# Enhanced Optical Magnetoelectric Effect in a Patterned Polar Ferrimagnet

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A simple method to dramatically enhance the optical magnetoelectric (ME) effect, i.e., nonreciprocal directional birefringence, is proposed and demonstrated for a polar ferrimagnet $GaFeO_3$ as a typical example. We patterned a simple grating with a period of 4 $\mu$m on a surface of $GaFeO_3$ crystal and used the diffracted light as a probe. The optical ME modulation signal for the Bragg spot of the order $n = 1$ becomes gigantic in the photon energy 1–4 eV and reaches 1–2% of the bare diffracted light intensity in a magnetic field of 500 Oe. This is amplified by more than 3 orders of magnitude compared to that for the reflection of bulk $GaFeO_3$. Fabricating a photonic crystal will make it possible to lead the way for the practical use of the optical ME effect.

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The magnetoelectric (ME) effect is a cross-correlation phenomenon that the polarization $P$ is induced by the magnetic field $H$ or inversely the magnetization $M$ by the electric field $E$. Recent revival of the study of ME effects has stimulated further interest in ferroelectric magnets, termed multiferroics, showing the strong coupling between ferroelectricity and magnetism [1]. Multiferroics lack in both space inversion and time reversal symmetries due to their ferroelectric (polar) and ferromagnetic natures, respectively. Light is known to individually interact with each broken symmetry, for example, the second-order nonlinear optical effect in ferroelectrics and the magneto-optical Kerr effect (MOKE) in ferromagnets, due to the presence of the finite component of the second-order nonlinear susceptibility and of the off-diagonal dielectric tensor, respectively. Simultaneous symmetry breaking also produces the pronounced ME effect at optical frequencies. This has been known experimentally [2–4] and theoretically [5–7] since the 1970s as the nonreciprocal birefringence or dichroism, which is referred to as magnetochiral (MCh) effect for chiral media or as optical ME effect for polar magnets. The optical ME effect is characterized by whether the propagation vector $k$ of light is parallel or antiparallel to the toroidal moment $T$ defined as the outer product of $P$ and $M$. (The MCh effect is, on the other hand, with respect to $k$ parallel or antiparallel to $M$.) The effect emerges as a change of transmission or reflection and its magnitude is proportional to the magnitude of $T$ projected along the $k$ direction, $\hat{k} \cdot T$ ($\hat{k} \equiv \frac{k}{|k|}$) [5,7]. Although there are a variety of candidate materials showing the optical ME effect [8,9] or MCh effect [10,11], their magnitudes are too weak even under the combination of high $E$ and $H$ (in the order of 10 kOe). Furthermore, their amplitudes in the reflection geometry tend to be smaller than that in the transmission geometry. These prevent the optical ME effect or MCh effect from possible applications.

Here we propose a simple method to dramatically enhance the optical ME effect. With use of a simple diffraction grating structure on a surface of a multiferroic $GaFeO_3$ crystal, we have succeeded in observing the pronounced optical ME modulation signal up to 1–2% of the bare diffracted light intensity in $H$ of 500 Oe, which exceeds 3 orders of magnitude compared to the case of the simple reflection. In contrast to usual experimental conditions to observe the optical ME effect, where $k$ is parallel to $T$, we adopt the experimental setup so that the direction of $T$ is parallel to the reciprocal lattice vector $G$ of the grating, in which an interference effect would occur when the grating period is close to the wavelength of light (Fig. 1). Therefore, the propagation vector $k_{out}$ of the diffracted light in the reflection geometry lies within the plane of scattering.

The polar (not ferroelectric but pyroelectric) ferrimagnet $GaFeO_3$ we used here exhibits both broken symmetries of space inversion and time reversal; Fe ions displace from the centrosymmetric position to produce the spontaneous polarization $P_s$ along the $b$ axis and the ferrimagnetic transition takes place around 205 K, yielding the spontaneous $M$ along the $c$ axis [see Fig. 3(b)]. This makes $GaFeO_3$ suitable for the detection of the optical ME effect, since $P_s$ is perpendicular to the easy axis of $M$ and thus $T$ ($\propto P_s \times M$) appears along the $a$ axis of the orthorhombic lattice (left panel of Fig. 1). Furthermore, $GaFeO_3$ is known to show the relatively large optical ME modulation signal in $H$ of 500 Oe at optical ($\sim 10^{-3}$) [12] and at x-ray ($\sim 10^{-4}$) [13] frequencies when the transmission geometry is adopted [see also Fig. 4(a)].

A single crystal of $GaFeO_3$ was grown by a floating zone method [14]. We patterned a simple grating structure along

 

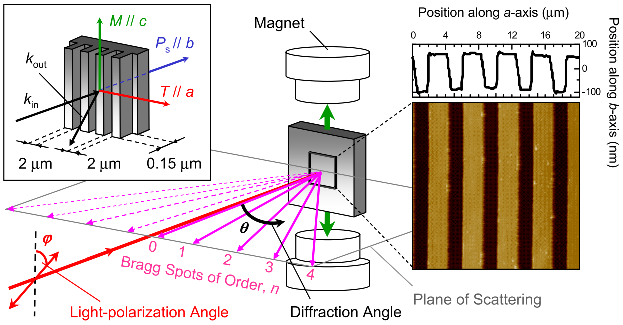


FIG. 1 (color online). Schematic illustration of our experimental setup [15]. Experimental configurations and each crystallographic axis are shown in the left panel. AFM image of the fabricated multiferroic grating and the cross-sectional depth profile along the $a$ axis are displayed in the right panel.

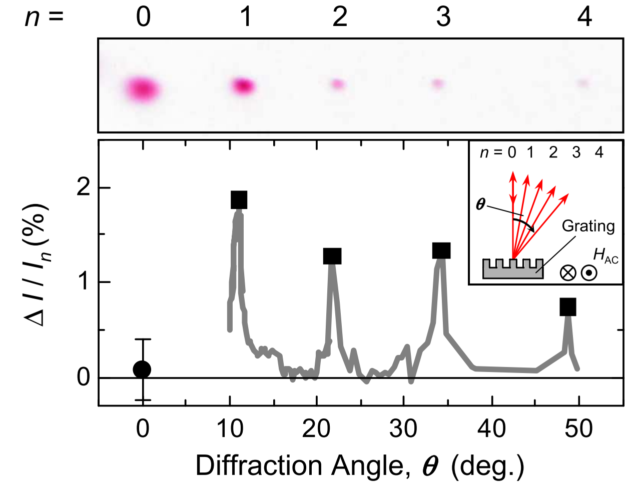


FIG. 2 (color online). Diffraction angle $\theta$ dependence of the modulation $\Delta I/I_n$ ($\lambda = 785$ nm) for $E^{\omega} \parallel H_{ac}$ (500 Oe and 3 Hz), measured at 50 K. Bragg spots of $n$ up to 4, which were projected on the screen, are also shown in the upper panel. Inset shows the schematic illustration of the trajectory of diffraction.

the $c$ axis on the (010) surface. The grating period $d$, the width, and the depth of the groove were set to be 4 $\mu$m, 2 $\mu$m, and 0.15 $\mu$m, respectively, (left panel of Fig. 1), which were confirmed by an atomic force microscopic (AFM) image (right panel of Fig. 1). This can act as a Bragg diffraction grating at optical frequencies. We employed Bragg diffraction magneto-optical setup in a transverse geometry, where $H$ was applied perpendicular to the plane of scattering. Figure 1 depicts the schematic layout for the diffraction experiments. $k$ was fixed parallel to $P_s$, while $T$ was set to lie within the plane of scattering. This configuration sets $k_{out}$ within the plane of scattering and $G$ parallel to $T$. If not stated explicitly, the linearly polarized cw laser with wavelength $\lambda$ of 785 nm was used as a light source. To obtain the intrinsic optical ME modulation signal, a sinusoidal magnetic field $H_{ac}$ of 500 Oe with an alternating frequency $f$ of 2–10 Hz was applied along the $c$ axis. The modulation of the diffracted light intensity $\Delta I$ was lock-in detected and was normalized by the average diffracted light intensity $I_n$ for Bragg spots of the order $n$. The change in $\Delta I$ with the reversal of a static magnetic field $H_{dc}$ of 500–2000 Oe was also detected and was normalized by $I_n$ without $H_{dc}$. We confirmed that $H$-modulation technique gives essentially identical results with those obtained by using $H_{dc}$ [see Fig. 3(a)]. Further details of our experimental setups are deposited elsewhere [15].

One can visually discern the reflected and diffracted spots when the cw laser was irradiated on the multiferroic grating. In the upper panel of Fig. 2, we display the spatial profile of the reflected and diffracted light intensity up to $n = 4$. Diffraction angle $\theta$ of the respective spots satisfies the Bragg diffraction law given by $d \sin\theta = n\lambda$. Figure 2 shows $\Delta I/I_n$ as a function of $\theta$. $\Delta I/I_n$ was measured by rotating the detector within the plane of scattering so that the detected position was changed arbitrarily while keeping the distance from the multiferroic grating constant. The electric field $E^{\omega}$ of the cw laser was set parallel to $H_{ac}$, where no transverse MOKE is expected to appear. We can see an oscillation of $\Delta I/I_n$ with $n$, which seems to have even-odd parity of $n$. Such an oscillation structure frequently emerges as a consequence of the interference effect in simple ferromagnetic gratings on various substrates [16,17]. This can be phenomenologically explained by the Fourier transformation of the grating structure with a geometrical phase shift depending on the depth of the groove [17]. Noticeable signatures in Fig. 2 are no detectable signal of $\Delta I/I_n$ for $n = 0$ indicated by a closed circle within the capability of our detection system ($\sim 10^{-4}$), as well as the appreciable magnitude of $\Delta I/I_n$ of the order of $10^{-2}$ for respective $n$ indicated by closed squares. The maximum value of $\Delta I/I_n$ is observed for $n = 1$ and its magnitude reaches as large as 2%.

To verify that the observed gigantic signal should come from the optical ME effect, the dependence of the modulation signal on the magnitude of $M$ and the polarity of $P_s$ were examined. We measured the temperature dependence of $\Delta I/I_{n=1}$ [closed circles in Fig. 3(b)] and for $M$ along the $c$ axis (a solid line). The temperature-dependent behavior of $\Delta I/I_{n=1}$ is nearly consistent with that of $M$. The slight difference of the onset temperatures between the both quantities is perhaps due to the degradation of the surface $M$ by the pattern fabrication process. We display in Fig. 3(a) $\Delta I/I_{n=1}$ as a function of the light-polarization angle $\varphi$ (circles). The configurations are schematically shown in the right panel of the figure. $\Delta I/I_{n=1}$ is nearly independent of $\varphi$, which is characteristic of the optical ME effect [6,7], excluding other possible $\varphi$-dependent effects such as MOKE as the origin of the signal. To confirm the effect of the reversal of $P_s$ on the modulation signal, we also fabricated another piece of multiferroic grating on the *counter surface* of the crystal. Namely, we cut the crystal ingot to produce a pair of (010) surfaces on the both of which the identical grating structures were patterned. This

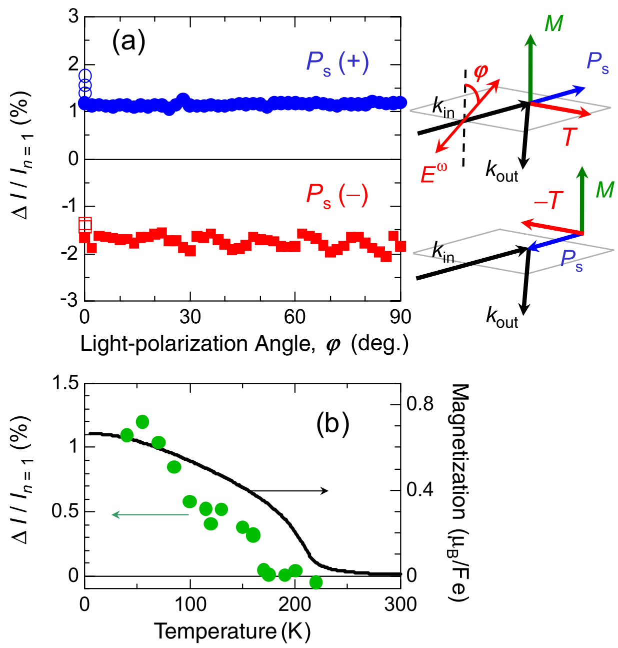


FIG. 3 (color online). (a) Light-polarization angle $\varphi$ dependence of the modulation $\Delta I/I_{n=1}$ (closed symbols) for $E^{\omega} \parallel H_{ac}$ (500 Oe and 2 Hz) at 70 K. $\Delta I/I_{n=1}$ (open symbols) observed by reversing $H_{dc}$ of 500 Oe for $E^{\omega} \parallel H_{dc}$ is also shown. Corresponding experimental configurations are shown in the respective right panels. (b) Temperature dependence of $\Delta I/I_{n=1}$ (closed circles) for $E^{\omega} \parallel H_{dc}$ with the reversal of $H_{dc}$ of 2000 Oe and $M$ (solid line) along the magnetic easy axis ($c$ axis) of bulk $GaFeO_3$ in $H_{dc}$ of 2000 Oe [15].

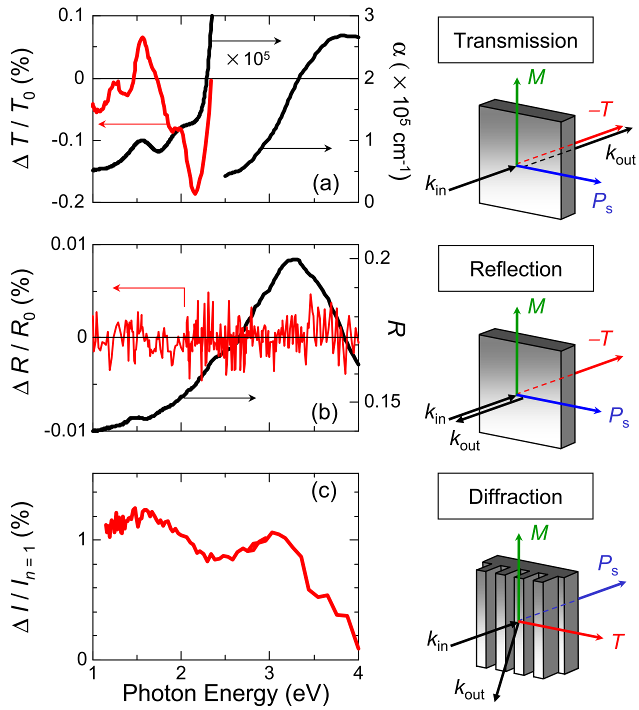


FIG. 4 (color online). Optical ME spectra for $E^{\omega} \parallel H_{ac}$ in various geometries; (a) transmission, (b) reflection, and (c) diffraction for Bragg spot of $n = 1$. Corresponding experimental configurations are also shown in the respective right panels. (a) Modulation in transmission $\Delta T/T_0$ with 30 $\mu$m thick of crystal plate in $H_{ac}$ of 800 Oe at $f$ of 5 Hz and at 10 K. (b) Modulation in reflection $\Delta R/R_0$ in $H_{ac}$ of 800 Oe at $f$ of 5 Hz and at 10 K. (c) Modulation in intensity $\Delta I/I_{n=1}$ in $H_{ac}$ of 500 Oe at $f$ of 3 Hz and at 70 K. Static spectra of (a) $\alpha$ and (b) $R$ for $E^{\omega} \parallel c$ (right scale) are also shown [15].

produces the reversal of the direction of $T$ because of the reversal of $P_s$, when the direction of $M$ and the detector position can be set unchanged [right panel of Fig. 3(a)]. Therefore, the sign reversal of $\Delta I/I_{n=1}$ is expected to show up in the case of the optical ME effect, while it would not be in the case of MOKE. We indeed confirmed such a unique characteristic of the optical ME effect, as indicated by squares in Fig. 3(a). The phase of $\Delta I/I_{n=1}$ changes by $\pi$ with each another (the absolute value of $\Delta I/I_{n=1}$ is slightly different possibly due to variance of the experimental sample condition). Combined with the results shown in Figs. 3(a) and 3(b), $\Delta I/I_{n=1}$ is proved to depend on both the directions of $P_s$ and $M$. We also confirmed that the sign of $\Delta I/I_n$ is reversed by changing the detector position between $n = 1$ and $n = -1$. All these results are the firm evidences that the observed gigantic signal arises from the optical ME effect. Therefore, the negligible value of $\Delta I/I_{n=0}$ (Fig. 2) is reasonable since $k_{out}$ in this case is perpendicular to $T$, where the optical ME effect is not allowed. This also ensures the normal incidence of the cw laser and excludes the possible violation of the strict relations, $k_{in} \perp M$ and $k_{in} \parallel P_s$ in our experimental setup.

The optical ME signal (1–2%) of the multiferroic grating presented here is by far larger than the previously reported values in the reflection geometry; $\sim 10^{-4}$ for $Cr_2O_3$ after applying $E$ of 2 kV/cm and $H$ of 2 kOe [3], $\sim 10^{-4}$ for $LiFe_5O_8$ in $H$ of 10 kOe [4], $\sim 10^{-10}$ for limonene in $H$ of 1 kOe [11], and $\sim 10^{-3}$ for the chevron shaped patterns made by $Ni_{80}Fe_{20}$ in $H$ of 500 Oe [18]. The observed optical ME signal in the present reflection (diffraction) geometry even exceeds the value in the transmission or emission geometry, e.g., $\sim 10^{-3}$ for $Eu(tfc)_3$ (tfc = trifluoroacetyl-camphorato) in $H$ of 10 kOe [10]. Here, let us compare the optical ME spectra of $GaFeO_3$ in transmission [12], reflection, and diffraction geometries. Relationships among $k_{in}$, $k_{out}$, $P_s$, $M$, and $T$ are shown in the panels of Fig. 4. Figure 4(c) displays $\Delta I/I_{n=1}(\omega)$. The light from a xenon discharged lamp through a monochromator was used. The detector was rotated so as to satisfy the Bragg diffraction law with $\lambda$ [15]. There are two spectral structures around 1.5 and 3 eV as can be clearly seen in the spectra (right scale) of the absorption coefficient $\alpha$ and the reflectivity $R$ of bulk $GaFeO_3$ shown in Figs. 4(a) and 4(b), respectively [15]. These peak structures correspond to the $d$-$d$ transitions from the high-spin ground state of $Fe^{3+}$ ($^6A_1$) to the $^4T_1$ state [19] and the charge-transfer (CT) transition from O $2p$ to Fe $3d$ states, respectively. The appreciable magnitude of $\Delta I/I_{n=1}$ continues up to the CT transition region $\sim 3$ eV and tends to decrease above 3 eV, while keeping the same sign over the measured photon energy. This is in sharp contrast to the case of the transmission geometry: Fig. 4(a) shows the relative modulation transmission spectrum $\Delta T/T_0(\omega)$ of bulk $GaFeO_3$ (left scale). We can see multiple changes of $\Delta T/T_0$ including the sign change near the *d*-*d* transition, which can be considered as a result of the interference effect between electric and magnetic dipole transitions [12]. On the contrary, $\Delta I/I_{n=1}(\omega)$ shows the enhancement below 2 eV, although the electric dipole moment is considerably weak, as can be seen in $\alpha(\omega)$ [Fig. 4(a)]. This may be ascribed to the Fabry-Perot–like constructive interference of reflections, where the light propagates along $T$ (see upper panel of AFM image of Fig. 1) and repeatedly reflects between the side wall of the groove and that of the next groove. However, quantitative estimate of the optical ME spectrum is not available even in the case of the simple reflection or transmission and thus its microscopic origin is left to be elucidated. Finally, we present in Fig. 4(b) relative modulation reflection spectrum $\Delta R/R_0(\omega)$ of bulk $GaFeO_3$ (left scale). No optical ME modulation signal can be discerned with our detection limit ($\sim10^{-5}$) when the reflection geometry was adopted. This gives the lower bound of the enhancement factor of the optical ME signal using the simple grating; the optical ME modulation signal is amplified at least by 3 orders of magnitude when Bragg spot is used as the probe [Fig. 4(c)].

Recently, Sawada and Nagaosa numerically solved the Maxwell's equation in the case of the multiferroic grating that we proposed here [20]. They derived the enhancement factor ($\sim10^2$–$10^3$) of the optical ME effect within the framework of the classical optics, whose magnitude is consistent with our results. The present systematic experiments as well as the support from the calculation can ensure the usefulness of multiferroic grating to strongly enhance the optical ME effect and the potential of such a "multiferroic photonic crystal" in future application.

In summary, gigantic enhancement of the optical ME effect has been demonstrated for a simple grating structure on a surface of a polar ferrimagnet $GaFeO_3$ crystal. The observed optical ME modulation signal is found to be 1–2% of the bare diffracted light intensity in $H$ of 500 Oe, which is amplified by 3 orders of magnitude or more compared to the case of bulk $GaFeO_3$. This simple method that we proposed and demonstrated here can also be applied to other multiferroics and may provide potential applications of the optical ME effect.

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