varied from 5×10^{12} to 2×10^{15} ions/ cm^2 .

The film structure was characterized by x-ray diffraction (XRD) with Cu $K\alpha$ radiation. Two types of scanning were performed to analyze the structure in detail: out-of-plane ω - 2θ scan and in-plane ϕ - $2\theta\chi$ scan. The schematic drawing of the two types of x-ray scans is shown in Fig. 1. The diffraction from lattice planes parallel and perpendicular to the film surface was analyzed by ω - 2θ and ϕ - $2\theta\chi$ scans, respectively. For the in-plane ϕ - $2\theta\chi$ scan, x-ray incident angle ω was set to be 0.4° . Hysteresis loops were measured by using an alternating gradient field magnetometer, and perpendicular anisotropy was estimated by a torque magnetometer applying a maximum field of 15 kOe at temperatures of 300–550 K.

III. RESULTS AND DISCUSSIONS

Figure 2 shows the temperature dependence of the amplitude of torque curve of a vacuum annealed CrPt₃ (20 nm) ordered alloy film onto a SiO₂ substrate. The torque curves

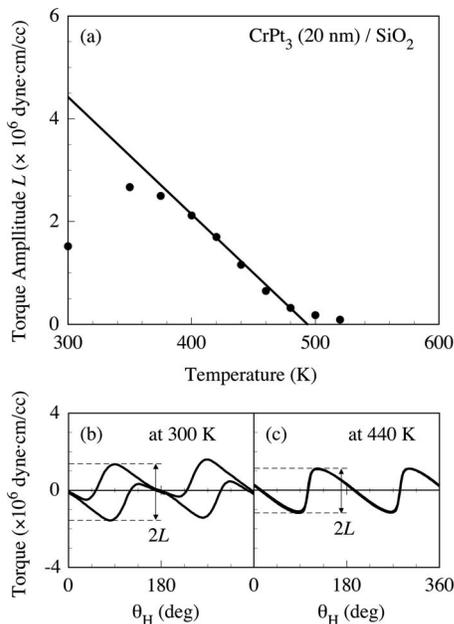


FIG. 2. Temperature dependence of the amplitude of torque curve of a vacuum annealed CrPt₃ (20 nm) ordered alloy film onto a fused quartz (SiO₂) substrate. The lower two figures (b) and (c) show torque curves taken at 300 and 440 K, respectively. The definition of the torque amplitude L was illustrated in (b) and (c).

taken at 300 and 440 K are shown in Figs. 2(b) and 2(c), respectively. The definition of the amplitude L was illustrated in the figures. As previously reported, the CrPt₃ (20 nm) onto SiO₂ exhibits a saturation magnetization M_s of 250 emu/cc and a large coercivity H_c of >12 kOe, which are suitable for the application to the perpendicular recording media.⁹ However, due to its large perpendicular anisotropy, the anisotropy constant at room temperature cannot be estimated simply by the amplitude of the torque curve because of the insufficient external field to rotate the magnetization (see the torque curve at 300 K). As shown in Fig. 2, the amplitude of the torque curve increased with reducing the temperature from 500 to 400 K, but it starts to decrease with further decrease in the temperature. This is due to the insufficient external field to rotate the magnetization during the torque curve measurement. To roughly estimate the perpendicular anisotropy of CrPt₃ (20 nm) onto SiO₂, the torque amplitude was extrapolated linearly to room temperature. The uniaxial anisotropy K_u of typical magnetic recording materials exhibits a nearly linear decrease in K_u as a function of temperature.¹³ Both classical single ion approach¹⁴ and more recent theoretical calculations¹⁵ predict the uniaxial anisotropy K_u scales as $K_u \sim M_s(T)^2$ (in the high temperature region). If we assume the decrease in M_s proportional to $T^{0.5}$ in the high temperature region just as the well-known mean field approximation, the linear decrease in K_u as a function of the temperature is described by these theories. Thus, we assumed the linear dependence of K_u on the temperature for the present CrPt₃ (20 nm) onto SiO₂. In this case, the effective anisotropy constant K_{eff} is also supposed to decrease linearly with temperature, and thus it was estimated to be 4.4×10^6 ergs/cc at room temperature. The perpendicular anisotropy constant K_u of CrPt₃ was represented by the sum of the K_{eff} and the shape anisotropy $2\pi M_s^2$, and resulted in 5×10^6 ergs/cc. The perpendicular anisotropy of CrPt₃ alloy films originates from a strain induced uniaxial anisotropy, and the strain is considered to result from the difference of the thermal expansion coefficient between the CrPt₃ and SiO₂ substrates. The strain will be relaxed in the thick film, and the relaxation of the strain results in the reduction in the perpendicular anisotropy. The CrPt₃ film studied here has total thicknesses of 20 nm, so that larger perpendicular anisotropy K_u than that of the previous report (total thickness 60 nm)¹² was obtained.

Figure 3 shows the dependence of the M_s , H_c , K_u , and saturation field H_k of the ion-irradiated CrPt₃ (20 nm) films on the 30 keV Kr⁺ ion dose amount. The saturation field H_k was calculated simply as $H_k = 2K_{\text{eff}}/M_s$, where $K_{\text{eff}} = K_u - 2\pi M_s^2$. The M_s , H_c , and K_u of CrPt₃ ordered alloy films start to decrease from the Kr⁺ irradiation dose of 10^{13} ions/ cm^2 , and these parameters became zero by a quite low ion dose of 2×10^{14} ions/ cm^2 . This ion dose of Kr⁺, 2×10^{14} ions/ cm^2 , is much lower than that of the previous report using 700 keV N⁺ ions.¹⁰ Atomic force microscope (AFM) measurements show that such a low ion dose of 2×10^{14} ions/ cm^2 has no influence on the surface topology and the film thickness. In the previous report on 22 keV Ga⁺ ion irradiation into Co/Pd MLs, Ga⁺ ion dose of 2×10^{15} ions/ cm^2 is reported to cause the film etching of

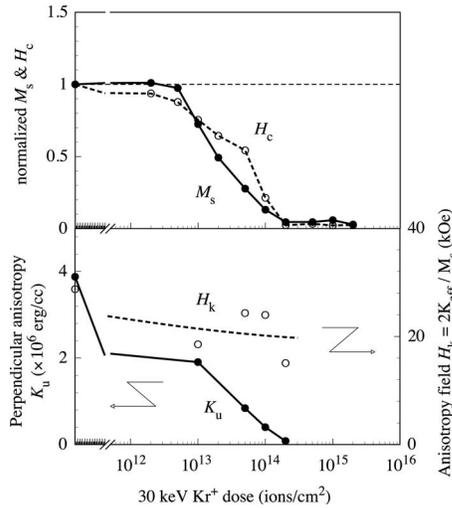


FIG. 3. 30 keV Kr^+ ion dose dependence of saturation magnetization M_s , coercivity H_c , perpendicular anisotropy K_u , and saturation field H_k of the ion-irradiated CrPt_3 (20 nm) films onto a thermally oxidized Si substrate. The M_s and H_c are normalized as the values before irradiation to be 1.

~ 2 nm.¹⁶ If we assume the same sputtering yield for the present 30 keV Kr^+ irradiation, the etching depth after the irradiation of 2×10^{14} ions/cm² is estimated to be 0.2 nm, which is lower than the resolution limit of our AFM, 0.5 nm. Unlike the dose dependence of M_s , H_c , and K_u , the H_k does not decrease significantly with the increase in the ion dose, and exhibits more than 20 kOe even at the ion dose of 1×10^{14} ions/cm². This indicates that the variation of K_u with increasing the ion dose is similar to that of M_s . The large H_k observed at an ion dose of 1×10^{14} ions/cm² may be related that the lattice distortion of CrPt_3 unchanged with the Kr^+ ion irradiation, which will be described later.

Figure 4 shows the (a) out-of-plane and (b) in-plane x-ray profiles of as-ordered CrPt_3 films on a thermally oxidized Si substrate. The as-ordered CrPt_3 film exhibits strong 111 and 222 reflections, which indicates the (111) orientation of the film. In the in-plane profile, 110 and 211 superlattice

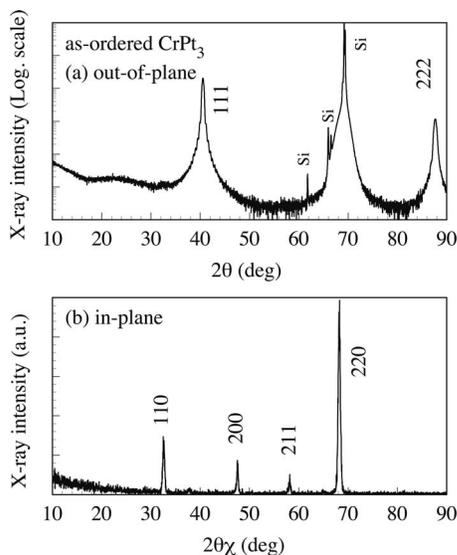


FIG. 4. (a) Out-of-plane and (b) in-plane x-ray profiles of as-ordered CrPt_3 films on a thermally oxidized Si substrate.

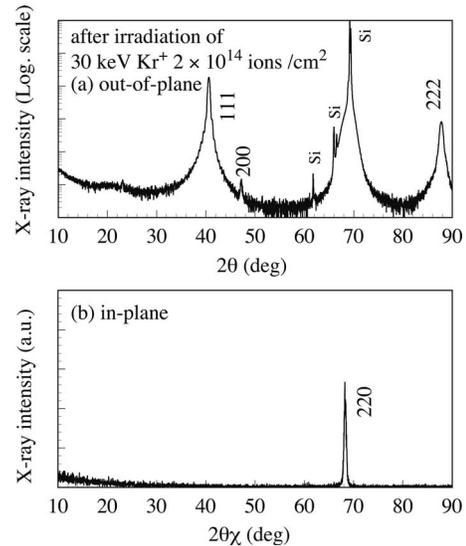


FIG. 5. (a) Out-of-plane and (b) in-plane x-ray profiles of CrPt_3 films after irradiation of 30 keV Kr^+ 2×10^{14} ions/cm².

lines, indicating the formation of the $L1_2$ phase, were clearly seen. No diffraction peaks indicating the oxidation of CrPt_3 and/or intermixing between the substrate (SiO_2) and the film were seen in Figs. 4(a) and 4(b). The order parameter estimated from the integrated intensities of the 110 superlattice line and 220 fundamental line was $S=0.9$, and thus the as-ordered CrPt_3 film is considered to have an almost perfect $L1_2$ ordered structure. The in-plane and out-of-plane lattice constants of CrPt_3 are $a_i=3.89$ Å and $a_p=3.85$ Å, respectively. Thus, the lattice distortion $\varepsilon=(a_i-a_p)/a_p$, contributing to the large perpendicular anisotropy of CrPt_3 , was estimated to be about 1%. Figure 5 shows the (a) out-of-plane and (b) in-plane profiles of a CrPt_3 (20 nm) film after Kr^+ ion irradiation of 2×10^{14} ions/cm². The out-of-plane x-ray profile after the ion irradiation shows a strong (111) orientation, and was almost identical to that of as-ordered CrPt_3 shown in Fig. 4(a). On the other hand, as shown in Fig. 5(b), the 110 and 211 superlattice lines completely disappeared after the ion dose of Kr^+ , 2×10^{14} ions/cm², which means the CrPt_3 film was transformed from a $L1_2$ phase to a disordered fcc phase by the ion irradiation. From Figs. 4 and 5, we found that the Kr^+ ion irradiation of 2×10^{14} ions/cm² does not affect the lattice distortion of CrPt_3 , contributing its large perpendicular anisotropy. The in-plane and out-of-plane lattice constants of CrPt_3 after the Kr^+ irradiation of 2×10^{14} ions/cm² remained $a_i=3.89$ Å and $a_p=3.85$ Å, respectively. When the CrPt_3 disordered by Kr^+ ion irradiation is annealed again at 850 °C, the CrPt_3 re-exhibits ferrimagnetism and the strong perpendicular anisotropy due to the recovery of the $L1_2$ ordering.

Figure 6 shows the dependence of the lattice distortion ε and order parameter S on the 30 keV Kr^+ ion dose amount. The lattice distortion was estimated from the out-of-plane and in-plane lattice constants, a_p and a_i , respectively, as $\varepsilon=(a_i-a_p)/a_p$. The order parameter S decreased with increasing Kr^+ ion dose. The decreases in the ordering parameter will cause the reduction in the Curie temperature of CrPt_3 , which will be observed as the decrease in M_s and K_u as

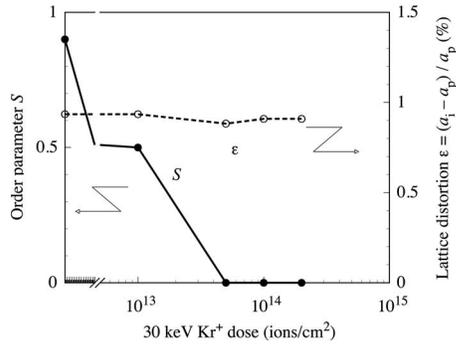


FIG. 6. Dependence of the lattice distortion ε and order parameter S on the 30 keV Kr^+ ion dose amount. The lattice distortion was estimated from the out-of-plane and in-plane lattice constants, a_p and a_i , respectively, as $\varepsilon = (a_i - a_p) / a_p$.

discussed in Fig. 3. The ordering parameter S became almost zero at the ion dose of 5×10^{13} ions/cm² as shown in Fig. 6. This ion dose is smaller than that necessary to suppress the magnetization of CrPt_3 , 2×10^{14} ions/cm² in Fig. 3. This is considered to be due to the limited penetration depth of incident x ray in the in-plane ϕ - $2\theta\chi$ scan. For fixing the incident x-ray $\omega = 0.4^\circ$, the x-ray probing depth will be limited to be about 10 nm from the surface. Thus the structure of CrPt_3 within this probing depth will be reflected in the in-plane profile. By TRIM code simulation, it was confirmed that the number of vacancies in CrPt_3 generated by the 30 keV Kr^+ ion impact decreased with increasing the distance from the surface. The lattice distortion ε was unchanged even when the Kr^+ ion dose was increased to 2×10^{14} ions/cm², as shown in Fig. 6. We consider this is the reason why a large anisotropy field H_k was observed even at a high Kr^+ ion dose as shown in Fig. 3, since the lattice distortion is the origin of the large perpendicular anisotropy of CrPt_3 . In order to apply the CrPt_3 to an ion-irradiated patterned medium, the relaxation of the lattice distortion by the ion irradiation will be a significant problem, because the irradiated (relaxed) region may affect the distortion and perpendicular anisotropy of the nonirradiated region. However, such a relaxation was not confirmed in the 30 keV Kr^+ ion irradiation up to 2×10^{14} ions/cm², and thus the CrPt_3 ordered alloy is considered to be a candidate for the future high density ion-irradiated bit patterned media.⁹

IV. CONCLUSION

$L1_2$ ordered CrPt_3 alloy films were prepared by postannealing of magnetron sputtered Cr/Pt MLs. The fabricated $L1_2$ CrPt_3 (20 nm) onto a SiO_2 substrate exhibited a quite large perpendicular anisotropy of 5×10^6 ergs/cc, due to the large lattice distortion of 1%. The CrPt_3 film was irradiated by 30 keV Kr^+ ions, and the modification of the structure and magnetic properties has been investigated in detail. The satu-

ration magnetization, coercivity, and perpendicular anisotropy decreased with increasing Kr^+ ion dose and became almost zero at a quite low ion dose of 2×10^{14} ions/cm², which is a quite useful property for the high density ion-irradiated patterned media. The suppression of the magnetic order of CrPt_3 by the Kr^+ ion irradiation is attributed to the transformation of the structure from the $L1_2$ phase to a disordered fcc phase, which was confirmed by XRD analysis. Unlike the magnetization and perpendicular anisotropy, the anisotropy field does not decrease significantly with the increase in the ion dose, and exhibits more than 20 kOe even at the ion dose of 1×10^{14} ions/cm². This is considered to be related to the fact that the lattice distortion of CrPt_3 unchanged even when the Kr^+ ion dose was increased to 2×10^{14} ions/cm². The results obtained here indicate that the Kr^+ ion irradiation onto CrPt_3 is a promising technology to develop future high density ion-irradiated bit patterned media.

ACKNOWLEDGMENTS

The authors are grateful to Dr. K. Matsumoto and Dr. T. Morikawa of Yamagata Fujitsu Ltd., and Dr. K. Ozaki of Fujitsu Laboratories Ltd. for helpful discussions, and would like to thank Mr. M. Kumazawa, and Mr. Y. Adachi of Nagoya University for assistance in the experiments and film composition analysis, respectively.

- ¹C. Chappert, H. Bernas, J. Ferré, V. Kottler, J.-P. Jamet, Y. Chen, E. Cambril, T. Devolder, F. Rousseaux, V. Mathet, and H. Launois, *Science* **280**, 1919 (1998).
- ²B. D. Terris, L. Folks, D. Weller, J. E. E. Baglin, J. Kellock, H. Rothuizen, and P. Vettiger, *Appl. Phys. Lett.* **75**, 403 (1999).
- ³J. Ferré, C. Chappert, H. Bernas, J.-P. Jamet, P. Meyer, O. Kaitasov, S. Lemerle, V. Mathet, F. Rousseaux, and H. Launois, *J. Magn. Magn. Mater.* **198–199**, 191 (1999).
- ⁴R. Hyndman, P. Warin, J. Gierak, J. Ferré, J. N. Chapman, J. P. Jamet, V. Mathet, and C. Chappert, *J. Appl. Phys.* **90**, 3843 (2001).
- ⁵E. Suharyadi, S. Natsume, T. Kato, S. Tsunashima, and S. Iwata, *IEEE Trans. Magn.* **41**, 3595 (2005).
- ⁶E. Suharyadi, T. Kato, S. Tsunashima, and S. Iwata, *IEEE Trans. Magn.* **42**, 2972 (2006).
- ⁷T. Blon, G. B. Assayag, J.-C. Qusset, B. Pecassou, A. Claverie, and E. Snoeck, *Nucl. Instrum. Methods Phys. Res. B* **257**, 374 (2007).
- ⁸M. Urbaniak, F. Stobiecki, D. Engel, B. Szymanski, and A. Ehresmann, *Acta Phys. Pol. A* **115**, 326 (2009).
- ⁹T. Kato, S. Iwata, Y. Yamauchi, S. Tsunashima, K. Matsumoto, T. Morikawa, K. Ozaki, *J. Appl. Phys.*, **105**, 07C117 (2009).
- ¹⁰O. Hellwig, D. Weller, A. J. Kellock, J. E. E. Baglin, and E. E. Fullerton, *Appl. Phys. Lett.* **79**, 1151 (2001).
- ¹¹J. Fassbender, D. Ravelosona, and Y. Samson, *J. Phys. D* **37**, R179 (2004).
- ¹²T. Kato, H. Ito, K. Sugihara, S. Tsunashima, and S. Iwata, *J. Magn. Magn. Mater.* **272–276**, 778 (2004).
- ¹³J.-U. Thiele, K. R. Coffey, M. F. Toney, J. A. Hedstrom, and A. J. K. Kellock, *J. Appl. Phys.* **91**, 6595 (2002).
- ¹⁴H. B. Callen and E. Callen, *J. Phys. Chem. Solids* **27**, 1271 (1966).
- ¹⁵R. Skomski, A. Kashyap, and D. J. Sellmyer, *IEEE Trans. Magn.* **39**, 2917 (2003).
- ¹⁶E. Suharyadi, S. Natsume, T. Kato, S. Tsunashima, and S. Iwata, *Trans. Magn. Soc. Jpn.* **5**, 125 (2005).