

Magnetolectric resonance with electromagnons in a perovskite helimagnet

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Maxwell's equations describe the interrelation between temporally changing electric (E) and magnetic (H) fields in a given medium. In materials that exhibit relativistic spin-orbit interactions, we also expect their polarization (P) and magnetization (M) to be dynamically coupled. This in turn could enable greater control over the cross-coupling between the electric and magnetic fields of light in the development of photonic devices¹. Such magnetolectric phenomena are expected to be enhanced within materials that support electromagnons—fundamental excitations that exhibit both electric and magnetic dipole moments. Here we report the discovery of electromagnons in the perovskite ($\text{Eu, Y})\text{MnO}_3$, which arise from fluctuations in the spontaneous polarization generated by cycloidal spin order^{2–5}. The resulting dynamical M - P cross-coupling causes the material to exhibit colossal directional dichroism—a difference in the absorption of light propagating in opposite directions—at the resonance frequency (sub-THz) associated with these excitations.

The magnetolectric effect, which is the cross-correlation between electricity and magnetism, is one of the central subjects in contemporary condensed-matter science. Although the magnetolectric effect itself has a long history of investigations⁶, the recent discovery of the large magnetolectric effect in multiferroics, in which particular spin orders produce ferroelectric polarization, accelerates the research of this area^{2,3,7}. Among them, a unique origin of ferroelectricity has been found in perovskite RMnO_3 (R = rare earth), where the helical spin structure produces the macroscopic electric polarization irrespective of the original lattice symmetry. The origin of this ferroelectricity is successfully explained by the spin-current model, or equivalently the inverse effect of the Dzyaloshinskii–Moriya interaction^{4,8}; the microscopic polarization around the bond connecting adjacent spins is produced by the term proportional to vector product $\mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$, where \mathbf{e}_{ij} is a unit vector connecting the adjacent spins \mathbf{S}_i and \mathbf{S}_j . In this model, the spontaneous spin current is viewed as flowing between the mutually canted spins (\mathbf{S}_i and \mathbf{S}_j) with the magnetization component along the direction of $(\mathbf{S}_i \times \mathbf{S}_j)$, which in turn generates the electronic polarization under the spin-orbit interaction. The summation of these microscopic polarizations gives rise to the ferroelectricity in the cycloidal (transverse-spiral) spin structure, where the static polarization lies in the helical spin plane and is directed perpendicular to the magnetic modulation vector \mathbf{q}_m (Fig. 1a).

Such magnetically driven ferroelectricity promises a concomitant collective spin excitation endowed with electric dipole activity, termed an electromagnon⁹. In the cycloidal spin phase, the

rotational vibration of the helical spin plane accompanies that of the ferroelectric polarization (mode I in Fig. 1b), leading to the electric activity of this magnetic excitation as the electromagnon⁵. However, the existence of this magnetic excitation generic for multiferroic order has remained controversial^{9–14}; spin excitations with transition electric dipoles have in fact been observed for several multiferroics, but their origins have now been identified to be different from the above cross-coupling electromagnon; that mode is induced by the magnetostriction mechanism, unrelated to spin-orbit interaction, and shows no magnetic-dipole activity^{15–19}. In this study, we have unravelled the cross-coupling electromagnon driven by the spin-current mechanism in a perovskite manganite $\text{Eu}_{0.55}\text{Y}_{0.45}\text{MnO}_3$ as well as its generic dynamical magnetolectric effect.

In $\text{Eu}_{0.55}\text{Y}_{0.45}\text{MnO}_3$, the spin structure exhibits the ab -plane cycloidal spin phase in the ground state with $\mathbf{q}_m \parallel b$ axis (Fig. 1a)²⁰. With the onset of the ab -plane cycloidal spin phase, the ferroelectric polarization directed along the a axis shows up as predicted by the spin-current model. In the terahertz regime, a strongly electric-dipole-active absorption is observed when the light electric vector \mathbf{E}^ω is parallel to the a axis as shown in Fig. 1c. This $\mathbf{E}^\omega \parallel a$ electromagnon is commonly observed in the helical spin phase in RMnO_3 compounds irrespective of the spin-spiral plane configuration (ab or bc plane), and has been elucidated in terms of the magnetostriction mechanism; the electric field of light acts on the local electric dipole produced by GaFeO_3 -type lattice distortion, which in turn modulates the exchange interaction between the adjacent spins and gives rise to the in-plane spin rotational vibration (mode III shown in Fig. 1b)^{18,19}. Although the electromagnon arising from this magnetostriction mechanism corresponds to the zone-edge magnon mode ($q = \pi$; refs 18,19; 8 meV for this material²¹), it has been shown²² that magnetic anisotropy terms induce the low-energy peak of the electromagnon with $q = \pi - 2q_m$ at 2.4 meV, as plotted in Fig. 1c. Note that such a magnetostriction-induced electromagnon mode with the electric-dipole activity is, however, not magnetic-dipole active owing to their $q \neq 0$ and $q \neq q_m$ nature, lacking in cross-coupling nature (see Supplementary Information).

The electromagnon of spin-current origin, on the other hand, should be active for $\mathbf{E}^\omega \parallel c$ polarization in the case of ab -plane cycloidal spin structure where the static ferroelectric polarization \mathbf{P} is directed along the a axis (mode I shown in Fig. 1b). Figure 1d,e shows the spectra of the imaginary parts of the refractive indices under the external magnetic field along the c axis. A distinct

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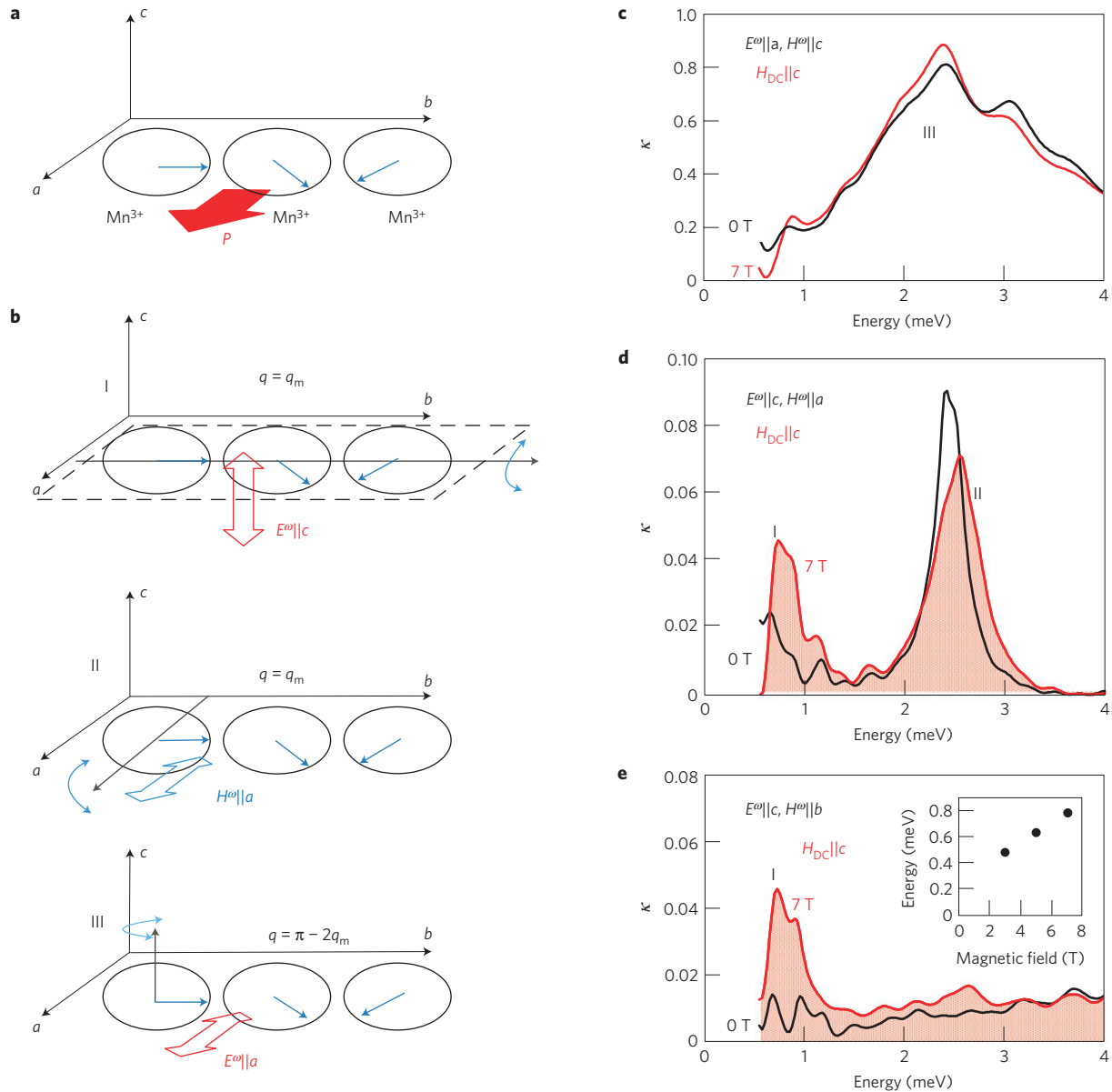


Figure 1 | Spin excitations in cycloidal spin state. **a**, Schematic spin structure and direction of static ferroelectric polarization in the ab -plane cycloidal spin phase. **b**, Illustration of the spin motion. Mode I (cross-coupling electromagnon): rotational vibration of the helical spin plane around the b axis. Mode II (magnon): rotational vibration of the spins around the a axis. Mode III (magnetostriction-induced electromagnon): rotational vibration of the spins around the c axis. Wavenumbers of the magnons are also indicated. **c–e**, Imaginary part κ of the refractive index for $(\mathbf{E}^\omega \parallel a, \mathbf{H}^\omega \parallel c)$ (**c**), $(\mathbf{E}^\omega \parallel c, \mathbf{H}^\omega \parallel a)$ (**d**) and $(\mathbf{E}^\omega \parallel c, \mathbf{H}^\omega \parallel b)$ (**e**) under the external magnetic field along the c axis at 4 K. The inset in **e** shows the magnetic-field dependence of the energy of mode I. (Tiny structures in κ around 1 meV at 0 T are background or artefact, showing little temperature dependence.)

peak structure appears at 0.8 meV (7 T) for both $(\mathbf{E}^\omega \parallel c, \mathbf{H}^\omega \parallel a)$ (Fig. 1d) and $(\mathbf{E}^\omega \parallel c, \mathbf{H}^\omega \parallel b)$ (Fig. 1e), signalling the electric activity of this mode. (Note that mode I is also active for $\mathbf{H}^\omega \parallel b$, and hence spectra in Fig. 1e may contain some amount of magnetically active component.) This makes a strong contrast with the peak structure at 2.7 meV in Fig. 1d, which is absent from Fig. 1e and has been attributed to the conventional antiferromagnetic resonance (AFMR) driven by the $\mathbf{H}^\omega \parallel a$ component of light (mode II in Fig. 1b). The electromagnon observed at about 0.8 meV can be identified as the Nambu–Goldstone mode of the cycloidal spin order^{5,13}; the magnetic anisotropy due to the energy difference between the ab - and bc -plane helical states as well as to the external magnetic field induces the finite resonance energy as observed. The resonance energy increases with the magnetic field (inset to Fig. 1e), because the magnetic field along the c axis

stabilizes the ab -plane helical state²⁰, leading to an increase of the magnetic anisotropy.

The energies of magnons including electromagnons observed by the terahertz spectroscopy are compatible with the result of polarized inelastic neutron scattering experiments on TbMnO_3 (refs 10,11), of which the magnon dispersion is plausibly similar to that of $\text{Eu}_{0.55}\text{Y}_{0.45}\text{MnO}_3$. The magnons in the bc -plane cycloidal spin state of TbMnO_3 exhibit three branches at $(0, q_m, 1)$. Two modes with out-of-plane dynamical spin component at about 1.1 and 2.5 meV can be ascribed to the spin-current-driven electromagnon (mode I) and AFMR (mode II), respectively, whereas the electromagnon due to magnetostriction (mode III) is attributed to the magnon at another k point.

One of the most intriguing aspects of this spin-current-driven electromagnon is its dynamical magnetoelectric effect,

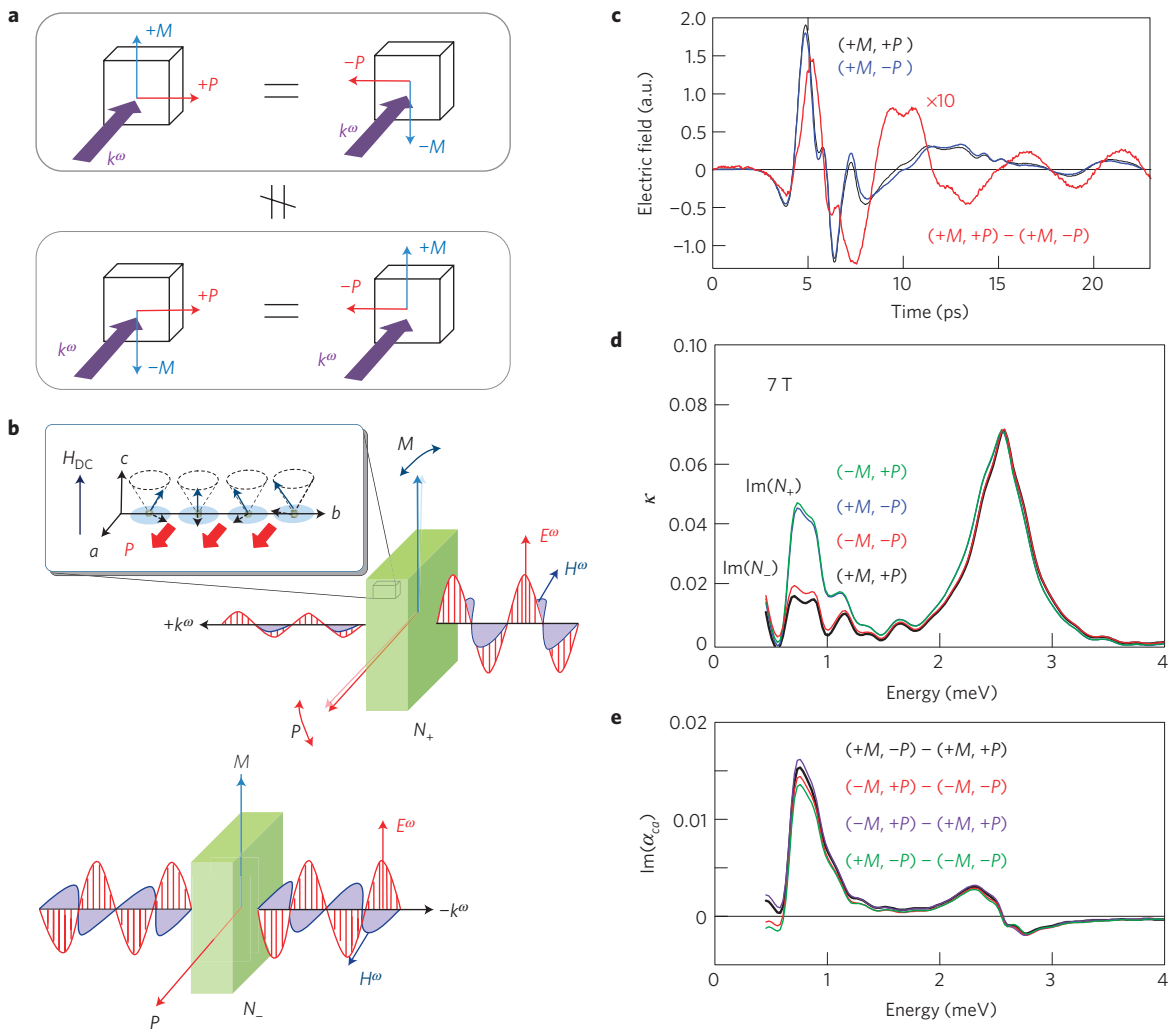


Figure 2 | Experimental observation of magnetoelectric resonance. **a**, Configurations of \mathbf{P} , \mathbf{M} and \mathbf{k}^ω and their relation in terms of the optical magnetoelectric effect (see text). **b**, Experimental configuration for the matter (\mathbf{P} and \mathbf{M}), and for the light (\mathbf{E}^ω , \mathbf{H}^ω and \mathbf{k}^ω) with respect to two different conditions of $\pm\mathbf{k}^\omega$ (N_\pm). \mathbf{E}^ω and \mathbf{H}^ω of light with $+\mathbf{k}^\omega$ can constructively vibrate both \mathbf{P} and \mathbf{M} (upper panel), whereas \mathbf{E}^ω and \mathbf{H}^ω of $-\mathbf{k}^\omega$ suppress the vibrations of \mathbf{P} and \mathbf{M} with each other (lower panel). **c**, Time-domain waveform of the transmitted terahertz pulses in different configurations in terms of the magnetoelectric effect (black and blue lines) and the difference between them (red line) at 7 T. **d**, Imaginary part of refractive index for $\mathbf{E}^\omega \parallel \mathbf{c}$, $\mathbf{H}^\omega \parallel \mathbf{a}$ under magnetic field along the c axis (7 T) at 4 K. Spectra for the four possible configurations in **a** are shown. **e**, Imaginary part of α_{ca} . Results for four possible combinations to obtain $\text{Im}(\alpha_{ca})$ are shown.

arising from the strong cross-coupling between the induced polarization \mathbf{P}^ω and magnetization \mathbf{M}^ω . The dynamical response of the first-order magnetoelectric coupling can be described by the dynamical magnetoelectric tensor $\alpha_{ij}(\omega)$ in addition to the conventional terms (dynamical dielectric and magnetic susceptibility) $\chi^{ee}(\omega)$ and $\chi^{mm}(\omega)$,

$$\begin{aligned} P_i^\omega &= \chi_{ii}^{ee}(\omega)E_i^\omega + \alpha_{ij}(\omega)H_j^\omega \\ M_j^\omega &= \chi_{jj}^{mm}(\omega)H_j^\omega + \alpha_{ij}(\omega)E_i^\omega \end{aligned} \quad (1)$$

To enable the existence of the magnetoelectric tensor, the symmetry consideration requires the simultaneous breaking of time-reversal and space-inversion symmetry, for example the configuration shown in Fig. 2a with the orthogonal relations among \mathbf{P} , \mathbf{M} and light wavevector \mathbf{k}^ω (ref. 23). (Note that \mathbf{P} and \mathbf{M} are the static quantities here.) When this condition is satisfied, the optical magnetoelectric effect for the linearly polarized light can appear with directional non-reciprocity; light with \mathbf{k}^ω vectors parallel and antiparallel to $\mathbf{P} \times \mathbf{M}$ experiences different optical responses (Fig. 2b). By

combining Maxwell's equations with equation (1), the directional complex refractive index is given for $\pm\mathbf{k}^\omega$:

$$N_\pm(\omega) = n(\omega) + i\kappa(\omega) = (n_0(\omega) + i\kappa_0(\omega)) \pm \alpha_{ij}(\omega) \quad (2)$$

where n_0 and κ_0 are the conventional (non-magnetoelectric) parts of the refractive index²⁴. The real and imaginary parts of $\alpha_{ij}(\omega)$ contribute to the directional birefringence and dichroism, respectively. The reversal of one of three parameters (\mathbf{P} , \mathbf{M} and \mathbf{k}^ω) results in a sign change of $\alpha_{ij}(\omega)$ in the refractive index $N_\pm(\omega)$. To observe the non-reciprocal dichroism, we carried out the experiments in four configurations of $(\mathbf{k}^\omega, \mathbf{P}, \mathbf{M})$, which are categorized into two equivalent configurations in terms of the sign of the product, $\mathbf{k}^\omega \cdot (\mathbf{P} \times \mathbf{M})$ (see Fig. 2a). As no spontaneous magnetization appears in the cycloidal spin state, we applied an external magnetic field along the c axis to induce the cone-like transverse helical spin structure and therefore cause a finite static magnetization $\mathbf{M} \parallel c$. As a result, the coexistence of spontaneous ferroelectric polarization $\mathbf{P} \parallel a$ and field-induced magnetization $\mathbf{M} \parallel c$ gives rise to a finite

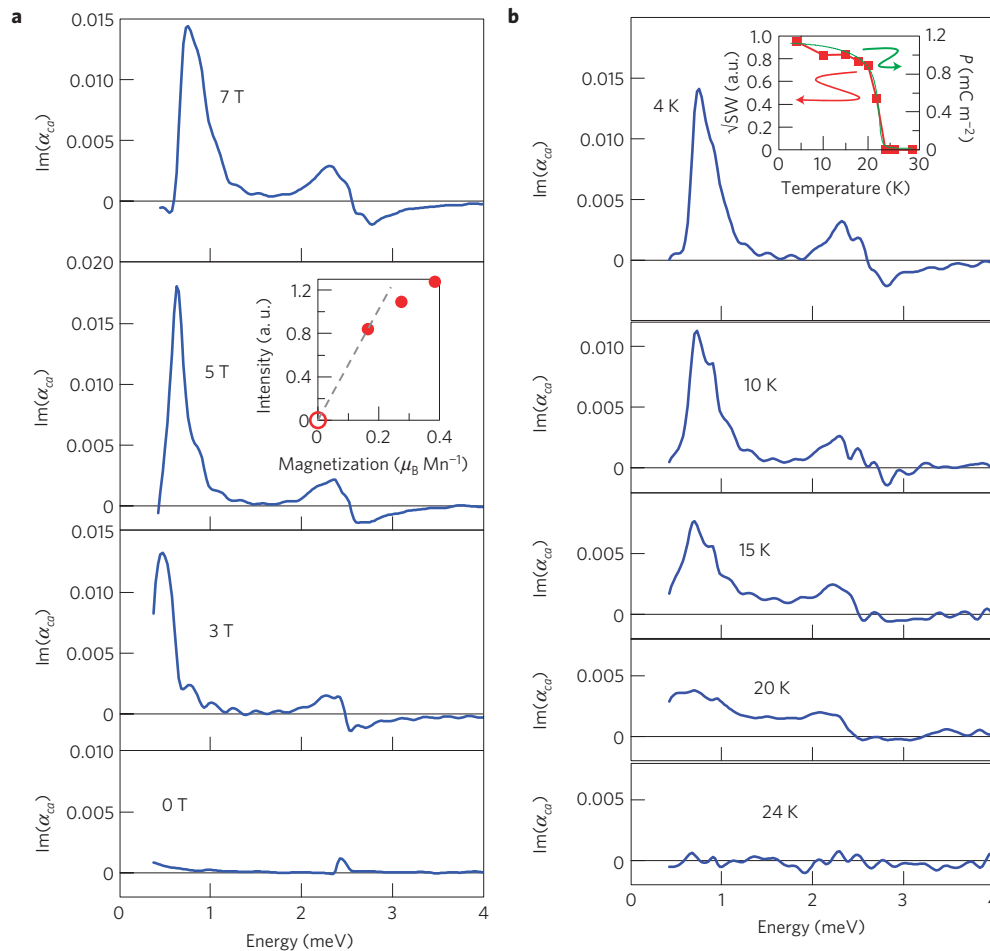


Figure 3 | Temperature and magnetic-field dependence of magnetolectric resonance spectra. a, Magnetic-field dependence of $\text{Im}(\alpha_{ca})$ at 4 K. Inset: The magnetization dependence of the integrated intensity of $\text{Im}(\alpha_{ca})$ at the electromagnon resonance (red filled circles), which shows the deviation from the linear magnetic-field dependence (grey dashed line). No magnetolectric effect is anticipated in zero magnetic field (as indicated with a red open circle), because the time-reversal invariance of the cycloid spin state prohibits the magnetolectric effect. **b,** Temperature dependence of $\text{Im}(\alpha_{ca})$ at 7 T. Inset: The temperature dependence of spontaneous polarization and square root of spectral weight (SW) of $\text{Im}(\alpha_{ca})$.

value of $\mathbf{P} \times \mathbf{M}$ ($\parallel \mathbf{b}$); the light polarization of ($\mathbf{E}^\omega \parallel c$, $\mathbf{H}^\omega \parallel a$) is selected to enable both magnetic and electric resonances with the spin-current-driven electromagnon with the light \mathbf{k}^ω -vector along the b axis.

As shown in Fig. 2c, the temporal waveform of the terahertz pulse transmitted through the sample shows a distinct difference depending on the sign of $\mathbf{P} \times \mathbf{M}$, indicating the occurrence of the dynamical magnetolectric effect. The subtracted waveform exhibits a low-frequency oscillation with a period of 5.2 ps, which corresponds to 0.8 meV in photon energy. In fact, the imaginary part of the refractive index κ shows a dramatic difference between the two inequivalent configurations at the electromagnon resonance, in contrast to the AFMR at 2.7 meV, which mostly conserves its intensity (Fig. 2d). The spectra of the 0.8 meV resonance exhibit the enhanced absorption ($\text{Im}(N_+)$) with \mathbf{k}^ω parallel to $\mathbf{P} \times \mathbf{M}$, but the suppressed one ($\text{Im}(N_-)$) for \mathbf{k}^ω antiparallel to $\mathbf{P} \times \mathbf{M}$. This difference directly gives the component of the magnetolectric tensor α_{ca} for light polarization of ($\mathbf{E}^\omega \parallel c$, $\mathbf{H}^\omega \parallel a$); $\text{Im}(\alpha_{ca}) = (\text{Im}(N_+) - \text{Im}(N_-))/2$ (see equation (2)). As shown in Fig. 2e, a colossal directional dichroism is discerned at the resonance of the spin-current-driven electromagnon even for small \mathbf{M} ($0.37 \mu_B/\text{Mn}^{3+}$ at 7 T), which is less than 10% of the fully spin-polarized value ($4 \mu_B/\text{Mn}^{3+}$). Although the maximum value of $\text{Im}(\alpha_{ca})$ is restricted by the second law of thermodynamics as given by $\kappa = \kappa_0 - \text{Im}(\alpha_{ca}) > 0$, the observed magnitude of $\text{Im}(\alpha_{ca})$

(~ 0.15) approaches roughly 50% of κ_0 ($=(\kappa_+ + \kappa_-)/2 \sim 0.03$) at 7 T, namely the same order as κ_0 . The first observation of the optical magnetolectric effect has been reported on the electronic transition on the isolated rare-earth ions in garnet with relatively small magnitude ($\sim 10^{-3}\%$; ref. 23). Recently, enhancement of the magnetolectric effect has been reported on several particular electronic and magnetic resonances, which are the transitions of the local d electron in the ligand field in X-ray²⁵, visible and near-infrared^{26,27} and far-infrared regions²⁸. In the present case, the colossal dynamical magnetolectric effect emerges at the resonance of the spin-current-driven electromagnon with both electric- and magnetic-dipole activities as the dynamical counterpart of the large static magnetolectric effect in the multiferroic system. Incidentally, this kind of dynamical magnetolectric effect can be distinguished from the other type of magnetolectric coupling on magnetostrictive–piezoelectric devices, where the magnetic resonance is coupled with the static electric field through strain^{6,29}. On the other hand, the magnetolectric resonance observed at present may be viewed as the dynamical resonance of toroidal moment³⁰, as the static toroidal (ferrotoroidic) moment is always anticipated under the existence of $\mathbf{P} \times \mathbf{M}$.

A relatively small but finite magnetolectric effect appears around 2.5 meV as shown in Fig. 2e, indicating that the originally genuine AFMR (mode II) gains magnetolectric coupling in the magnetic field. Although the AFMR shows basically no electric

activity, this result suggests that the finite electric activity shows up in the \mathbf{H} -induced conical spin state. In fact, the electric component of AFMR for $\mathbf{E}^\omega \parallel c$ is manifested by the tiny structures around 2.6 meV at 7 T as shown in Fig. 1e. The electric activity of AFMR may be induced by the transfer of the spectral weight from the electromagnons through the H -induced coupling among these excitations.

Figure 3a shows the magnetic-field dependence of $\text{Im}(\alpha_{ca})$, which is relevant to the sign and magnitude of $\mathbf{P} \times \mathbf{M}$. In the present condition, \mathbf{P} is nearly constant and \mathbf{M} is proportional to the magnetic field. Near $H = 0$, $\text{Im}(\alpha_{ca})$ should be proportional to \mathbf{M} , because $\text{Im}(\alpha_{ca})$ originates from the first-order magnetoelectric effect with respect to \mathbf{M} and \mathbf{P} . Indeed, $\text{Im}(\alpha_{ca})$ increases with magnetic field (Fig. 3a); however, a deviation from linear \mathbf{M} dependence is observed (inset to Fig. 3a), indicating that the $\text{Im}(\alpha_{ca})$ goes beyond the linear regime to \mathbf{M} .

Figure 3b shows the temperature dependence of the dynamical magnetoelectric effect. Apparently, the magnetoelectric effect emerges below 23 K (ref. 20), where the helical spin state appears. The temperature dependence of the spectral weight of $\text{Im}(\alpha_{ca})$ is plotted in the inset of Fig. 3b, showing a good agreement with the temperature dependence of \mathbf{P}^2 . Because this electromagnon has its origin in the fluctuation of the spontaneous \mathbf{P} , this coincidence confirms the assignment of this electromagnon to the rotational vibration mode of the helical spin plane (Fig. 1b1).

A magnetoelectric resonance originating from the fluctuation in \mathbf{P} as observed in the present study should be ubiquitous for any magnetically driven ferroelectrics (for example multiferroic helimagnets), and is always expected to give rise to the dynamical magnetoelectric effect in a gigahertz-to-terahertz frequency region. The extension of the magnetoelectric effect toward the dynamical region may open a new arena in the control of electromagnetism of materials beyond the conventional optics.

Methods

Single crystals of orthorhombic perovskite $\text{Eu}_{0.55}\text{Y}_{0.45}\text{MnO}_3$ were grown by the floating-zone method. To obtain the value of ferroelectric polarization, pyroelectric current was measured after cooling with the electric field of $\pm 400 \text{ V cm}^{-1}$ along the a axis and the magnetic field of 7 T along the c axis. Terahertz time-domain spectroscopy was carried out under the magnetic field applied perpendicular to the light propagation direction (in the Voigt configuration) to determine the complex refractive index in the energy range from 0.4 to 4 meV. A femtosecond laser pulse was split into two paths to generate and detect the terahertz wave. The terahertz pulse was emitted from a bow-tie-shape photoconductive antenna and detected by a dipole antenna. To observe the dynamical magnetoelectric effect due to the spin-current-driven electromagnon, the polarization configuration of ($\mathbf{E}^\omega \parallel c$, $\mathbf{H}^\omega \parallel a$) was employed (Fig. 2b). The sample was placed in a superconducting magnet, which can apply magnetic field up to 7 T. To measure the optical magnetoelectric effect, d.c. electric field of 1 kV cm^{-1} was applied to the sample during the cooling process and then switched off at 4 K, so that a single ferroelectric domain was formed. During the terahertz measurements, no d.c. electric field was applied to the sample.

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Author contributions

Y. Takahashi carried out optical terahertz spectroscopy and analysed data. Y.K. carried out crystal growth. H.M. measured pyroelectric current. The results were discussed and interpreted by Y. Takahashi, R.S. and Y. Tokura.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturephysics. Reprints and permissions information is available online at <http://www.nature.com/reprints>. Correspondence and requests for materials should be addressed to Y. Takahashi.