Microwave magnetochiral effect in Cu$_2$OSeO$_3$

Masahito Mochizuki$^{1,2}$

$^1$Department of Physics and Mathematics, Aoyama Gakuin University, Sagamihara, Kanagawa 229-8558, Japan
$^2$PRESTO, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan

We theoretically predict that in a multiferroic chiral magnet Cu$_2$OSeO$_3$, resonant magnetic excitations are coupled to collective oscillation of electric polarization, and thereby attain simultaneous activity to ac magnetic field and ac electric field. Because of interference between these magnetic and electric activation processes, this material hosts gigantic magnetochiral dichroism on microwaves, that is, the directional dichroism at gigahertz frequencies in Faraday geometry. The absorption intensity of microwave differs by as much as $\sim$30% depending on whether its propagation direction is parallel or antiparallel to the external magnetic field.

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Collective excitations of spins in magnets, so-called magnons or spin waves, can be activated not only via a direct process with ac magnetic field $H^\omega$ coupled to magnetizations but also via an electric excitation by ac electric field $E^\omega$ coupled to charge degrees of freedom. When the magnon or spin-wave modes have simultaneous activity to the $H^\omega$ and $E^\omega$ components of electromagnetic waves, interference between the two activation processes, that is, the magnetically activating and the electrically activating processes, gives rise to peculiar optical and/or microwave phenomena, so-called optical ME effect. One of the most important examples is the directional dichroism, that is, oppositely propagating electromagnetic waves exhibit different absorptions.

Multiferroic materials with concurrent magnetic and ferroelectric orders provide an opportunity to realize the electric-dipole active magnons (so-called electromagnons), and thus the optical ME effect via the magnetoelectric coupling. Indeed observations of the directional dichroism have been reported for several multiferroic materials such as Ba$_2$CoGe$_2$O$_7$[17–19], RMnO$_3$ ($R$=rare-earth ions) [20, 21], and CuFe$_{1-x}$Ga$_x$O$_2$[22], in which nontrivial spin orders induce the ferroelectric polarization via the relativistic spin-orbit interaction. In these materials, the optical ME effect is observed at the electromagnon resonance frequencies in the terahertz (THz) regime.

The directional dichroism is observed also at higher frequencies, i.e., x-ray and visible-light regimes in several polar magnets, which is caused by electron transitions among the spin-orbit multiplets [23–29]. However, observations of the effect at gigahertz (GHz) frequencies are quite limited and the effect observed so far is very tiny whose difference in absorption intensity is only 2.5% at most [32], while the directional dichroism at GHz frequencies is anticipated for application to microwave devices [30]. This is because most of the well-known multiferroic materials based on simple spiral or antiferromagnetic spin structures with short-period modulation tend to have relatively large spin-wave gaps of several meV, which inevitably results in rather high resonance frequencies in THz regime. To achieve the microwave ME effect, long-period magnetic textures with tiny spin-wave gap should be examined, and slowly modulating magnetic structures induced by the Dzyaloshinskii-Moriya interaction (DMI) are promising for this purpose. Indeed, the nonreciprocal directional dichroism of microwaves in Voigt geometry was theoretically predicted for the DMI-induced skyrmion phase [31].

In this Letter, we theoretically predict that electromagnon excitations in magnetically ordered phases of Cu$_2$OSeO$_3$ exhibits unprecedentedly large magnetochiral dichroism at GHz frequencies, that is, the microwave directional dichroism in Faraday geometry. In the presence of $H$, the conical spin phase or the field-polarized ferrimagnetic phase emerges in the bulk samples depending on the strength of $H$. When $H$ is applied in a certain direction, and a microwave is irradiated parallel or antiparallel to $H$, the absorption intensities for the oppositely propagating microwaves differ by as much as 30%. Such a huge directional dichroism at microwave frequencies has never been observed in a single material. This effect is traced back to the resonantly enhanced magnetoelectric coupling, and therefore essentially distinct in microscopic mechanism from the traditional microwave non-reciprocal device based on microwave polarization, potentially leading to a unique microwave device.

The crystal and magnetic structures of Cu$_2$OSeO$_3$ consist of a network of tetrahedra composed of four Cu$^{2+}$ (S=1/2) ions at their apexes as shown in Figs. 1(a) and (b). In each tetrahedron, three-up and one-down collinear spin configuration is realized below $T_c$~58 K [33–34]. This four-spin assembly can be regarded as a magnetic unit, and is described by a classical magnetization vector $m$, whose norm $m$ is unity. We employ a classical Heisenberg model on a cubic lattice to describe the magnetism in a bulk specimen of Cu$_2$OSeO$_3$ [35–37], which contains the ferromagnetic-exchange interaction and the Dzyaloshinskii-Moriya interaction among the effective magnetizations $m_i$ and the Zeeman coupling to
the spin textures considered here are slowly varying in space, and thus the coupling to the background crystal structure is significantly weak. This justifies our treatment with a spin model on the cubic lattice after the coarse graining of magnetizations.

The presence or absence of the ferroelectric polarization \( \mathbf{P} \) and, if any, its direction can be known from the symmetry consideration \cite{38, 40}. The crystal structure of Cu$_2$OSeO$_4$ belongs to the chiral cubic P2$_1$3 point group, which has three-fold rotation axes, 3, along (111), and two-fold screw axis, 2$_1$, along (100) as shown in Fig. 1(e). This crystal symmetry is not polar, and thus there exists no spontaneous \( \mathbf{P} \). Although the conical and the ferromagnetic spin states are not polar, either, combination of the crystal and the magnetic symmetries renders the system polar, and allows the emergence of \( \mathbf{P} \). As shown in Fig. 1(f), the emergence of \( \mathbf{P} \parallel [001] \) perpendicular to the net magnetization \( \mathbf{M} \parallel \mathbf{H} \) is expected for the conical and the ferromagnetic states formed under \( \mathbf{H} \parallel [110] \) since only the 2$_1$ axis along [001] remains as a symmetry axis. On the other hand, the emergence of \( \mathbf{P} \) is forbidden under \( \mathbf{H} \parallel [010] \) since three 2$_1$ axes survive as shown in Fig. 1(g).

Microscopically the local polarization \( \mathbf{p}_i \) at the \( i \)th tetrahedron is given using the magnetization components \( m_{ia}, m_{ib}, \) and \( m_{ic} \) as,

\[
\mathbf{p}_i = (p_{ia}, p_{ib}, p_{ic}) = \lambda (m_{ib}m_{ic}, m_{ic}m_{ia}, m_{ia}m_{ib}) .
\]

The net magnetization \( \mathbf{M} \) and the ferroelectric polarization \( \mathbf{P} \) are calculated by sums of the local contributions as \( \mathbf{M} = \frac{1}{N} \sum_{i=1}^{N} \mathbf{m}_i \) and \( \mathbf{P} = \frac{1}{N} \sum_{i=1}^{N} \mathbf{p}_i \), respectively, where the index \( i \) runs over the Cu-ion tetrahedra, \( N \) is the number of the tetrahedra, and \( V = 1.76 \times 10^{-28} \) m$^3$ is the volume per tetrahedron. The coupling constant \( \lambda \) is evaluated as \( \lambda = 5.64 \times 10^{-27} \) µCm from the experimental data \cite{31}. Figure 1(h) shows calculated net polarization \( \mathbf{P} \) in the field-polarized ferromagnetic state when \( \mathbf{H} \) is applied within the c-plane as a function of the angle \( \theta \) between \( \mathbf{H} \) and the \( b \) axis (see the inset). We find that the positive (negative) \( \mathbf{P} \) emerges along [001] when \( \theta \) is negative (positive).

According to Fig. 1(h), one realizes that oscillation of \( \mathbf{M} \) (\( \Delta \mathbf{M}^r \parallel [001] \)) induces oscillation of \( \mathbf{P} \) (\( \Delta \mathbf{P}^r \parallel [001] \)) when \( \mathbf{M} \parallel \mathbf{H} \parallel [010] \) [see Figs. 2(a)-(c)], and conversely \( \Delta \mathbf{P}^r \parallel [001] \) induces \( \Delta \mathbf{M}^r \parallel [001] \) via the magnetoelectric coupling. This means that both \( \mathbf{H}^r \parallel [100] \) and \( \mathbf{E}^r \parallel [001] \) components of microwave can activate the coupled oscillation of \( \mathbf{M} \) and \( \mathbf{P} \) (Note that the response time of \( \mathbf{M} \) (\( \mathbf{P} \)) against the change of \( \mathbf{P} \) (\( \mathbf{M} \)) is governed by the electron transitions among the orbital multiplets, and thus is much shorter than the typical time scale of the oscillations). To see this, we calculate the following dynamical susceptibilities by numerically solving the Landau-Lifshitz-Gilbert (LLG) equation using the fourth-order

\[
\mathcal{H}_0 = -J \sum_{<i,j>} \mathbf{m}_i \cdot \mathbf{m}_j - D \sum_{i} \mathbf{m}_i \times \mathbf{m}_i + g\mu_B H \sum_i \mathbf{m}_i ,
\]
FIG. 2: (color online). (a)-(c) In the presence of net magnetization $M_t^f$ under $H\|\langle 010 \rangle$, oscillating magnetization component $\Delta M^\omega\|\langle 100 \rangle$ is accompanied by the oscillating polarization component $\Delta P^\omega\|\langle 001 \rangle$. (d) Configuration of microwave $H^\omega$ and $E^\omega$ components, for which the magnetochiral dichroism occurs under $H\|\langle 010 \rangle$: $k^\omega\|\pm H$, $H^\omega\|\langle 100 \rangle$ and $E^\omega\|\langle 001 \rangle$.

Runge-Kutta method:

$\chi^{\mu\nu}_{\alpha\beta}(\omega) = \frac{\Delta M_{\omega}}{\mu_0 \Delta H_{\beta}}$ magnetic susceptibility,

$\chi^{ee}_{\alpha\beta}(\omega) = \frac{\Delta P_{\omega}}{\epsilon_0 \Delta E_{\beta}}$ dielectric susceptibility,

$\chi^{em}_{\alpha\beta}(\omega) = \frac{\Delta P_{\omega}}{\sqrt{\epsilon_0 \mu_0 \Delta H_{\beta}}} \frac{\Delta E_{\beta}}{}$ magnetoelectric susceptibility,

$\chi^{me}_{\alpha\beta}(\omega) = \sqrt{\frac{\mu_0}{\epsilon_0}} \frac{\Delta M_{\omega}}{\Delta E_{\beta}}$ electromagnetic susceptibility.

Because of the symmetry, the relation $\chi^{em}_{\alpha\beta}(\omega) = \chi^{me}_{\beta\alpha}(\omega)$ holds. The LLG equation is given by

$$\frac{d\mathbf{m}_i}{dt} = -\mathbf{m}_i \times H_i^{\text{eff}} + \frac{\alpha_G}{m} \mathbf{m}_i \times \frac{d\mathbf{m}_i}{dt},$$

where $\alpha_G (= 0.04)$ is the Gilbert-damping coefficient. The effective field $H_i^{\text{eff}}$ is calculated from the Hamiltonian $H' = \mathcal{H}_0 + H'(t)$ as $H_i^{\text{eff}} = -\partial \mathcal{H}/\partial \mathbf{m}_i$. The first term $\mathcal{H}_0$ is the model Hamiltonian given by Eq. (1). The perturbation term $H'(t)$ represents a short rectangular pulse of magnetic field $\Delta H(t)$ or electric field $\Delta E(t)$, which are given, respectively, by

$$H'(t) = -g \mu_0 \sum_i \Delta H(t) \cdot \mathbf{m}_i$$

and

$$H'(t) = -\sum_i \Delta E(t) \cdot \mathbf{p}_i.$$
The expression of the complex refractive index \( N(\omega) \) is derived by solving the Fourier-formed Maxwell’s equation as [17],

\[
N(\omega) = \frac{c}{\omega} k^{\omega} \sim \sqrt{\kappa_{cc}^{\omega} + \chi_{cc}^{\omega}(\omega)} \left[ \mu_{aa}^{\omega} + \chi_{aa}^{em}(\omega) \right] + sgn(Re k^{\omega}) \left[ \chi_{ac}^{me}(\omega) + \chi_{ca}^{em}(\omega) \right]/2, \tag{6}
\]

for \( k^{\omega} = k^{\omega} \mathbf{b} \parallel [010], \ H^{\omega} \parallel [100] \parallel \mathbf{a} \) and \( \mathbf{E}^{\omega} \parallel [001] \parallel \mathbf{c} \). The absorption coefficient \( \alpha(\omega) \) is related to \( N(\omega) \) as,

\[
\alpha(\omega) = \frac{2\omega \kappa(\omega)}{c} = \frac{2\omega}{c} \text{Im} N(\omega), \tag{7}
\]

and thus attains the directional dependence via the sign of \( \text{Re} k^{\omega} \). Here \( \kappa(\omega) = \text{Im} N(\omega) \) is the extinction coefficient.

Figures 3(a) and (b) display calculated frequency-dependence of \( \alpha_{+} \) and \( \alpha_{-} \) in the conical phase and the ferromagnetic phase, respectively, for several values of \( H \), where \( \alpha_{+} \) and \( \alpha_{-} \) are the absorption coefficients for microwaves with parallel and antiparallel to \( H^{\omega} \parallel [010] \), respectively. In the calculation, we take \( c^2=8 \) and \( \mu_{zz}^{\omega}=1 \) according to the experimental data [12, 43]. We find that the microwave absorption is resonantly enhanced at the field-frequency of the electromagnon excitation, and there exists significant difference between \( \alpha_{+} \) and \( \alpha_{-} \).

Calculated magnitude of the directional dichroism \( \Delta \alpha/\alpha_{-} \) where \( \Delta \alpha = \alpha_{+} - \alpha_{-} \) is plotted in Fig. 4. This quantity is governed by the amplitude of ferroelectric polarization \( P \), which is nearly proportional to the square of the net magnetization \( M^2 \). The dichroism increases in the conical state with the growth of \( P \) and \( M \) as the magnetic field increases. In contrast, once the system enters the ferromagnetic state, the saturated \( M \) gives nearly constant directional dichroism although there still exists slight field-dependence due to the field-dependent resonant frequency via \( \omega \) in Eq. (7). The directional dichroism is enhanced at the phase boundary between the conical and the ferromagnetic phases, and its magnitude reaches as much as \(~30\%\). Calculated electromagnon resonance frequency plotted in Fig. 4 shows decreasing behavior in the conical state, while increasing behavior in the ferromagnetic state as the magnetic field increases. This behavior is in good agreement with the experimental observations [32, 41] and coincides with analytical formula of the spin-wave gap given in Ref. [44].

In summary, we have theoretically predicted that magnetically ordered phases in the chiral multiferroics \( \text{Cu}_2\text{OSeO}_3 \) host gigantic microwave magnetochiral dichroism. This phenomenon results from interference between the magnetic and electric activation processes of electromagnons with GHz resonance frequencies. It has been demonstrated that long-period magnetic structures in the chiral multiferroics without inversion symmetry can host gigantic dynamical magnetoelectric phenomena at GHz regime. In order to further enhance the effect, search for novel chiral multiferroics with larger \( P \) or stronger magnetoelectric coupling is needed. For example, chiral multiferroics based on the inverse Dzyaloshinskii-Moriya mechanism [2] as an origin of its \( P \) is worth trying to search because this mechanism tends to induce large \( P \) relative to the spin-dependent metal-ligand hybridization mechanism in \( \text{Cu}_2\text{OSeO}_3 \). Metamaterials, thin films, and synthetic nanomaterials are an-
other promising candidate to realize enhanced microwave ME effects through artificially designing large magneto-electric susceptibilities and intense electromagnon resonances [45, 50].

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