HOKKAIDO UNIVERSITY

Title	Nanopatterns induced by pulsed laser irradiation on the surface of an Fe-Al alloy and their magnetic properties
Author(s)	Yoshida, Yutaka; Oosawa, Kazuya; Watanabe, Seiichi; Kaiju, Hideo; Kondo, Kenji; Ishibashi, Akira; Yoshimi, Kyosuke
Citation	Applied Physics Letters, 102, 183109 https://doi.org/10.1063/1.4804363
Issue Date	2013-05-07
Doc URL	http://hdl.handle.net/2115/52675
Rights	Copyright 2013 American Institute of Physics. This article may be downloaded for personal use only. Any other use requires prior permission of the author and the American Institute of Physics. The following article appeared in Applied Physics Letters 102, 183109 (2013) and may be found at https://dx.doi.org/10.1063/1.4804363
Type	article
File Information	_ojpstmp_stampPdf_d_07T10_18_APPLAB_102_18_183109_1.pdf

Instructions for use

Hokkaido University Collection of Scholarly and Academic Papers : HUSCAP

AIP | Applied Physics Letters

Nanopatterns induced by pulsed laser irradiation on the surface of an Fe-Al alloy and their magnetic properties

Yutaka Yoshida, Kazuya Oosawa, Seiichi Watanabe, Hideo Kaiju, Kenji Kondo et al.

Citation: Appl. Phys. Lett. **102**, 183109 (2013); doi: 10.1063/1.4804363
View online: http://dx.doi.org/10.1063/1.4804363
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v102/i18
Published by the American Institute of Physics.

Additional information on Appl. Phys. Lett.

Journal Homepage: http://apl.aip.org/
Journal Information: http://apl.aip.org/about/about_the_journal
Top downloads: http://apl.aip.org/features/most_downloaded
Information for Authors: http://apl.aip.org/authors

ADVERTISEMENT

Nanopatterns induced by pulsed laser irradiation on the surface of an Fe-Al alloy and their magnetic properties

Yutaka Yoshida,[1] Kazuya Oosawa,[2,a)] Seiichi Watanabe,[1,2,b)] Hideo Kaiju,[3,4] Kenji Kondo,[3] Akira Ishibashi,[3] and Kyosuke Yoshimi[5]

[1]*Center for Advanced Research of Energy and Materials, Faculty of Engineering, Hokkaido University, Kita-8, Nishi-13, Kita-ku, Sapporo, Hokkaido 060-8628, Japan*
[2]*Division of Quantum Science and Engineering, Graduate School of Engineering, Hokkaido University, Kita-8, Nishi-13, Kita-ku, Sapporo, Hokkaido 060-8628, Japan*
[3]*Laboratory of Nano-Structure Physics, Research Institute for Electronic Science, Hokkaido University, Sapporo, Hokkaido 001-0020, Japan*
[4]*Japan Science and Technology Agency, PRESTO, Kawaguchi, Saitama 332-0012, Japan*
[5]*Graduate School of Engineering, Tohoku University, Sendai, 6-6-02 Aramaki Aza Aoba, Miyagi 980-8579, Japan*

(Received 25 March 2013; accepted 19 April 2013; published online 7 May 2013)

We have studied nanopatterns induced by nanosecond pulsed laser irradiation on (111) plane surfaces of a polycrystalline iron-aluminum alloy and evaluated their magnetic properties. Multiple nanosecond pulsed laser irradiation induces a wavelength-dependent surface transformation of the lattice structure from a B2-type to a supersaturated body centered cubic lattice. The selective formation of surface nanopatterns consisting of holes, stripes, polygonal networks, and dot-like nanoprotrusions can be observed. Furthermore, focused magneto-optical Kerr effect measurements reveal that the magnetic properties of the resultant nanostructured region changes from a paramagnetic to a ferromagnetic phase in accordance with the number of laser pulses. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4804363]

Nanostructures have attracted interest since nanodot arrays and nanopatterns, for example, have been found to possess many enhanced functional properties.[1–8] Quantum beam technologies, such as ion beams[1,2] and lasers,[9–17] have often been used for the efficient development of nanostructured materials. Lasers, in particular, are expected to serve as an increasingly useful tool for obtaining nanostructured materials because nanostructured surface morphologies can be fabricated by laser irradiation[9–14] and laser-assisted direct imprinting.[16] Recently, we have proposed an approach whereby unique nanostructure patterns can be fabricated in a gold-silicon system by pulsed laser irradiation.[14] This approach is thought to be applicable to other alloy systems as well. We herein focus on an iron-aluminum (Fe-Al) alloy, which exhibits transitions of the internal structure in thermal equilibrium states through changes in Al concentration at an atomic scale.[18,19] Magnetic properties in this system can also be controlled by adjusting the composition. For an Al concentration of 0%–28%, the material exists in a ferromagnetic phase because of the lattice atomic structure being body centered cubic (bcc). In the range between 28% and 33%, the structure transforms from a bcc lattice to a bcc + B2-type structure. The magnetic properties of the alloy indicate soft-magnetism owing to the existing phase separation. When the Al concentration is 33%–48%, the material exists in a paramagnetic phase because the system possesses a B2-type structure.[18,19]

On the other hand, nonequilibrium phenomena such as thermal quenching do not obey the rules pertaining to thermal equilibrium state diagrams. As a typical example, surface morphologies of the B2-type Fe-Al alloy have been modified by thermal treatment, which has formed surface self-patterning structures through thermal quenching.[20,21] Laser beam irradiation has also induced self-organization by laser quenching.[12–14]

Therefore, for this alloy, laser beam irradiation may control not only surface morphologies but also magnetic properties. In this report, we demonstrate the formation of nanopatterns of holes, stripes, and polygonal networks and dot-like nanoprotrusions on the (111) plane of a polycrystalline Fe-48%Al alloy, induced by nanosecond pulsed laser irradiation. We also show their magnetic properties of this material, in which the transition from paramagnetism to ferromagnetism is observed in accordance with the number of laser pulses.

Fe-Al alloy button ingots, having Al concentration of 48%, were prepared using an arc-melting technique in an argon gas atmosphere. The polycrystalline Fe-Al alloy was synthesized as a B2-type structure. For laser irradiation, the surface was polished to a mirror finish. The specimens were ultrasonically cleaned and then subjected to pulsed laser irradiation. The B2-type Fe-Al alloy surface was irradiated in air with an Nd:YAG pulse laser (Continuum Co., Ltd., Inlite II) at a wavelength of 532 nm, a repetition frequency of 2 Hz, and a pulse width of 5–7 ns at room temperature. The laser beam diameter was 6 mm, and irradiation was conducted normal to the surface with linearly polarized light having an average laser energy density of 1.24 kJ/m^2.

After laser irradiation, the surface morphology of the specimens was observed using a scanning electron microscope (SEM: JEOL JSM-7001F). In order to examine the crystalline orientation, the electron backscatter diffraction (EBSD) pattern was measured. Microstructural and

a)Present address: Japan Nuclear Fuel, Ltd., 4-108 Aza Okitsuke, Oaza Obuchi, Rokkasho-mura, Kamikita-gun, Aomori-ken 039-3212, Japan.
b)Author to whom correspondence should be addressed. Electronic mail: sw004@eng.hokudai.ac.jp

 © 2013 AIP Publishing LLC

microchemical analyses were performed using a transmission electron microscope (TEM: JEOL JEM-2010F) and energy-dispersive X-ray spectroscopy (EDS: Noran Vantage) systems. The thin foil specimens for TEM observation were prepared using a focused ion beam system.

The magnetization curves were measured by focused magneto-optic Kerr effect (MOKE) equipment (NEOARK, BH-PI920-HU) under a magnetic field of up to 1 kOe at room temperature.[23] The spot size for the observation of the focused MOKE signal was set to 3 μm.

First, the development of nanopatterns on the laser-irradiated sample was examined using SEM. Then, since nanopattern development of the polycrystalline Fe-Al alloy is dependent on the crystalline orientation, an investigation of the crystalline orientation of each grain by EBSD analysis was conducted. Figures 1(a)–1(d) show SEM images and the results of EBSD analysis of an irradiated polycrystalline Fe-Al alloy surface after 200 pulses. Nanopattern development by this method is observed to occur more rapidly on the (111) plane, compared to other planes such as the (100) and (110) planes, as shown in Figs. 1(a)–1(c). Moreover, it is proposed that the fabrication of the observed periodic nanopattern on the (111)-oriented surface is attributable not only to laser irradiation but also to the crystalline orientation because the direction of the periodic pattern corresponds with the [110] direction, as shown in Figs. 1(c) and 1(d). In fact, the formation of the nanopattern is predominately attributed to the crystalline orientation. Hereafter, this study shall focus on the irradiated (111) plane surface because the development of nanopatterns by laser beam irradiation of the (111) plane is expected to have a more pronounced effect.

Figure 2 shows SEM images of nanopatterns on the (111) surface of the polycrystalline Fe-Al alloy after (a) 0, (b) 10, (c) 50, (d) 200, (e) 300, and (f) 2000 pulses, respectively. The surface is smooth before irradiation, as shown in Fig. 2(a). After irradiation with 10 pulses, small holes are produced randomly on the surface, having diameter between 0.5 and 1.5 μm, as shown in Fig. 2(b). As shown in Fig. 2(c), an aligned pattern of small holes is formed locally after 50 pulses. These aligned strip patterns can be observed along the [110] direction on the (111)-oriented plane. As shown in Fig. 2(d), the stripe pattern is formed after 200 pulses by surface self-assembly of holes, having an area of more than 200 μm^2. The experimental results suggest that the formation of small holes is attributed to the dewetting process[23] because the surface is of a smoothly curved shaped and not a faceted plane, such as the case for the (100) plane.[21,22] Later, the adjacent stripe lines are connected with increasing number of laser pulses, and the nanopattern develops into a polygonal network structure, as shown in Fig. 2(e). The network structure begins to break up with an increasing number of pulses in excess of 300 pulses. The transformation from a stripe to a polygonal network structure occurs through connections formed between lines from the other directions, such as the [101] and [011] directions. Finally, a periodic nanodot-like pattern results as the number of laser pulses approaches 2000, as shown in Fig. 2(f). The position of a nanodot-like pattern on the surface stabilizes with periodical distance. A polygonal network nanopattern and a nanodot-like pattern are observed to occur over approximately a 25 μm^2 area. The magnetic properties of the surface are

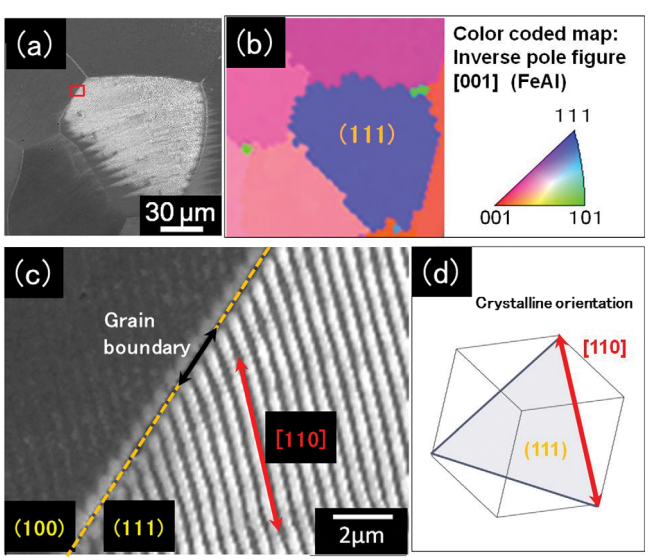

FIG. 1. SEM images and EBSD analysis of various plane surfaces of the polycrystalline Fe-48%Al alloy irradiated with 200 pulses.

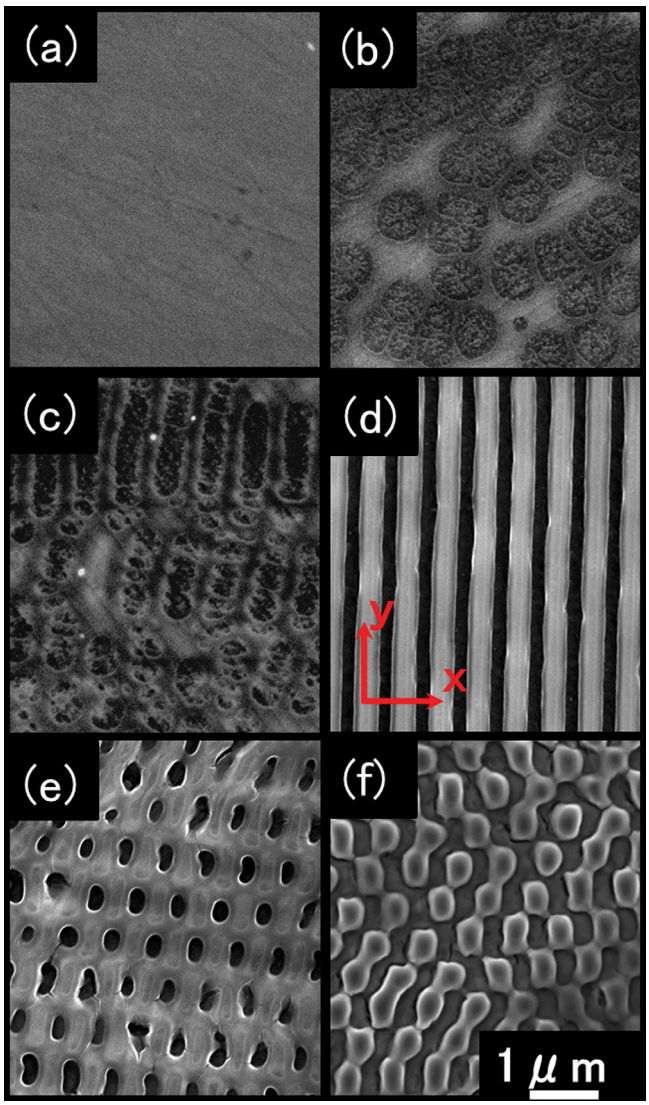

FIG. 2. SEM images of the (111) plane surface of the Fe-48%Al alloy irradiated with (a) 0, (b) 10, (c) 50, (d) 200, (e) 300, and (f) 2000 pulses.

expected to be enhanced by the periodically arrayed structure of the resulting nanodot-like pattern.

In addition, to investigate the details of the periodic nanopattern, we carried out cross-sectional TEM observations of the internal structures of stripe lines in samples irradiated for 200 pulses. Figure 3 shows a cross-sectional TEM image along the x-direction illustrated in Fig. 2(d) in (a), diffraction patterns along the [110] direction in a bulk Fe-Al alloy in (b) and (c), and the results of EDS point analyses in (d) and (e). The spacing between the stripe lines was an average of 530 nm (whose distance is equivalent to the incident laser wavelength), and the periodic surface roughness consisted of a uniformly waving structure that was an average of 150–200 nm in height. The dimensions of the structure of a stripe line was an average of 244 nm in width and a maximum 240 nm in depth by darkly contrasted part, as suggested by Fig. 3(a). The smoothly curved shape of the surface and the internal roughness of the periodic nanostructure suggest that the formation is attributed to the dewetting process.[24] Figures 3(b) and 3(c) show diffraction patterns along the [110] direction given by arrows *b* and *c*, respectively, given in Fig. 3(a). The nanostructure of the darkly contrasted surface section is epitaxial because the spot angles of the diffraction pattern are the same. However, the (111) spots of the B2-type structure disappear from the diffraction pattern of the darkly contrasted region, as shown in Fig. 3(b). From these results, it is clear that the darkly contrasted region is the melting zone produced by the thermal effect of laser irradiation wherein the temperature exceeds 1700 K (the alloy's melting temperature). Elementary analyses of the stripe structure, at point *b*, and the bulk, at point *c* (as shown in Fig. 3(a)), by EDS provide almost equivalent results, as shown in Figs. 3(d) and 3(e). These results conclude that the lattice structure on the Fe-Al alloy surface transforms from a B2-type to a supersaturated bcc structure.

Finally, focused MOKE measurements were conducted for investigating the magnetic properties of the fabricated nanopatterns. Figure 4 shows the magnetization curves of nanopatterns on the (111) surface of the polycrystalline Fe-Al alloy after (a) 0, (b) 10, (c) 50, (d) 200, (e) 300, and (f) 2000 pulses. The magnetic field was applied in the y-direction for each sample, as shown in Fig. 2. Figure 4(a) indicates that Kerr rotation is nearly zero degrees, and there is no coercive force. This behavior indicates paramagnetism, which has been already confirmed as being characteristic of Fe-48%Al, B2-type structures.[18,19] In comparison, a magnetic hysteresis loop is visible in Figs. 4(b) and 4(c), and coercive forces of 53 and 51 Oe are evident in the samples irradiated with 10 and 50 pulses, respectively. The Kerr rotations in the saturated field are measured as 0.6 and 1.0 m degrees, respectively. The appearance of the coercive force and Kerr signal is due to the existence of ferromagnetic phases. This indicates the coexistence of paramagnetic and ferromagnetic phases, which are attributed to Fe-48%Al, B2-type structures, and supersaturated bcc structures, respectively. These results are consistent with the fact that the measured coercive forces show good agreement with typical values obtained from Fe films[25] and that the Kerr rotations are small since only a few Fe bcc structures are formed on the surface of the sample. Then, as shown in Fig. 4(d), the

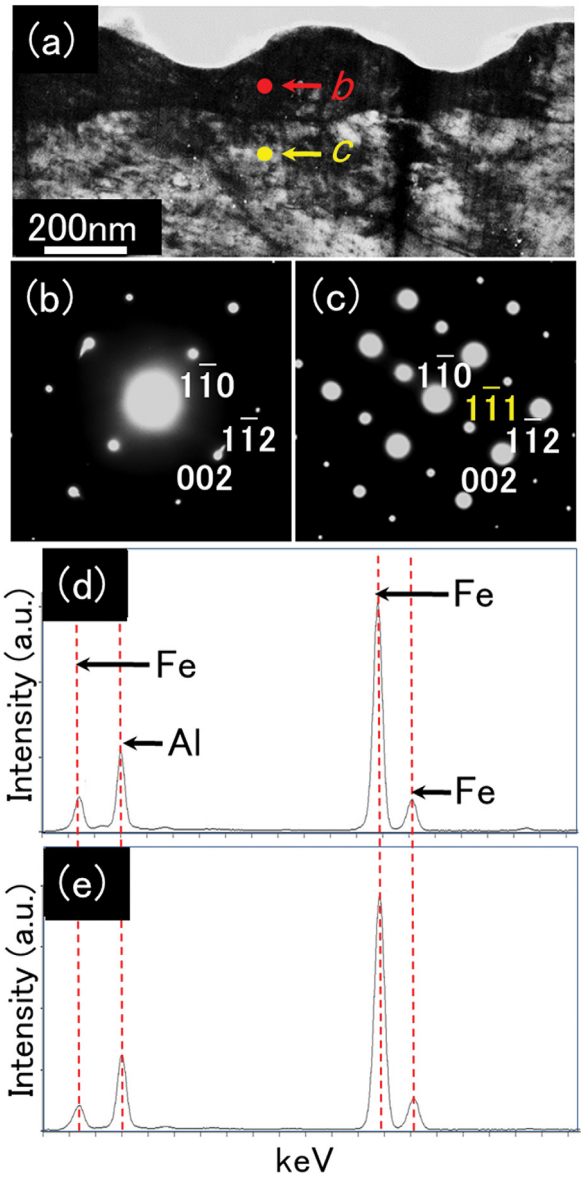

FIG. 3. TEM images, diffraction patterns, and EDS point analyses of the Fe-48%Al alloy surface after irradiation with 200 pulses. (a) A cross-sectional TEM image taken along the x-direction shown in Fig. 2(d). (b), (c) Diffraction patterns examined at the points in Fig. 3(a) given by the arrows *b* and *c*. (d), (e) EDS analyses at the points in Fig. 3(a) given by the arrows *b* and *c*.

coercive force expands to as large a value as 85 Oe, and the Kerr rotation in the saturated field increases to 15.0 m degrees. The evident large Kerr rotation is attributable to Fe bcc structures being formed over the surface of the sample, which is confirmed by the cross-sectional TEM observations shown in Fig. 3. The enhancement of the coercive force can be explained by the formation of Fe bcc structures and a strong shape magnetic anisotropy. As is well known, the shape magnetic anisotropy becomes strong when the magnetic field is applied in a direction parallel to the longitudinal direction of the stripe shape. This strong shape magnetic anisotropy enhances the coercive force. In Fig. 4(e), the coercive force decreases down to a value of 40 Oe, which is less than half the value obtained for the sample irradiated for 200 pulses, as shown in Fig. 4(d), while the Kerr rotation is nearly identical to that shown in Fig. 4(d). The observed reduction of the coercive force is attributable to the

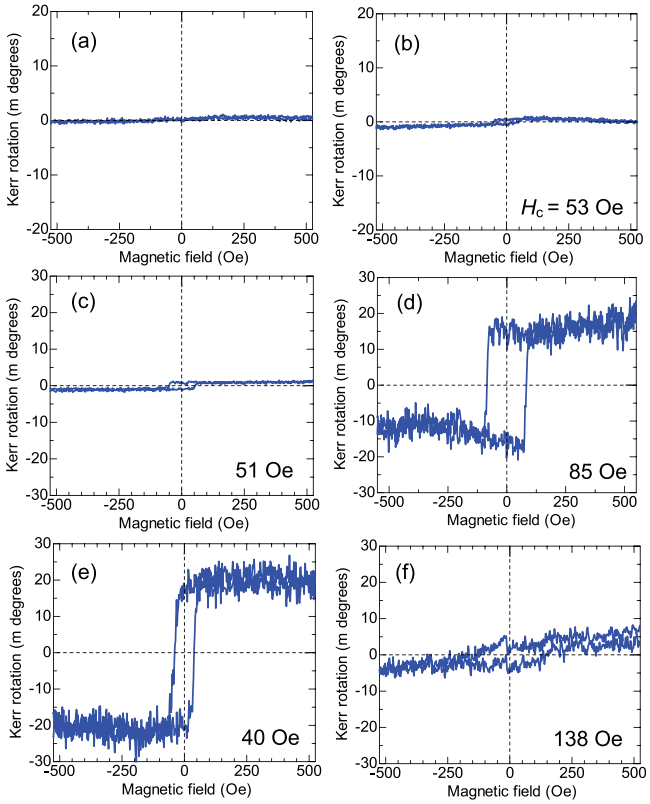

FIG. 4. Magnetization curves of the Fe-48%Al alloy after laser irradiation with (a) 0, (b) 10, (c) 50, (d) 200, (e) 300, and (f) 2000 pulses.

emergence of complex structures, i.e., polygonal networks, which are apparent in Fig. 2(e). Such complex structures in magnetic materials can form magnetic multidomains, which decrease the coercive force. In Fig. 4(f), the coercive force is as large as 138 Oe, and the Kerr rotation in the saturated field is as small as 3.5 m degrees. The enhancement of the coercive force originates from the formation of the dot-like protrusions evident in Fig. 2(f). For example, the increased coercivity observed for Fe nanodots is a well-known phenomenon for fine magnetic nanostructures.[7] The magnetic hysteresis loop of nanopattern at 300 K is much more sheared with larger coercive forces, H_c, of 110 Oe.[8] According to recent results, the coercive force of nanopatterns consisting of dot-like protrusions increases as compared with that of uniform films.[26] This is because the small dimensions of particles impede the formation of multidomains, and magnetization reversal proceeds through rotation. This explanation can be applied to the case illustrated by Fig. 4(f) since the dot-like structure can be formed as shown in Fig. 2(f). The reduction in the Kerr rotation is considered to be due to the formation of ultrathin metal-oxide layers, such as FeO, Fe_2O_3, Fe_3O_4, and Al_2O_3, on the surface of the sample. FeO and Al_2O_3 exhibit paramagnetism, γ-Fe_2O_3 and Fe_3O_4 exhibit ferrimagnetism, and α-Fe_2O_3 exhibits antiferromagnetism at room temperature.[27–29] The appearance of paramagnetic, ferrimagnetic, and antiferromagnetic phases on the surface of the sample causes a decrease in the magnetization, resulting in the reduction of the Kerr rotation.

We studied the surface nanopatterns formed on (111) plane surfaces of a polycrystalline Fe-48%Al alloy by pulsed laser irradiation and evaluated their magnetic properties. The development of nanopatterns can be controlled by the number of laser pulses, which result in the formation of holes, stripes, polygonal networks, and dot-like nanoprotrusions. Additionally, the surface magnetic properties transit from paramagnetism to ferromagnetism.

The authors thank Mr. Melbert Jeem, Dr. Muhammad Rafiq Mirza Bin Julaihi, and Professors Shigeo Yatsu, Tamaki Shibayama, and Norihito Sakaguchi for helpful discussions. We also thank Mr. Yuusuke Nishida, Mr. Junya Wajima, and Mr. Kenji Okubo for their experimental assistance. This research was partially supported by Precursory Research for Embryonic Science and Technology program (PRESTO) from the Japan Science and Technology Agency (JST).

[1]H. Liao, W. Wen, and G. K. L. Wong, J. Opt. Soc. Am. B **23**, 2518 (2006).
[2]S. Dhara, R. Kesavamoorthy, P. Magudapathy, M. Premila, B. K. Panigrahi, K. G. M. Nair, C. T. Wu, K. H. Chen, and L. C. Chen, Chem. Phys. Lett. **370**, 254 (2003).
[3]T. M. Parker, L. K. Wilson, N. G. Condon, and F. M. Leibsle, Phys. Rev. B **56**, 6458 (1997).
[4]J. Shen, R. Skomski, M. Klaua, H. Jenniches, S. S. Manoharan, and J. Kirschner, Phys. Rev. B **56**, 2340 (1997).
[5]J. Shen, M. Klaua, P. Ohresser, H. Jenniches, J. Barthel, Ch. V. Mohan, and J. Kirschner, Phys. Rev. B **56**, 11134 (1997).
[6]F. Komori, K. D. Lee, K. Nakatsuji, T. Iimori, and Y. Q. Cai, Phys. Rev. B **63**, 214420 (2001).
[7]T. Miyazaki and N. Tezuka, J. Mag. Mag. Mater. **139**, L231 (1995).
[8]B. D. Cullity, in *Introduction to Magnetic Materials*, edited by M. Cohen (Addison-Wesley, London, UK, 1972), Chap. 11.
[9]K. Liu, J. Noguēs, H. Masuda, K. Nishio, I. V. Roshchin, and I. K. Schuller, Appl. Phys. Lett. **81**, 4434 (2002).
[10]M. Birnbaum, J. Appl. Phys. **36**, 3688 (1965).
[11]J. F. Young, J. S. Preston, H. M. Vandriel, and J. E. Sipe, Phys. Rev. B **27**, 1155 (1983).
[12]J. E. Sipe, J. F. Young, J. S. Preston, and H. M. Vandriel, Phys. Rev. B **27**, 1141 (1983).
[13]S. Watanabe, Y. Yoshida, S. Kayashima, S. Yatsu, M. Kawai, and T. Kato, J. Appl. Phys. **108**, 103510 (2010).
[14]Y. Yoshida, N. Sakaguchi, S. Watanabe, and T. Kato, Appl. Phys. Express **4**, 055202 (2011).
[15]Y. Yoshida, S. Watanabe, Y. Nishijima, K. Ueno, H. Misawa, and T. Kato, Nanotechnology **22**, 375607 (2011).
[16]B. Cui, C. Kemimel, and S. Y. Chou, Nanotechnology **21**, 045303 (2010).
[17]J. Trice, D. Thomas, C. Favazza, R. Sureshkumar, and R. Kalyanaraman, Phys. Rev. B **75**, 235439 (2007).
[18]C. Favazza, J. Trice, and R. Kalyanaraman, Appl. Phys. Lett. **91**, 043105 (2007).
[19]A. Taylor and R. M. Jones, J. Phys. Chem. Solids **6**, 16 (1958).
[20]R. D. Shull, H. Okamoto, and P. A. Beck, Solid State Commun. **20**, 863 (1976).
[21]K. Yoshimi, M. Tsunekane, R. Nakamura, A. Yamauchi, and S. Hanada, Appl. Phys. Lett. **89**, 073110 (2006).
[22]M. Tsunekane, K. Yoshimi, and K. Murayama, Acta Mater. **56**, 3162 (2008).
[23]H. Kaiju, T. Abe, K. Kondo, and A. Ishibashi, J. Appl. Phys. **111**, 07C104 (2012).
[24]U. Thiele, M. Mertig, and W. Pompe, Phys. Rev. Lett. **80**, 2869 (1998).
[25]F. B. Wyart, P. Martin, and C. Redon, Langmuir **9**, 3682 (1993).
[26]R. K. Dumas, C.-P. Li, I. V. Roshchin, I. K. Schuller, and K. Liu, Phys. Rev. B **75**, 134405 (2007).
[27]D. V. Dimitrov, K. Unruh, G. C. Hadjipanayis, V. Papaefthymiou, and A. Simopoulos, Phys. Rev. B **59**, 14499 (1999).
[28]G. Yang, D. Gao, J. Zhang, J. Zhang, Z. Shi, and D. Xue, J. Phys. Chem. C **115**, 16814 (2011).
[29]S. V. Stolyar, O. A. Bayukov, Y. L. Gurevich, E. A. Denisova, R. S. Iskhakov, V. P. Ladygina, A. P. Puzyr, P. P. Pustoshilov, and M. A. Bitekhtina, Inorg. Mater. **42**, 763 (2006).