Artificially controlled magnetic domain structures in ferromagnetic dots/ferroelectric heterostructures

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A variation in the magnetization and magnetic domain structures of epitaxial Fe dots on a single crystal $BaTiO_3$ substrate is demonstrated in association with the structural phase transition of ferroelectric $BaTiO_3$. The temperature dependent magnetization of Fe dots drops suddenly at 282 K and increases again at 189 K with decreasing temperature. The variations clearly correspond to the successive structural phase transitions of $BaTiO_3$ from tetragonal to orthorhombic phases and from orthorhombic to rhombohedral phases. After a thermal cycle between room temperature and 150 K passing through these phase transitions, the initial magnetic domain structure of Fe dots with an enclosed magnetic flux structure changes to a single-domain-like structure due to interfacial strain between Fe and $BaTiO_3$ arising from possible switching of *c*-axis orientation of $BaTiO_3$ substrate.
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I. INTRODUCTION

Artificial control of magnetic domain structures using an electric means instead of a magnetic field is of crucial importance for developing novel spin electronic devices. To realize the electric control, ferromagnet/ferroelectric heterostructures can be one of the most promising candidates since ferroelectrics can induce a huge strain at the ferromagnet/ferroelectric interface under the application of voltage, achieving magnetization switching through a strain induced magnetoelectric coupling effect. A number of studies were reported using ferromagnet/ferroelectric heterostructures such as $La_{2/3}Sr_{1/3}MnO_3/BaTiO_3$ (BTO) heterostructures,[1,2] $Fe/BaTiO_3$ heterostructures,[3,4] CoPd/PZT heterostructures (where PZT denotes lead zirconate titanate),[5] $CoFe/BiFeO_3$ heterostructures,[6] Permalloy/$YMnO_3$/Permalloy heterostructures,[7] and GaMnAs/PZT ceramics[8] in this view. On the other hand, a tremendous theoretical prediction has also been reported, where the magnetic moments of Fe show a sizable change due to an $Fe-BaTiO_3$ bonding effect at the $Fe/BaTiO_3$ interface.[9]

In this study, we report on the magnetoelectric effect of Fe dots on ferroelectric $BaTiO_3$ substrates with a view to developing a new approach to control the magnetic domain structures. In fact, we observe a clear variation in the magnetization and magnetic domain structures of Fe dots when the structural phase transition of ferroelectric $BaTiO_3$ occurs at 282 and 186 K. The results provide a promising basis for designing spin electronic devices with a ferromagnetic/ferroelectric heterointerface.

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II. EXPERIMENTS

30-nm-thick Fe dot arrays with different aspect ratios were fabricated on BTO (001) single crystal substrates using ultrahigh vacuum molecular beam epitaxy and electron-beam lithography. The deposition of Fe was done at room temperature. The long axis of all the dots was defined to be along the [100] direction of the BTO substrate. Since the *a*-axis lattice spacing of BTO is 3.99 Å in the tetragonal phase at room temperature, Fe having a lattice constant of 2.87 Å can be grown epitaxially on BTO with a lattice mismatch of 1.4% when the principal axis is rotated by an angle of 45° to each other, i.e., Fe [110] is parallel to BTO [100]. Magnetization of Fe dots was measured in a superconducting quantum interference device magnetometer at temperatures between 300 and 150 K. Magnetic domain structures were observed at room temperature using magnetic force microscopy (MFM). As shown in Fig. 1, the magnetic domain structure of Fe dots changes depending on the aspect ratio of each dot, e.g., the domain structure of an Fe dot with an aspect ratio of 6.5

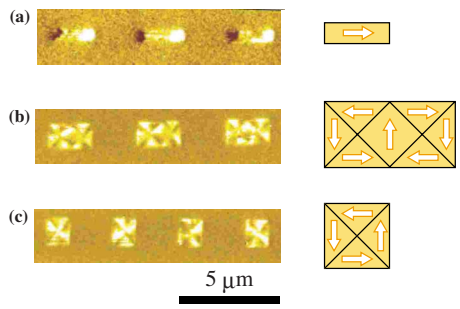

FIG. 1. (Color online) MFM images of epitaxial Fe dots on $BaTiO_3$ with dimensions of (a) 2.0×0.3 μm^2, (b) 1.9×0.9 μm^2, and (c) 1.0×1.0 μm^2 and the corresponding aspect ratios of (a) 6.5, (b) 2, and (c) 1, respectively.

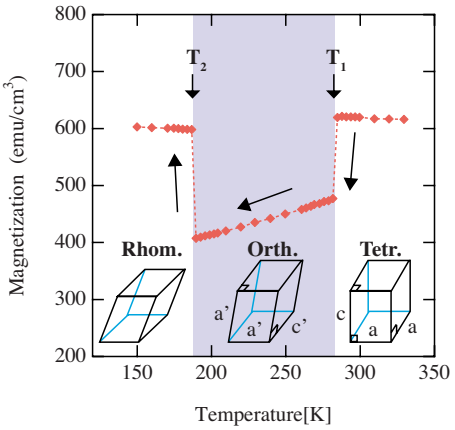

FIG. 2. (Color online) Temperature dependent magnetization of Fe dots/$BaTiO_3$ heterostructures with an aspect ratio of 2 in a magnetic field of 50 Oe along the long axis of Fe dots. Tetr., Orth., and Rhom. denote the tetragonal, orthorhombic, and rhombohedral structures of BTO, respectively.

shows a single domain structure, while those with aspect ratios of 2 and 1 exhibit well-defined symmetrical enclosed magnetic flux structures at room temperature, ensuring the good quality of Fe dots we fabricate. In order to see the magnetostriction effect on the magnetic domain structures of Fe/BTO due to the structural phase transition of BTO, the magnetic domain structures of the Fe dots were again observed *in situ* after being subjected to a thermal cycle between 300 and 150 K.

III. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of in-plane magnetization of Fe dots/BTO with an aspect ratio of 2 shown in Fig. 1(b), measured in a magnetic field of 50 Oe along the long axis of Fe dots. The magnetization exhibits a sudden drop at $T_1=282$ K and again increases up to the almost initial value at $T_2=186$ K with decreasing temperature. The temperatures showing these magnetization jumps clearly correspond to the successive structural phase transitions of BTO from tetragonal to orthorhombic and from orthorhombic to rhombohedral structures, respectively, and the huge change in the magnetization arises from the lattice strain at the Fe/BTO interface associated with the phase transition of BTO. Also, the sign of the change in the magnetization indicates that tensile stress occurs at T_1 while compressive stress occurs at T_2, considering the negative magnetostriction constant $\lambda_S=-1.07\times10^{-5}$ of Fe along $\langle110\rangle$.[10]

Shown in Fig. 3 is the field dependence of in-plane magnetization at 223 and 298 K, where BTO is in the orthorhombic phase and tetragonal phase, respectively. Although the change is not very significant, a variation in the magnetization curves is clearly observed, depending on the crystal structure of the BTO substrate. The results indicate that the strain at the Fe/BTO interface modifies the magnetic anisotropy of Fe due to the magnetostriction effect. It should also be noted that a rapid decrease in the magnetization is seen around the remanent state when the magnetic field is reduced from its magnetic saturation. Such a magnetization reduction is compatible with the remanent enclosed flux magnetic domain structure as depicted in Fig. 1(b), where oppositely oriented magnetic domains cancel out the total magnetization along the magnetic field direction.

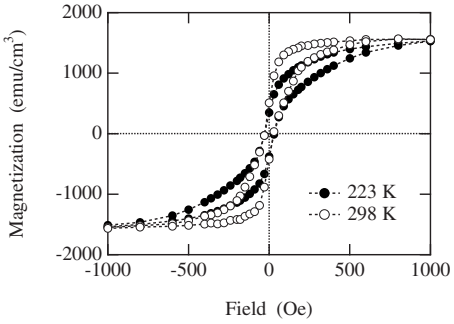

FIG. 3. Magnetic field dependence of the magnetization of Fe dots/$BaTiO_3$ heterostructure with an aspect ratio of 2 measured at 223 and 298 K.

Another interesting magnetic property of the Fe dots/BTO is a significant change in the magnetic domain structures after a thermal cycle between 300 and 150 K. Once the sample experiences the thermal cycle, the magnetic domain structure at room temperature varies from the initial enclosed flux magnetic domain structure to a single-domain-like structure as shown in Fig. 4. In order to clarify the cause of the change in the magnetic domain structure, we observe the x-ray diffraction pattern of a 30-nm-thick Fe film/BTO sample which was fabricated in the same manner as dots samples (see Fig. 5). The (002) peak of the Fe shifts slightly toward higher angles, while (003) and (300) diffractions of the BTO substrate remain at the same position, indicating that the Fe lattice expands in plane by a magnitude of 0.035%. Also, we note that the intensity of the (300) diffraction peak of the BTO substrate increases after the thermal cycle, suggesting that the number of ferroelectric BTO domains lying in plane increases based on the fact that the *c*-axis and *a*-axis lattice spacings of BTO are 4.03 and 3.99 Å at room temperature, respectively.[11] In other words, local switching of the *c* axis of BTO from out of plane to in plane occurs after the thermal cycle and the switching results in an expansion of the in-plane lattice of Fe, accordingly. These combined results of Fe dots and Fe films indicate that a strain at the $Fe/BaTiO_3(001)$ interface induces an additional magnetic anisotropy due to magnetostriction effect.

In order to compare the magnetoelectric effects occur-

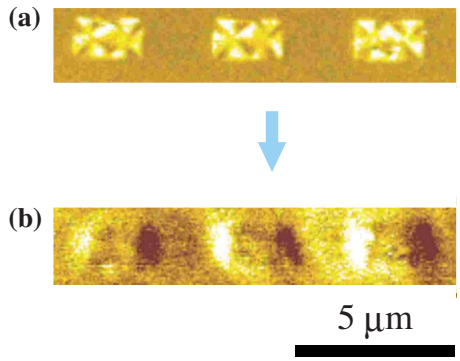

FIG. 4. (Color online) Variation in the magnetic domain structures at room temperature after a thermal cycle between 300 and 150 K.

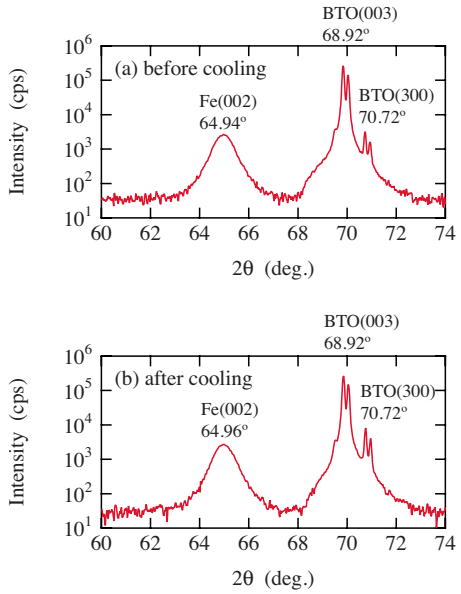

FIG. 5. (Color online) X-ray diffraction patterns of an Fe film/$BaTiO_3$ heterostructure (a) before and (b) after a thermal cycle between 300 and 150 K.

ring at the phase transition and after the thermal cycle, simple calculations of the magnetoelastic energy given below are worthwhile. Since the lattice strain along the a axis is induced by the structural change $\Delta\epsilon=(a'-a)/a=0.6\%$ at T_1,[11] the in-plane interfacial stress σ can be calculated to be 1.3×10^9 N/m^3 using the Young modulus of Fe=2.1×10^{11} J/m^3, estimating a value of the magnetoelastic energy $E_\sigma=3.5\times10^4$ J/m^3. Also, if we use the experimentally obtained in-plane Fe lattice expansion of 0.035% after the thermal cycle, the in-plane stress σ and magnetoelastic energy E_σ are calculated to be 7.4×10^7 N/m^3 and 1.1×10^3 J/m^3, respectively. These values obtained are comparable to the cubic magnetic anisotropy energy constant of Fe ($K_1=4.8\times10^4$ J/m^3), in particular, the value obtained for the magnetoelectric effect at the phase transition from tetragonal to orthorhombic phases. Moreover, our previous work reported on the magnetoelectric effect of Fe dots/$BaTiO_3$(001) heterostructures under the application of local electric voltage using a scanning probe microscope and gave an additional magnetoelastic energy of 1.9×10^3 J/m^3 due to the local voltage application.[3] The value is also comparable to the values obtained in this work, ensuring that both phase transition and local voltage application can provide a promising approach for controlling the magnetic domain structures of Fe dots.

IV. SUMMARY

We have observed a clear variation in the magnetization of Fe dots on $BaTiO_3$(001) substrates at the phase transition temperatures of $BaTiO_3$ from tetragonal to orthorhombic and from orthorhombic to rhombohedral structures. Once the sample is subjected to a thermal cycle between 300 and 150 K, the room-temperature magnetic domain structure changes significantly from an enclosed flux magnetic domain structure into a single-domain-like structure. In order to get a deeper insight into the cause of the variation in the magnetization and magnetic domain structures, we have estimated the magnetoelastic energy for both cases and gave the values comparable to the magnetic anisotropy energy of Fe. These results indicate that Fe/$BaTiO_3$ heterostructures can be a potential candidate for manipulating magnetic domain structures using interfacial strain.

ACKNOWLEDGMENTS

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