Theoretically predicted picosecond optical switching of spin chirality in multiferroics

Masahito Mochizuki1,2 and Naoto Nagaosa1,3

1Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan
2Multiferroics Project, ERATO, Japan Science and Technology Agency (JST), Tokyo 113-8656, Japan
3Cross-Correlated Materials Research Group (CMRG) and Correlated Electron Research Group (CERG), RIKEN-ASI, Saitama 351-0198, Japan

We show theoretically with an accurate spin Hamiltonian describing the multiferroic Mn perovskites that the application of the picosecond optical pulse with a terahertz frequency can switch the spin chirality through intensely exciting the electromagnons. There are four states with different spin chiralities, i.e. clockwise and counterclockwise ab/bc-plane spin spirals, and by tuning the strength, shape and length of the pulse, the switching among these states can be controlled at will. Dynamical pattern formation during the switching is also discussed.

PACS numbers: 75.80.+q, 75.85.+t, 77.80.Fm, 75.10.Hk

Chirality, i.e., the right- and left-handedness of structure, is one of the fundamental concepts penetrating through the whole of science. In solids, electron spins sometimes form a chiral order, which offers an opportunity to manipulate the chirality by external parameters. This issue is of vital importance in spintronics, which aims at the electric control of spins by an electric current or electric field (E). Multiferroics provides us an ideal system for this purpose, in which the spin chirality is directly related to the electric polarization. In the spin-current model, two mutually canted spins \( S_i \) and \( S_j \) generate the polarization \( p_{ij} \) as

\[
p_{ij} \propto e_{ij} \times (S_i \times S_j),
\]

(1)

with \( e_{ij} \) being the vector connecting \( i \)th and \( j \)th sites. Here the vector product \( S_i \times S_j \) is the spin chirality, which characterizes a direction of the spin rotation. In multiferroic Mn perovskites such as TbMnO\(_3\) and DyMnO\(_3\), the Mn spins are rotating within the bc plane to form a cycloid (bc-plane spiral) propagating along the +b direction (Pbnm setting) as shown in Fig. 1(a) \[3, 4\].

Theoretically predicted picosecond optical switching of spin chirality in multiferroics

 FIG. 1: (Color online) (a) Spin configuration, spin chirality \( S_i \times S_{i+b} \) and ferroelectric polarization \( P \) in the clockwise bc-plane spin spiral (upper panel) and those in the counterclockwise one (lower panel). (b) Modulation of the in-plane ferromagnetic exchanges under \( E_{//} \pm a \). Upper (lower) signs in front of \( \Delta J_{ab} \) correspond to the modulations under \( E_{//} + a (E_{//} - a) \). (c) Calculated electromagnon spectrum. Inset shows the experimental spectrum for TbMnO\(_3\) \[13\].

In this Letter, we theoretically propose a picosecond optical switching of the spin chirality in TbMnO\(_3\) as a representative material. However, the mechanism is rather general and would be ubiquitous in spiral magnets. It is found that the oscillating \( E \) component of the optical pulse activates collective rotations of the spin-spiral planes via magnetoelastic (ME) coupling, and their inertial motions result in the chirality reversal or flop. By tuning the strength, shape and length of the pulse, the spin chirality is shown to be controlled at will.

When the electric polarization is driven by the spin order, it is naturally expected that the collective excitation of spins (magnon) has an infrared activity \[7, 8\]. Indeed, strong optical absorptions were experimentally observed in \( R \)MnO\(_3\) (\( R=\)Tb, Dy, Eu\(_{1-x}Y_x\), etc) at THz frequencies, and they were ascribed to magnons activated by the \( E \) component of the light, i.e., electromagnons \[7, 9–14\]. In the early stage, the corresponding magnon modes were interpreted as rotation of the spin-spiral plane with oscillating \( p_{ij} \) in Eq. (1) \[8\]. However, this interpretation contradicts the experimental observation about the selection rule in terms of the light polarization \[11, 12\]. Afterwards, it turned out that the polarization \( p_{ij} \) due to the conventional magnetostirction, i.e.,

\[
p_{ij} = \pi_{ij}(S_i, S_j)
\]

(2)

is relevant to the infrared-activity of magnons in \( R \)MnO\(_3\) \[13–17\]. Here the vector \( \pi_{ij} \) is nonzero because of the orthorhombic lattice distortion without inversion symmetry at the center of the Mn-O-Mn bond. The puzzling electromagnon optical spectrum with two specific peaks was successfully explained by this mechanism \[17\].

Under this circumstance, the photo-induced phenom-
ena become a challenging issue. Since \( p_{ij} \) in Eq. (2) does not require the spin-orbit interaction, its magnitude is much larger than that of \( p_{ij} \) in Eq. (1), which enables the intense and fast optical excitation of magnons. This offers a unique opportunity to study the nonlinear dynamics of the spin system. In addition, the light can locally activate or modify the spin structure with a squeezed light spot in contrast to the magnetic field.

We are now ready to attack such phenomena in \( RMnO_3 \) theoretically ahead of experiments for the following reasons. First, we know that the optical pulse activates mostly the spins only via the ME coupling, which allows us to neglect electronic excitations at much higher energies (>1.5 eV). Second, we have an accurate spin Hamiltonian, which describes competitions among various phases in \( RMnO_3 \) \cite{18}, so that the optical switchings among them and dynamics after the light irradiation can be simulated in a reliable way.

We employ a classical Heisenberg model on a cubic lattice, which contains not only the frustrating spin exchange (\( H_{\text{ex}} \)) but also the single-ion spin anisotropy (\( H_{\text{dia}}^D \) and \( H_{\text{dia}}^E \)), Dzyaloshinskii-Moriya (DM) interaction (\( H_{\text{DM}} \)) and biquadratic interaction (\( H_{\text{biq}} \)) as,

\[
H = H_{\text{ex}} + H_{\text{dia}}^D + H_{\text{dia}}^E + H_{\text{DM}} + H_{\text{biq}}
\]

\[
= \sum_{<i,j>} J_{ij} S_i \cdot S_j + D \sum_i S_i^2 + E \sum_i (-1)^i S_i^x S_i^y + \sum_{<i,j>} \sum_{a,b} d_{ij} \cdot (S_i \times S_j) - B_{\text{biq}} \sum_{<i,j>} (S_i \cdot S_j)^2.
\]  

(3)

The frustration between ferromagnetic (FM) exchange \( J_{ab} \) and antiferromagnetic (AFM) exchange \( J_{c} \) results in the in-plane spiral spin orders, while the interplane AFM exchange \( J_{e} \) causes their staggered stacking. The DM vectors \( d_{ij} \) are expressed using five DM parameters, \( \alpha_{ab}, \beta_{ab}, \gamma_{ab}, \alpha_{c} \) and \( \beta_{c} \), as given in Ref. \cite{19}. Crucial roles of the biquadratic interaction in \( RMnO_3 \) were uncovered in recent theoretical studies \cite{21, 22}. For more detail of the model, see Ref. \cite{17}. We adopt the following parameters: \( J_{ab} = -0.74, J_{b} = 0.64, J_{c} = 1.0 \) (\( \alpha_{ab}, \beta_{ab}, \gamma_{ab} \))=(0.1, 0.1, 0.14), \( (\alpha_{c}, \beta_{c}) \)=(0.48, 0.1), \( D = 0.2 \), \( E = 0.25 \), and \( B_{\text{biq}} = 0.025 \), where the energy unit is meV.

This parameter set gives the bc-plane spin spiral with a wave number \( q_0 = 0.3 \pi \) at low temperatures, which resembles the spin structure in \( TbMnO_3 \) \( (q_0 = 0.29 \pi) \) \cite{23, 24}.

We trace dynamics of the Mn spins by numerically solving the Landau-Lifshitz-Gilbert equation using the fourth-order Runge-Kutta method. We derive an effective magnetic field \( H_{\text{eff}}^S \) acting on the spin \( S_i \) from the Hamiltonian \( \mathcal{H} \) as \( H_{\text{eff}}^S = -\partial \mathcal{H} / \partial S_i \). Considering the observed reduced Mn moment \( |S_i| = 1.4 \), we set for the norm of the spin vector \( |S_i| \) 40×40×6 in size with the periodic boundary condition. For the ME coupling, we consider \(-E \cdot p_{ij} \) with \( p_{ij} \) given in Eq. (2) \cite{13, 17}. This coupling effectively modulates the in-plane FM exchanges from \( J_{ab} S_i \cdot S_j \) to \( (J_{ab} - E \cdot \pi_{ij}) S_i \cdot S_j \). Consequently, the applied \( E|\pm a \) modulates the spin exchanges as shown in Fig. 1(b). Here \( |\pi_{ij}^a| \) is calculated to be \( 3.5 \times 10^{-50} \mu C m \) from the lattice parameters \cite{22} and the observed ferroelectric polarization \( P(\approx 5000 \mu C/cm^2) \) for \( RMnO_3 \) with an up-up-down-down spin order \cite{23}. This means that \( E_a = 1 MV/cm \) induces the modulation \( |\Delta J_{ab}| = |E_a | \pi_{ij}^a| \approx 0.022 \) meV.

The infrared-absorption spectrum is calculated as the response to a weak \( \delta \)-function pulse. (For technical detail, see Ref. \cite{17}). The calculated spectrum is displayed in Fig. 1(c), which has two peaks at \( \omega = 0.94 \) THz and 2.1 THz, and reproduces well the experimental spectrum of \( TbMnO_3 \). The Gilbert-damping coefficient \( \alpha_G \) is chosen to be 0.1 so as to reproduce the observed peak width, which guarantees the under-damped spin oscillations.

Now we theoretically demonstrate switching of the spin chirality by the optical pulse. There are two kinds of bc-plane spirals with different spin chiralities, i.e., CW and CCW ones. Their spin chiralities \( C \) point in the \(-a \) and \(+a \) directions (\( C \parallel -a \) and \( C \parallel +a \)) so that they are referred to as \( bc_- \) and \( bc_+ \), respectively. Here the chirality \( C \) is defined as a sum of the local contributions \( C_{i,i+x} = S_i \times S_{i+x} \) and \( C_{i,i+y} = S_i \times S_{i+y} \) as

\[
C = 1/2 \sum_i (C_{i,i+x} + C_{i,i+y}) / S_i^2.
\]

Note that \( C_{i,i+x} \) is zero because of the AFM stacking in z-direction. The CW and CCW ab-plane spirals are also possible although they are slightly higher in energy than the bc-plane ones without external fields. They have \( C \parallel -c \) and \( C \parallel +c \), and thus are referred to as \( ab_- \) and \( ab_+ \), respectively.

Starting with \( bc_- \) with \( C \parallel -a \), we apply an intense pulse of \( E = |E_0 (t, 0, 0) \) along the \( a \) axis, where

\[
E_0(t) = -E_0 \sin \omega t \exp\left[-\frac{(t-t_0)^2}{2\sigma^2}\right],
\]

with \( t_0 = 1 \) psec. Here the frequency \( \omega \) is fixed at 2.1 THz, which corresponds to the higher-energy electromagnon peak, while the full width of the half maximum for the Gaussian envelope, \( 2\sqrt{2\ln 2} \sigma \), is taken to be 0.5 psec (see inset of Fig. 2). In Fig. 2 we display calculated time evolutions of the \( a \)-, \( b \)- and \( c \)-axis components of \( C \). When \( E_0 = +14 MV/cm \), we find a reversal of \( C \) from \( bc_- \) (\( C \parallel -a \)) to \( bc_+ \) (\( C \parallel +c \)) as shown in Fig. 2(a). This reversal takes place via \( ab_- \) with \( C \parallel +c \) and \( C \parallel -c \). In addition, as shown in Fig. 2(b), when we apply a slightly weaker pulse of \( E_0 = +13 MV/cm \), a chirality flop occurs from \( bc_- \) (\( C \parallel -a \)) to \( ab_+ \) (\( C \parallel +c \)). These switchings occur very fast, typically within 5-7 psec.

To understand these phenomena, we first consider energies of the four chirality states, i.e., \( ab_- \) and \( bc_- \). When \( E = 0 \), the \( ab_- \) and \( bc_- \) are degenerate, and are higher in energy than the ground-state bc-plane spirals. Application of \( E|\pm a \) lifts this degeneracy, and the \( ab_+ \) (\( ab_- \)) becomes the lowest energy under the strong \( E|\pm a \).
FIG. 2: Time evolutions of the spin chirality $C=(C_a, C_b, C_c)$ after application of the pulse with (a) $E_0=+14$ MV/cm and (b) $E_0=+13$ MV/cm. Fig.2(a) shows the chirality-reversal from $bc_-$ ($C_a<0$) to $bc_+$ ($C_a>0$), while Fig.2(b) shows the chirality flop from $bc_-$ ($C_\parallel \approx -\alpha$) to $ab_+$ (C$_\parallel +\alpha$). Insets show spin states before or after applying the pulse, and time profile of the applied pulse $E_a(t)$.

FIG. 3: Modulations of spin turn angles (thin red arrows) and the local spin chiralities $\pm \Delta C_{i,j}$ under $E\parallel a$ for (a) $ab_+$ and (b) $ab_-$ states. (c) Arrangement of the $c$-axis components of DM vectors $\pm d_{i,j}^c$.

\( (E\parallel -a) \). This can be understood as follows. As shown in Figs. 3(a) and 3(b), the modified in-plane FM exchanges, $J_{ab} \pm \Delta J_{ab}$, under $E\parallel a$ cause changes in the spin turn angles (thin red arrows) and hence staggered modulations of the local spin chiralities as $C_{i,j} \pm \Delta C_{i,j}$ (thick brown arrows). Under $E\parallel +a$, the modulations $\pm \Delta C_{i,j}$ in the $ab_+$ ($ab_-$) are always antiparallel (parallel) to the staggered $c$-axis components of the DM vectors $\pm d_{i,j}^c$ [blue arrows in Fig. 3(c)]. Since the DM coupling favors the antiparallel configurations of $\pm d_{i,j}^c$ and $\pm C_{i,j}$, the energy decreases (increases) in the $ab_+$ ($ab_-$). Note that this mechanism is distinct from the direct coupling between $E$ and $p_{ij}$ given in Eq. (1), which is much weaker in the THz-frequency regime. On the other hand, the energies of $bc_\pm$ are not affected by $E$ because the $a$-axis components of the DM vectors are alternately stacked, by which the DM energy always cancels out.

In Fig. 3(a), we illustrate schematic time evolution of the potential as a function of $\theta$ during the chirality-reversal process. For time profile of the applied pulse with $E_0>0$, see the inset of Fig. 3(b). Here $\theta$ is the angle between the chirality $C$ and the $a$ axis. At $t=t_1$, the system is located in the minimum at $\theta=180^\circ$ ($bc_-$). When $E_a(t)>0$ as at $t=t_2$, $\theta=90^\circ$ ($ab_+$) becomes a new energy minimum, so that the chirality $C$ starts rotating or the angle $\theta$ starts decreasing towards this minimum. Importantly the chirality does not stop its rotation at $\theta=90^\circ$ immediately, but passes through that minimum or oscillates around it because of the inertial force. The DM interaction and the single-ion anisotropy originating from the spin-orbit interaction make the rotation of spin-spiral plane massive, resulting in its inertial motion. Then the $E_a(t)$ becomes negative as at $t=t_3$, which makes $ab_+$ ($\theta=90^\circ$) the highest in energy. Consequently the system starts falling into the minimum at $\theta=0^\circ$ ($bc_+$). At last ($t=t_4$), the chirality reversal is completed being settled in $bc_+$ ($\theta=0^\circ$).

On the other hand, the chirality flop for $E_0=+13$ MV/cm is a rather subtle process. If the system is within the domain of metastability of $\theta=90^\circ$ when $E_a(t)$ becomes almost zero at $t=t_4$, the system can be trapped in the local minimum of $ab_+$. Since the $ab_+$ are metastable, the system should decay into the ground-state $bc_+$ or $bc_-$ eventually due to thermal fluctuations.

This ultrafast chirality switching is distinct from the
conventional polarization reversal, and is accompanied by the dynamical spatial-pattern formation, i.e., dynamical stripes of chirality domains, through the nonlinear photoexcitation of electromagnons. In Figs. 5(a)-(f), we show the calculated real-time dynamics of the local chiralities $C_{i,j}$ on the Mn-O plane for the chirality-reversal process. Starting from $bc_-$ (blue) at $t=0$, the optical pulse generates $ab_+$ (green) domains as at $t=1.2$ psec. Subsequently stripes of $bc_+$ (red) and $bc_-$ (blue) domains emerge after the pulse ends at $t=2$ psec, and the chiralities $C_{i,j}$ oscillate in each domain between $\theta_{ij}=0^\circ$ and $\theta_{ij}=180^\circ$ as seen in $2<t(\text{psec})<3.4$. Among the spirally ordered spins, those directing (nearly) along the propagation vector cannot flip and become nodes of the spin oscillations to form the chirality domains. At last ($t=6$ psec) the system gradually gets settled in the $bc_+$ state.

Finally we discuss conditions for the chirality switching. First, the switching occurs only at a frequency of the higher-energy electromagnon resonance ($\omega \sim 2.1$ THz in the present case), and does not occur at the lower-energy peak ($\omega \sim 1$ THz). Second, we need a rather large peak height of the pulse, $|E_0| \gtrsim 10$ MV/cm, in the present simulation. Experimentally maximum peak height exceeding 100 MV/cm is available for 10-72 THz, but below 3 THz, it reaches only $\sim 1$ MV/cm at present. We expect that the optical pulse with $|E_0| \gtrsim 10$ MV/cm at $\sim 2$ THz will be realized in the near future. Besides, the threshold value of $|E_0|$ can be reduced if we properly choose the target materials: For example, solid solutions Tb$_{1-x}$Gd$_x$MnO$_3$ locating near the boundary between the $ab$- and $bc$-plane spiral phases are promising candidates. Importantly there are optimal ranges of the electric strength $|E_0|$, and a larger $|E_0|$ cannot necessarily induce the switching. This can be understood as follows. To achieve the chirality reversal, for instance, from $\theta=180^\circ$ ($bc_-$) to $\theta=0^\circ$ ($bc_+$), the chirality vector oscillating around the energy minimum at $\theta=90^\circ$ ($ab_+$) should be in the range $0^\circ < \theta < 90^\circ$ when $E$ is reversed from $E_0(t)>0$ to $E_0(t)<0$ in order to fall into the another minimum at $\theta=0^\circ$ ($bc_+$) instead of $\theta=180^\circ$ ($bc_-$).

This means that we need to adjust depth of the energy minimum of $ab_+$ at $t=t_2$ by tuning the strength of $E_0$ in order to synchronize the timing between the chirality oscillation and the reversal of $E$. Therefore, the switching processes show highly nonlinear behaviors with respect to strength and shape of the pulse. In addition, if we adopt a negative $E_0$ in Eq. (4), the lowest-lying state at $t=t_2$ becomes $ab_-$ with $\theta=270^\circ$. Then the chirality reversal occurs via $ab_-$ instead of $ab_+$. The chirality flops to $ab_-$ from $bc_+$ become also possible for a slightly weaker $|E_0|$. Relationships between the switching processes and the sign of $E_0$ are summarized in Fig. 4(b).

To summarize, we have theoretically studied the ultrafast optical switching of spin chirality by exciting the electromagnons in the multiferroic Mn perovskite. We have revealed that the oscillating $E$ component of the light activates the collective rotations of the spin-spiral planes with a THz frequency via the ME coupling, and their inertial motions result in chirality reversal or flop. By tuning strength, shape and length of the pulse, the spin chirality is shown to be controlled at will.

The authors are grateful to N. Kida, Y. Tokura, N. Furukawa, R. Shimano and I. Kezmarki for discussions. This work was supported by Grant-in-Aid (Grants No. 22740214, No. 2144053, No. 17105002, No. 19048015, and No. 19048008) and G-COE Program “Physical Sciences Frontier” from MEXT Japan, and Funding Program for World-Leading Innovative R&D on Science and Technology (FIRST Program) from JSPS.

[1] Y. Tokura, J. Magn. Magn. Mater. 310, 1145 (2007); S.-W. Cheong and M. Mostovoy, Nat. Mater. 6, 13 (2007); D. I. Khomskii, J. Magn. Magn. Mater. 306, 1 (2006).
[2] T. Kimura et al., Nature (London) 426, 55 (2003).
[3] H. Katsura, N. Nagaosa, and A. V. Balatsky, Phys. Rev. Lett. 95, 057205 (2005).
[4] M. Mostovoy, Phys. Rev. Lett. 96, 067601 (2006).
[5] M. Kenzelmann et al., Phys. Rev. Lett. 95, 087206 (2005).
[6] Y. Yamashiki et al., Phys. Rev. Lett. 98, 147204 (2007).
[7] A. Pimenov et al., Nat. Phys. 2, 97 (2006).
[8] H. Katsura, A. V. Balatsky, and N. Nagaosa, Phys. Rev. Lett. 98, 027203 (2007).
[9] A. Pimenov et al., Phys. Rev. B 74, 100403(R) (2006).
[10] A. Pimenov et al., Phys. Rev. B 77, 014438 (2008).
[11] N. Kida et al., Phys. Rev. B 78, 104414 (2008).
[12] N. Kida et al., J. Phys. Soc. Jpn. 77, 123704 (2008).
[13] Y. Takahashi et al., Phys. Rev. Lett. 101, 187201 (2008).
[14] N. Kida et al., J. Opt. Soc. Am. B 26, A35 (2009).
[15] R. Valdez Aguilar et al., Phys. Rev. Lett. 102, 047203 (2009).
[16] S. Miyahara and N. Furukawa, arXiv:0811.4082.
[17] M. Mochizuki, N. Furukawa, and N. Nagaosa, Phys. Rev. Lett. 104, 177206 (2010).
[18] M. Mochizuki, and N. Furukawa, J. Phys. Soc. Jpn. 78, 053704 (2009); Phys. Rev. B 80, 134416 (2009).
[19] I. Solovyev, N. Hamada, and K. Terakura, Phys. Rev. Lett. 76, 4825 (1996).
[20] T. A. Kaplan, Phys. Rev. B 80, 012407 (2009).
[21] T. Arima et al., Phys. Rev. Lett. 96, 097202 (2006).
[22] J. A. Alonso, M. J. Martínez-Lope, M. T. Casais, and M. T. Fernández-Díaz, Inorg. Chem. 39, 917 (2000).
[23] S. Ishiwata et al., Phys. Rev. B 81, 100411(R) (2010).
[24] A. Sell, A. Leitenstorfer, and R. Huber, Optics Lett. 33, 2767 (2008).
[25] T. Goto et al., Phys. Rev. B 72, 220403(R) (2005).