Electrically driven spin excitation at THz frequencies in \textit{bc} and \textit{ab} spiral spin phases of perovskite manganites

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Abstract. Here we overview the low-energy (1–10 meV) electrodynamics of spin excitations in a family of ferroelectric perovskite manganites, DyMnO\textsubscript{3} and Gd\textsubscript{0.7}Tb\textsubscript{0.3}MnO\textsubscript{3}. By using THz time-domain spectroscopy, we show the compelling evidence that the electric-dipole active spin excitation around 2 meV emerges only along the \textit{a}-axis, irrespective of the direction of the spiral spin plane (\textit{bc} or \textit{ab}). We sum up the general features of the spin excitations, as observed at THz frequencies in ferroelectric perovskite manganites, by taking DyMnO\textsubscript{3} and Gd\textsubscript{0.7}Tb\textsubscript{0.3}MnO\textsubscript{3} as typical examples.

1. Introduction

Recently, there is revival interest in the study of the magnetoelectric (ME) effect, in which the ferroelectric polarization \(P_s\) can be induced by the magnetic field \(H\) or inversely the magnetization can be induced by the electric field [1]. This boom is stimulated by the discovery of the ferroelectricity in perovskite manganites, general formula \(RMnO_3\) (\(R = \text{Tb, Dy, Gd, and their mixed crystals}\)) [2,3]. Below the ferroelectric (FE) transition temperature \(T_c\), the ferroelectricity emerges along the \(c\)-axis. In this FE phase, the spiral spin order was experimentally confirmed [4], being consistent with the prediction based on the spin-current model [5]; the spiral spin structure characterized by the vector chirality \((S_i \times S_j)\) can produce the \(P_s\) along the direction perpendicular to the modulation wavevector (\(b\)-axis) and within the spiral spin plane, i.e., \(P_s \parallel c\) in \(bc\) spiral and \(P_s \parallel a\) in \(ab\) spiral. This is simply formulated by \(P_s \propto e_{ij} \times (S_i \times S_j)\), where \(e_{ij}\) is the unit vector connecting the nearest spins, \(S_i\) and \(S_j\). As a manifestation of this strong coupling between magnetism and ferroelectricity, the direction of \(P_s\) can

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be controlled from \( c \) to \( a \)-axis (\( P \), flop by 90 deg.) by applying \( H \) for TbMnO\(_3\) and DyMnO\(_3\) [2,3]. This phase transition can be regarded as the \( H \) control of the spiral spin plane from \( bc \) to \( ab \).

Such a strong ME coupling would also produce the intriguing low-lying spin excitation, as theoretically anticipated to exist since 1970s [6]. In R\( \text{MnO}_3 \), there are experimental [7] and theoretical [8] arguments on the possibility of the presence of the electrically driven spin excitation at THz frequencies, called \textit{electromagnon}. Optical spectroscopy has provided the useful information for the electrodynamics in a family of R\( \text{MnO}_3 \), i.e., TbMnO\(_3\) [7,9], DyMnO\(_3\) [10], GdMnO\(_3\) [7,11], Gd\(_{0.7}\)Tb\(_{0.3}\)MnO\(_3\) [12], and Eu\(_{1-x}\)Y\(_x\)MnO\(_3\) [13,14], revealing that the electric-dipole active spin excitation indeed emerges at THz frequencies below the optical phonon frequencies of the perovskite structure.

In this conference proceeding, we show our comprehensive optical investigations on the spin excitation in a variety of the spin ordered phases (including FE \( bc \) and \( ab \) spiral spin ordered phases), as realized in DyMnO\(_3\) and Gd\(_{0.7}\)Tb\(_{0.3}\)MnO\(_3\).

2. Experimental

We adopted the THz time-domain spectroscopy in transmission geometry to extract the complex optical constants \( n \). Since the spin excitation in R\( \text{MnO}_3 \) can be driven by electric field \( E^o \) or magnetic field \( H^o \) of light, we used the quantity of \( \varepsilon\mu(\omega)(=n^2) \), where \( \varepsilon \) is the complex dielectric constant and \( \mu \) the complex magnetic permeability. The validity of this approach and detailed experimental setup can be found in Ref. [10]. All samples used here were grown by the floating-zone method. The available \( ac \), \( ab \), and \( bc \) surfaces of the crystals were cut from the ingot and were polished to the thickness of 100–800 \( \mu \)m. We measured the complete set of the light-polarization (both \( E^o \) and \( H^o \)) dependence using \( ac \), \( ab \), and \( bc \) surfaces of the crystals.

3. Results and discussion

First, we overview a variety of the spin excitations at THz frequencies in the FE \( bc \) spiral spin phase of DyMnO\(_3\) [10]. In DyMnO\(_3\), the FE \( bc \) spiral spin order is developed below \( T_s=19 \) K in zero \( H \), followed by the paraelectric (PE) collinear spin order along the \( b \)-axis below \( T_c=42 \) K [3]. Among a family of R\( \text{MnO}_3 \), DyMnO\(_3\) exhibits the largest ME response, as exemplified by the gigantic change of \( \varepsilon \) (\( \sim \)500\% at 10 kHz) during the \( P \), flop [3]. Figure 1(a) displays the imaginary part of \( \varepsilon\mu \) spectrum (Im[\( \varepsilon\mu \))], measured at 10 K for \( E^o||a \) and \( H^o||c \) (we used the \( ac \) surface of the crystal). There is a noticeable broad continuum-like absorption in Im[\( \varepsilon\mu \)] spectrum in the measured energy range (1–10 meV); it seems to consist of low- (\( \sim \)2 meV) and high- (\( \sim \)6 meV) lying peak structures. The position of the low-lying peak structure is nearly identical to those observed for other R\( \text{MnO}_3 \) [7-14]. The magnitude of Im[\( \varepsilon\mu \)] reaches the maximum about 10 in the FE \( bc \) spiral spin phase. Apparently, the low-lying peak structure can be discerned even in the PE collinear spin phase below \( T_s \), as shown in Im[\( \varepsilon\mu \)] spectrum at 28 K [Fig. 1(a)], indicating that the observed absorption is magnetic in origin. Such a broad absorption was confirmed to become active for \( E^o||a \) on the basis of the measurements using the \( ac \) and \( ab \) surfaces of the crystal [10]. Therefore, it can be assigned to the electric-dipole active spin excitation, as in the same cases of other R\( \text{MnO}_3 \) [7-14]. On the contrary, the single sharp peak structure is identified around 2 meV in Im[\( \varepsilon\mu \)] spectrum at 14 K [Fig. 1(b)] for \( E^o||c \) and \( H^o||a \). Its position nearly matches the position of the low-lying peak structure for \( E^o||a \). The magnitude of Im[\( \varepsilon\mu \)] was estimated to be about 0.4. Based on the light-polarization dependence using \( ac \) and \( ab \) surfaces of crystals [10], this peak structure can be interpreted as the antiferromagnetic resonance (AFMR) of Mn ions driven by \( H^o||a \). Concerning the broad continuum-like spin excitation for \( E^o||a \), we confirmed the negligible effect of \( H \); it survives even in the FE \( ab \) spiral spin plane induced by applying \( H \) along the \( b \)-axis [10]. This apparently contradicts the interpretation of the inelastic neutron scattering spectra in the \( bc \) spiral spin phase of TbMnO\(_3\) [15], in which one of the low-energy magnon band around 2 meV at \( k=0 \) can be attributed to the rotation mode of the spiral spin plane. For TbMnO\(_3\), DyMnO\(_3\), and GdMnO\(_3\), the relatively high \( H \) is required to induce the FE \( ab \) spiral spin phase, which restricts the possible optical configurations to further clarify the nature of the spin excitations.
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In Gd$_{0.7}$Tb$_{0.3}$MnO$_3$, we next introduced here, is a rare example showing the FE $ab$ spiral spin order in zero $H$. In Gd$_{0.7}$Tb$_{0.3}$MnO$_3$, the PE collinear spin order evolves below $T_N$ of 42 K along the $b$-axis, as in the same manner to other RMnO$_3$. Between 15 K and 24 K, the $ab$ spiral spin order ($P_a||a$) was recently confirmed by polarized neutron scattering experiments [16]. Furthermore, the modulation wavevector $q_b$ of Mn ions was found to be 0.25, which is identical to $q_b$ of the FE $ab$ spiral spin phase of TbMnO$_3$ induced by $H$ [17]. As a typical example, we show in Fig. 1(c) the Im[$\varepsilon_{\mu}$] spectrum of Gd$_{0.7}$Tb$_{0.3}$MnO$_3$ in the thermally induced FE $ab$ spiral spin phase, measured at 17 K for $E^a||a$ and $H^a||a$, respectively. We also plot the Im[$\varepsilon_{\mu}$] spectrum for $E^a||a$ in the paraelectric collinear spin phase.

Based on experimental results described here, we can extract the general features of the spin excitation in a family of RMnO$_3$. First, the electric-dipole active spin excitation becomes active only along the $a$-axis, irrespective of the direction of the spiral spin plane ($bc$ or $ab$). This can clearly exclude the possibility that the observed spin excitation can be ascribed to the rotation mode of the spiral spin plane, as formally suggested by theoretical [8] and experimental [15,18] arguments. Accordingly, the presence of the electric-dipole active spin excitation along the $a$-axis has been reported for Eu$_{1-x}$Y$_x$MnO$_3$ [12,13] even though $P_a$ in this system appears along the $a$-axis. Second, the observed excitation seems to be regarded as the broad continuum-like band, rather than the single peak structure previously clarified in TbMnO$_3$ [7] and GdMnO$_3$ [7,11]. Third, the AFMR of Mn ions driven by $H^a||a$ stimulation shows up at the nearly same position of the electric-dipole active spin excitation for $E^a||a$. This AFMR can be ascribed to one of the lower branch of the magnon band around 2 meV in FE $bc$ and $ab$ spiral spin phases of TbMnO$_3$ [15,18]. Contrary to the case of AFMR, the broad spectral shape of the electric-dipole active spin excitation for $E^a||a$ cannot be simply explained by assuming the $k=0$ one magnon alone. Although the more elaborated theory is needed, one plausible scenario is the model of the two-magnon excitation, as partly discussed in Refs. [9,10,12].
4. Summary
By using THz time-domain spectroscopy, we clarified the low-energy electrodynamics of spin excitations in $bc$ and $ab$ spiral spin phases of $RMnO_3$, by taking DyMnO$_3$ and Gd$_{0.7}$Tb$_{0.3}$MnO$_3$ as typical examples. Based on the measurements of the light-polarization dependence, we firmly confirmed that the electric-dipole active spin excitation in $RMnO_3$ emerges only along the $a$-axis, irrespective of the direction of the spiral spin plane.

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