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Electric, Magnetic, and Optical Control of Multiferroics

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Abstract

The manipulation of domains by external fields in ferroic materials is of major interest for applications. In multiferroics with strongly coupled magnetic and electric order, however, the magnetoelectric coupling on the level of the domains is largely unexplored. By using optical second harmonic generation microscopy we visualized what happened to the multiferroic domains in the archetypal spin-spiral $TbMnO_3$ under the application of electric, magnetic, and optical fields. The results show the deterministic ferroelectric polarization flop by magnetic field, which leads to the transformation of neutral into nominally charged domain walls, and the reversible optical switching of multiferroic domains. Such magnetoelectric functionalities in spin-driven ferroelectrics may lead to domain wall–based nanoelectronics device and new concept for local control of antiferromagnetism by means of light.

1. Introduction

Multiferroic materials support intertwined ferromagnetic and ferroelectric orders, with the magnetic field capable of controlling the electric order and vice versa. Among them spin-spiral multiferroics exhibit a strong coupling between the electric and magnetic subsystems which is of potential interest for technological applications. Although these systems have been investigated for more than a decade, the magnetoelectric domain evolution under external fields is still largely unknown. Using optical second harmonic generation (SHG) we resolve how electric, magnetic, and optical fields affect the multiferroic domains in the archetypal spin-spiral multiferroic $TbMnO_3$ [1, 2].

In $TbMnO_3$, a cycloidal spin structure emerges below the transition temperature T_C = 27 K and breaks inversion symmetry, giving rise to a spontaneous electric polarization, $\boldsymbol{P}_c$, along the *c* axis [3]. The polarization in $TbMnO_3$ is described as the product of the unit vector $\boldsymbol{e}_{ij}$ that connects neighboring spins at sites *i* and *j*, and their vector chirality, $\boldsymbol{S}_i \times \boldsymbol{S}_j$, according to $\boldsymbol{P} \propto \boldsymbol{e}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$ [4, 5]. Thus, changes in the spin arrangement and the polarization are rigidly connected. For example, a polarization flop ($\boldsymbol{P}_c \rightarrow \boldsymbol{P}_a$) can be triggered by spin reordering in a magnetic field along the *b* axis. This occurs as a first-order phase transition and has been studied for more than a decade. Data documenting the actual reorientation in the form of the evolving magnetoelectric domains are, however, unavailable. It remains an open question to which extent the transition and the associated functionalities are controllable.

2. Experimental Results

Experimental Method

We addressed this fundamental question by tracking the evolution of ferroelectric spin-cycloidal domains under external fields in noninvasive imaging experiments. Imaging was done after zero-field cooling or after electric-field cooling below T_C, using SHG microscopy. SHG, the frequency doubling of a light wave in a material, is an optical process allowed in the leading order only in materials without inversion symmetry. It thus became an established, spatially resolving tool for probing the ferroelectric state. Domains with the spontaneous polarization $\pm\boldsymbol{P}$ emit SHG light waves $\pm\boldsymbol{E}_P(2\omega)$ with a relative 180° phase shift. If a SHG reference light wave is superimposed according to $\boldsymbol{E}_{\mathrm{ref}}(2\omega)\pm\boldsymbol{E}_P(2\omega)$, opposite ferroelectric domain states can be imaged as regions of different brightness. This technique is used throughout our work [1, 2].

Results

In consecutive electric switching cycles, varying multidomain patterns emerge before a single-domain state is obtained (Fig. 1). This includes domain nucleation, forward

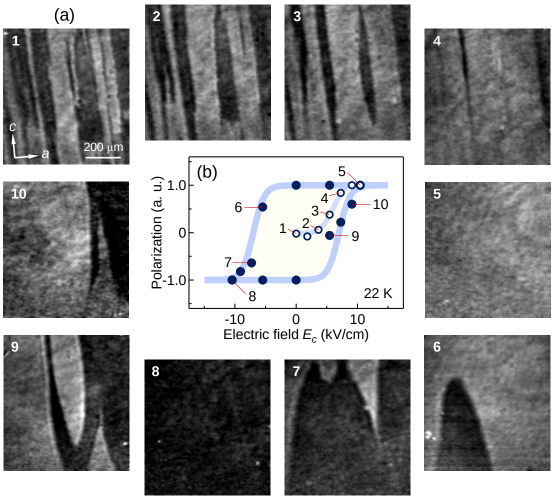

Fig. 1. Electric-field control of multiferroic domains in $TbMnO_3$. (a) Progression of multiferroic domain structure in a cycled electric field E_c along the *c* axis. Bright and dark regions correspond to $+\boldsymbol{P}_c$ and $-\boldsymbol{P}_c$ domains, respectively. (b) Ferroelectric hysteresis loop derived from the areal ratio of $+\boldsymbol{P}_c$ to $-\boldsymbol{P}_c$ domains in SHG images.

growth with walls parallel to the electric field and P_c, and subsequent sideways growth and corroborates the ferroelectric aspect of the domains. This observation reflects that the domain walls can easily move without being pinned by, e.g., structural defects.

In striking contrast to the electric-field response, multidomain patterns persist when the polarization direction is flopped ($P_c \rightarrow P_a$) by applied magnetic fields (Fig. 2). Here, the domain walls, initially parallel to the polarization vector, does not change their shape or position. Thus, a uniform polarization rotation is observed within all domains, which incorporates a transformation of neutral into nominally charged domain walls. Landau-Lifshitz-Gilbert simulations reveal that this behavior is intrinsic and provide first evidence for the scalability of macroscopic magnetoelectric properties onto the level of domains.

In a proof-of-principle experiment we demonstrate that optical switching of multiferroic domains is possible (Fig. 3). We reverse the multiferroic order parameter in spin-spiral $\mathrm{TbMnO_3}$ repeatedly, using light pulses of two different colors. Switching depends on a unique relation between the wavelength of the light, its optical absorption and the electric polarization field induced by the spin-spiral order of $\mathrm{TbMnO_3}$. We also demonstrate sequential laser-controlled writing and erasure of multiferroic (antiferromagnetic spin-spiral) domains. Opto-magnetism is thus complemented by an important degree of freedom, namely local control of antiferromagnetism by means of light.

3. Conclusions

We have investigated the multiferroic domain properties in $\mathrm{TbMnO_3}$ by nonlinear optical techniques. We succeeded in visualizing the multiferroic domains in TbMnO3 for the first time and full electric-field switching and deterministic polarization flop by magnetic field were observed. Furthermore, we could optically switch the multiferroic domains without any bias fields. These results may help in the development of future multiferroic devides.

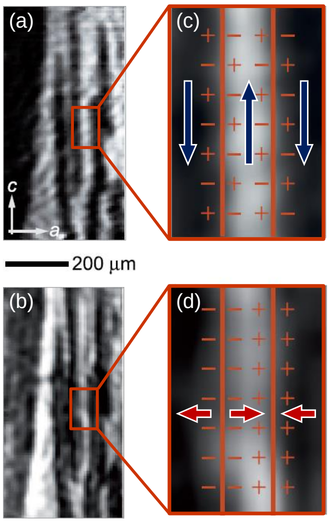

Fig. 2. Ferroelectric polarization flop by magnetic field in $\mathrm{TbMnO_3}$. Multiferroic domain structure in P_c phase (a) and P_a phas (b). The magnified regions show that side-by-side domain walls in P_c phase (c) are changed to head-to-head/tail-to-tail walls in P_a phase (d) by the polarization flop, leading to nominally charged domain walls.

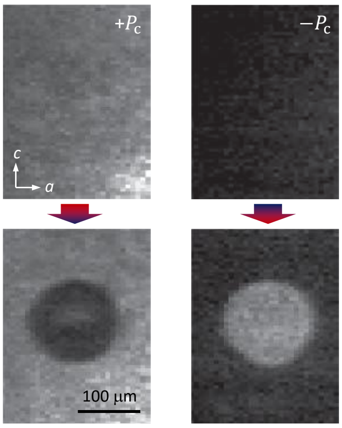

Fig. 3. Optical switching of multiferroic domains in $\mathrm{TbMnO_3}$. Single +P_c (-P_c) domain is locally switched to –P_c (+P_c) domain by light irradiation. (Left) $+P_c \rightarrow -P_c$. (Right) $-P_c \rightarrow +P_c$.

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Appendix

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