Editors' Suggestion

Electrical Switching of the Nonreciprocal Directional Microwave Response in a Triplon Bose-Einstein Condensate

Shojiro Kimura*
Institute for Materials Research, Tohoku University, Katahira 2-1-1, Sendai 980-8577, Japan

Masashige Matsumoto
Department of Physics, Shizuoka University, Shizuoka 422-8529, Japan

Hidekazu Tanaka
Department of Physics, Tokyo Institute of Technology, Tokyo 152-8551, Japan

(Received 11 December 2019; revised manuscript received 9 April 2020; accepted 29 April 2020; published 27 May 2020)

We present a microwave electron spin resonance study of the quantum spin dimer system $TlCuCl_3$, which shows the magnetic-field-induced ordering with both antiferromagnetic spin order and ferroelectricity by the Bose-Einstein condensation (BEC) of triplon quasiparticles. Our main achievement is an electrical switching of the nonreciprocal directional microwave response in the triplon BEC phase. High-speed directional control of microwave absorption by applying an electric field has been accomplished in this Letter. The strength of the observed nonreciprocal microwave response well agrees with the calculation based on Kubo theory with the parameters, evaluated from the static electric polarization in this material.

DOI: 10.1103/PhysRevLett.124.217401

Magnetoelectric (ME) multiferroic materials with both magnetic order and ferroelectricity are representative of systems with broken space-inversion and time-reversal symmetries. In materials with such broken symmetries, responses to quantum particles traveling in opposite directions can be distinguished from each other [1,2]. By utilizing this nonreciprocal directional response, rectification of electromagnetic wave propagation is achieved. In particular, in a ME multiferroic material, the directional response is reversed by reversing its electric polarization. In this Letter, we report electrical switching of the nonreciprocal directional microwave response in the quantum magnet $TlCuCl_3$ in the triplon Bose-Einstein condensation (BEC) phase with multiferroic ordering. It is demonstrated that by applying electrical voltage the directional response of microwave absorption due to the excitation of the Nambu-Goldstone magnon can be immediately reversed, thereby achieving fast interchange between transparent and absorbing directions of microwave propagation. Therefore, the triplon Bose-Einstein condensate works as an electrically controlled directional microwave switch.

The nonreciprocal directional dichroism (NDD), a change in optical absorption by reversal of the direction of electromagnetic wave propagation, has vast implications for photonic devices, such as an electromagnetic wave isolator, which is necessary for optical or wireless communication technology. Therefore, considerable efforts have been made to achieve efficient control of the directional electromagnetic wave propagation [3–5]. Dynamical coupling between spins and electric dipoles in ME multiferroic materials offers a key to realize strong NDD [5–22]. Through the dynamical ME coupling, oscillation of the spins, caused by magnetic fields of electromagnetic wave, generates oscillation of electric dipoles, and then interference between the oscillating electric dipoles, induced by the magnetic and electric components of the electromagnetic wave, causes NDD. From the symmetry point of view, nonreciprocal propagation occurs when all symmetries, connecting the opposite propagation directions, are broken [1]. This symmetry condition is fulfilled when the propagation vector $\boldsymbol{k}$ is parallel or antiparallel to the direction of $\boldsymbol{P}\times\boldsymbol{M}$ in a material [1]. Here, $\boldsymbol{P}$ and $\boldsymbol{M}$ are the macroscopic electric polarization and magnetization, respectively. In this case, modulation of an absorption coefficient $\Delta\alpha = \gamma\boldsymbol{k}\cdot(\boldsymbol{P}\times\boldsymbol{M})$ (γ being a constant), whose sign change due to the reversal of $\boldsymbol{k}$ causes a NDD, is expected [1,2,8,9,12,13,20,21]. It is noteworthy that the sign of $\Delta\alpha$ also changes by reversal of $\boldsymbol{P}$ or $\boldsymbol{M}$. Thus, the direction of $\boldsymbol{k}$ with strong light absorption is reversed by reversal of $\boldsymbol{P}$ or $\boldsymbol{M}$, resulting in the reversal of the nonreciprocal directional response. Reversal of $\boldsymbol{P}$ by applying the dc electric field $\boldsymbol{E}$, which can immediately switch the nonreciprocal directional response with no energy consumption, is more effective than that of $\boldsymbol{M}$, which requires sweeping of the dc magnetic field $\boldsymbol{H}$. However, such immediate switching by $\boldsymbol{E}$ is difficult to achieve because reversal of $\boldsymbol{P}$ in ME multiferroic materials often demands high electric voltage. Therefore, so far, the reversal of $\boldsymbol{P}$ has been achieved by the

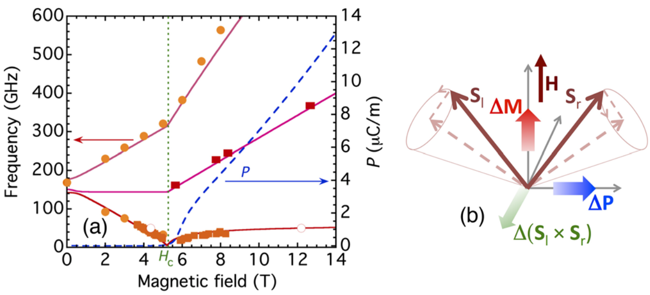

FIG. 1. (a) Magnetic-field dependence of the magnetic excitation energies at the magnetic zone center in $TlCuCl_3$ for $\boldsymbol{H}\|[201]$, calculated based on the bond operator formulation. The dotted line represents the critical field H_c. Open circles represent the ESR points at 50 GHz, where the NDD measurements are performed in this study. Closed circles are the data, obtained from the previous neutron scattering [28]. Closed squares below 80 GHz are the data obtained from the microwave ESR measurements [32], and those above 160 GHz are the data obtained in this study. Three branches of the triplon excitation, which are triply degenerated at zero magnetic field in the case of an isotropic system, split by a rhombic anisotropy. Owing to the triplon BEC, the field dependence of the two higher energy modes is renormalized, whereas the lowest mode changes to the Nambu-Goldstone mode for $H > H_c$ and the anisotropy causes reopening of the excitation gap in the Nambu-Goldstone mode. As shown by a dashed curve, the spontaneous electric polarization $\boldsymbol{P}$ perpendicular to the $(10\bar{2})$ plane appears for $H > H_c$. Thus, the triplon BEC brings about a finite $\boldsymbol{P}\times\boldsymbol{M}$ parallel to [010] in $TlCuCl_3$ for $\boldsymbol{H}\|[201]$. (b) Precession motion of the two-sublattice-ordered spin moments, driven by the excitation of the Nambu-Goldstone magnon for $H > H_c$.

reversal of poling fields in the cooling procedure under $\boldsymbol{E}$ and $\boldsymbol{H}$ from a nonpolar phase [12,13,19,21].

Recent studies have revealed that the interacting dimer system $TlCuCl_3$ exhibits soft ferroelectricity, which is desirable for the reversal of $\boldsymbol{P}$ by $\boldsymbol{E}$, in its magnetic-field-induced triplon BEC phase [23,24]. In this compound, spin $S = 1/2$ antiferromagnetic dimers, composed of two Cu^{2+} ions, are coupled by relatively weak interdimer interactions forming a three-dimensional dimer network. Reflecting the dimer structure, the ground state of $TlCuCl_3$ at zero magnetic field is a nonmagnetic singlet state, and thus remains quantum paramagnet down to the lowest temperature [25]. The magnetic excitation from the singlet state is a bosonic quasiparticle called a triplon, which is an excited triplet with $S = 1$ on a dimer propagating through the lattice [26–30]. In $\boldsymbol{H}$, the triply degenerated triplon modes split into three branches, as shown in Fig. 1(a). The energy gap between the ground state and the lowest energy triplon mode is closed at a critical field $H_c \simeq 5.5$ T. At this H_c, the quantum phase transition by the BEC of the triplon takes place [25–30]. The triplon BEC in $TlCuCl_3$ leads not only to a long-range two-sublattice ordering of the transverse spin components but also to ferroelectricity [23,24]. This is because coherent superposition of the singlet and triplet states on a dimer, realized in the BEC phase, has a finite expectation value of an outer product $\langle \boldsymbol{S}_l \times \boldsymbol{S}_r \rangle$, called the vector spin chirality. This generates a local electric polarization on the dimer, with $\boldsymbol{S}_l$ and $\boldsymbol{S}_r$ being the spins on the dimer. Actually, spontaneous electric polarization $\boldsymbol{P}$ develops above H_c in proportion to an absolute value of the expectation value of $\langle \boldsymbol{S}_l \times \boldsymbol{S}_r \rangle$ in the ground state of $TlCuCl_3$ [23,24]. The resulting ferroelectricity in $TlCuCl_3$ is very soft. Reversal of $\boldsymbol{P}$, which causes the reversal of the antiferromagnetic domain, can be induced by the dc electric field $E_c \simeq 0.03$ MV/m, which is the lowest cohesive field among those of the ME multiferroic materials. Dynamical coupling between spins and electric fields was also found in the interacting spin dimer system. The triplon excitation below H_c can be induced by the oscillating electric field of electromagnetic wave [31], indicating that the ferroelectricity results from the phase coherence of the electroactive triplons in the Bose-Einstein condensate. Above H_c, owing to the breaking of O(2) symmetry by the triplon BEC, the lowest energy triplon changes to the Nambu-Goldstone magnon, which is the phase oscillation of the order parameter in a Bose-Einstein condensate [28–30]. Because of inevitable magnetic anisotropy, the Nambu-Goldstone magnon, which is gapless in an isotropic case, acquires a small excitation gap at the magnetic zone center in $TlCuCl_3$ and thus can be excited by microwave illumination [32]. An important point is that the Nambu-Goldstone magnon is both magneto- and electroactive. As shown in Fig. 1(b), excitation of the Nambu-Goldstone magnon drives the precession motion of the two-sublattice-ordered magnetic moments with phase difference of π between them around those equivalent directions [29]. This precession motion induces the longitudinal oscillation $\Delta\boldsymbol{M}$ of the total magnetization along $\boldsymbol{H}$ and also the oscillation of $\boldsymbol{S}_l \times \boldsymbol{S}_r$ in the plane perpendicular to $\boldsymbol{H}$. If the oscillation of $\boldsymbol{S}_l \times \boldsymbol{S}_r$ induces dynamical electric polarization $\Delta\boldsymbol{P}$, oscillating perpendicular to $\Delta\boldsymbol{M}$, NDD occurs [12,13,20].

To observe the switching of the nonreciprocal directional microwave response in the triplon BEC phase, we measured the electron spin resonance (ESR) at frequency 50 GHz in high magnetic field for $\boldsymbol{H}\|[201]$. A single-crystal $TlCuCl_3$, which was grown by the Bridgman method, was cut into thin plates with the widest plane parallel to the cleavage (010) plane and the lateral cleavage $(10\bar{2})$ plane, and then a silver paste was applied on the $(10\bar{2})$ plane of the crystal as electrodes. The thickness of the sample was 1.1 mm, and the area of the widest plane of the sample was 3.5×5.0 mm^2. The NDD was measured by utilizing a homemade transmission-type ESR system with a 15 T superconducting magnet. A Gunn oscillator (Millitech GDV-19-1010IR) and a Schottky diode detector (Millitech DET-19-S) were used as a microwave source and detector,

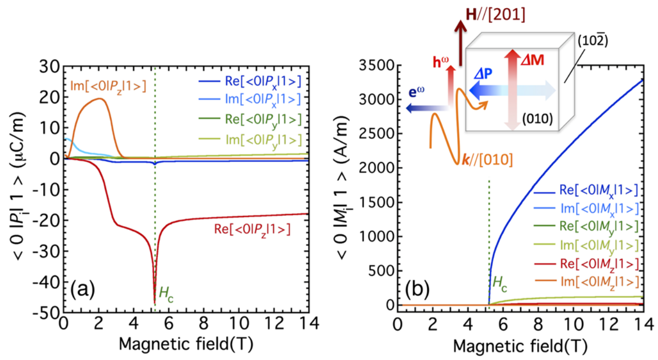

FIG. 2. Transition matrix elements (a) $\langle 0|P_i|1\rangle$ and (b) $\langle 0|M_i|1\rangle$, $(i = x,\ y,\ z)$ between the ground state $|0\rangle$ and the excited state $|1\rangle$ for the excitation of the lowest energy triplon or the Nambu-Goldstone magnon, calculated for $\boldsymbol{H}\|[201]$. (Inset) Both longitudinal oscillation of $\boldsymbol{M}$ along [201] and transverse oscillation of $\boldsymbol{P}$ perpendicular to $(10\bar{2})$ plane are induced by the excitation of the Nambu-Goldstone magnon for $H > H_c$. In the ESR experiment, the linearly polarized microwave with the oscillating magnetic field $\boldsymbol{h}\|[201]$ and the electric field $\boldsymbol{e}\perp(10\bar{2})$ is illuminated to a single crystal of $TlCuCl_3$ (see inset).

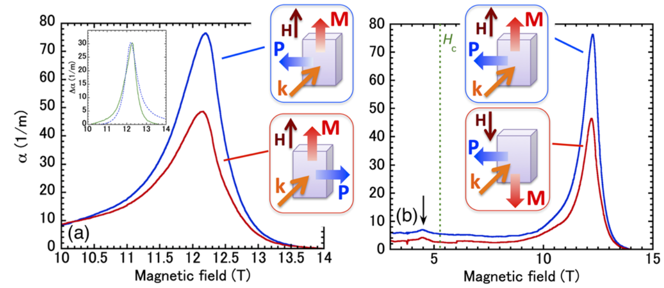

FIG. 3. (a) Microwave absorption due to the excitation of the Nambu-Goldstone mode at 50 GHz at 2.0 K. Reversal of $\boldsymbol{P}$ causes large change in the absorption coefficient. (Inset) Change in the absorption coefficient $\Delta\alpha = \alpha_+ - \alpha_-$ by the reversal of $\boldsymbol{P}$. Solid and dashed curves represent the experimental and theoretical results, respectively. (b) Microwave absorption spectra observed in reversed magnetic fields. The black arrow indicates the absorption due to the excitation of the triplon below H_c. Change in the absorption due to the excitation of the Nambu-Goldstone mode by the reversal of $\boldsymbol{M}$, as well as by the reversal of $\boldsymbol{P}$ for $H > H_c$, is observed, whereas the absorption due to the excitation of the triplon shows almost no change.

respectively. A wire grid polarizer is set in front of the single-crystal sample to illuminate a sample with the linearly polarized microwave. ESR spectra were observed under static electric field $E = \pm 0.058$ MV/m, applied perpendicular to the $(10\bar{2})$ plane of the single-crystal sample. The absorption coefficient is calculated from $\alpha = ln(I/I_0)/z$ with I_0 microwave transmission at 14 T and $z = 1.1 \times 10^{-3}$ m the sample thickness. The ESR data above 160 GHz, plotted with closed squares in Fig. 1(a), are obtained from the measurements, using 81 and 114 GHz Gunn oscillators (Millitech GDM-10-1019R and VCSS CD0-08-115) combined with a doubler (VDI WR5.1 × 2), a submillimeter wave source (VDI 490 GHz Modular Tx) and an InSb hot electron bolometer (QMC QFI/2BI).

Figure 1(a) shows the field dependence of the magnetic excitation energy, calculated based on a bond operator formulation. As demonstrated in Refs. [29,30], the bond operator formulation can quantitatively explain both the excited and ground state properties of $TlCuCl_3$. By taking into account the magnetic anisotropy, the reopening of the excitation gap of the Nambu-Goldstone mode above H_c is also reproduced, as shown in Fig. 1(a). Based on this bond operator formulation, the transition matrix elements $\langle 0|P_i|1\rangle$ and $\langle 0|M_i|1\rangle$, $(i = x,\ y,\ z)$, between the ground state $|0\rangle$ and the excited state $|1\rangle$ for excitation of the lowest energy triplon or the Nambu-Goldstone magnon are calculated as shown in Figs. 2(a) and 2(b), respectively. Here, $\boldsymbol{M} = g\mu_B \boldsymbol{S}$ with $g = 2.06$ [33] and Bohr magneton μ_B, and $\boldsymbol{P} = \tilde{C}(\boldsymbol{S}_l \times \boldsymbol{S}_r)$ with a second rank tensor $\tilde{C}$. The tensor components C^α_β, evaluated from the static electric polarization, are used for the calculations (Supplemental Material [34]). We define the x and y axes parallel to [201] and [010], respectively, and the z axis perpendicular to the $(10\bar{2})$ plane. Figures 2(a) and 2(b) show that only the transition matrix $\langle 0|P_i|1\rangle$'s are finite for $H < H_c$, indicating that the triplon is electroactive. On the other hand, both $\langle 0|M_i|1\rangle$'s and $\langle 0|P_i|1\rangle$'s become finite for the Nambu-Goldstone magnon above H_c, showing that the Nambu-Goldstone magnon is both magneto- and electroactive. The large absolute values of $\langle 0|M_x|1\rangle$ and $\langle 0|P_z|1\rangle$ indicate that $\boldsymbol{M}$ and $\boldsymbol{P}$ oscillate almost parallel to the [201] axis and perpendicular to the $(10\bar{2})$ plane, respectively, when the Nambu-Goldstone magnon is excited. Therefore, we illuminate the linearly polarized microwave with the oscillating magnetic field $\boldsymbol{h}\|[201]$ and the electric field $\boldsymbol{e}\perp(10\bar{2})$ to a single crystal of $TlCuCl_3$ to examine NDD. Results of the observation are shown in Fig. 3. As shown in Fig. 3(a), microwave absorption due to the excitation of the Nambu-Goldstone magnon at around 12.2 T largely changes by reversing $\boldsymbol{P}$, as expected. More than 45% change in the absorption by reversing $\boldsymbol{P}$ is observed. The change in absorption coefficient $\Delta\alpha = \alpha_+ - \alpha_-$ is determined by the dynamical magnetoelectric susceptibility χ^{em}_{zx} as follows [15]:

$$\Delta\alpha = (4\omega_0/c)\mathrm{Im}\chi^{em}_{zx}, \quad (1)$$

where $\omega_0 = 2\pi \times 50$ GHz. According to the Kubo theory, χ^{em}_{zx} is calculated as [15]

$$\chi_{zx}^{em} = \frac{2}{\hbar N}\sqrt{\frac{\mu_0}{\epsilon_0}}\frac{\omega_0 \mathrm{Re}\{\langle 0|P_z|1\rangle\langle 1|M_x|0\rangle\} + i\omega \mathrm{Im}\{\langle 0|P_z|1\rangle\langle 1|M_x|0\rangle\}}{\omega_0^2 - \omega^2 - 2i\omega\delta}. \tag{2}$$

Here, $\hbar$ is the Dirac constant, N is the total number of the dimers in 1 m^3, ϵ_0 is the electric constant, μ_0 is the magnetic constant, and $\omega = \omega_0 + 2\pi g\mu_B\ (H - H_{res})$ with the resonance field H_{res} of the Nambu-Goldstone magnon at 50 GHz. The dashed curve in the inset of Fig. 3(a) is the theoretical curve of $\Delta\alpha$ with $\mathrm{Re}\{\langle 0|P_z|1\rangle\} = -18.2\ \mu\mathrm{C/m}$, $\mathrm{Re}\{\langle 0|M_x|1\rangle\}$=2910A/m and $\mathrm{Im}\{\langle 0|P_z|1\rangle\} = \mathrm{Im}\{\langle 0|M_x|1\rangle\} = 0$ at H_{res}, calculated from the bond operator formulation, and $\delta = 2\pi \times 10.3$ GHz. The theoretical curve well agrees with the experimental result. Change in the microwave absorption by reversal of $\boldsymbol{M}$ is also observed. Figure 3(b) shows the ESR absorption spectra, observed in reversed magnetic fields. The absorption by triplon excitation at around 4.5 T, which is only electroactive, shows almost no change by reversal of $\boldsymbol{H}$, whereas large change in the absorption is seen for the Nambu-Goldstone magnon at 12.2 T, similar to the case by the reversal of $\boldsymbol{P}$. Compared to the reversal of $\boldsymbol{M}$ by $\boldsymbol{H}$, however, $\boldsymbol{P}$ can be more quickly reversed by $\boldsymbol{E}$. Figure 4(a) shows the change in the absorption coefficient at 12.2 T, where the excitation of the Nambu-Goldstone mode shows the absorption peak, by applying reversed electric fields $E = \pm 0.058$ MV/m. An immediate change in the absorption, along with the reversal of the polarity of $\boldsymbol{E}$, is observed. This change occurs due to reversal of $\boldsymbol{k}$ with stronger microwave absorption by the reversal of $\boldsymbol{P}$ as shown in Fig. 4(b). Thus, our result demonstrates that the rapid switching of the nonreciprocal directional microwave response by $\boldsymbol{E}$ is achieved. This switching of the nonreciprocal directional response occurs owing to 180° rotation of the antiferromagnetic domain around $\boldsymbol{H}$, caused by the reversal of $\boldsymbol{P}$. The rotation of the antiferromagnetic domain results in a reversal of the sign of the vector spin chirality $\boldsymbol{S}_l \times \boldsymbol{S}_r$ [23]. In $TlCuCl_3$, $\boldsymbol{P} = \tilde{C}(\boldsymbol{S}_l \times \boldsymbol{S}_r)$, so the reversal of the vector spin chirality causes a change in the sign of $\langle 0|P_z|1\rangle$ in χ_{zx}^{em}, thereby resulting in the reversal of the directional response by the sign change of $\Delta\alpha$ [see Eq. (1)]. The electric-field dependence of α, shown in the Supplemental Material [34], indicates that the application of $\boldsymbol{E}$ higher than the cohesive field $E_c \simeq 30$ V/mm is demanded for the directional switching of the microwave response. This $E_c \simeq 30$ V/mm is much lower than the cohesive fields in the known quantum multiferroic materials such as Ba_2CoGeO_7 [44] and $NiCl_2$-$4SC(NH_2)_2$ (DTN) [45]. Our recent experiments under pressure for $TlCuCl_3$ indicated that quantum fluctuation inherent in the magnon Bose-Einstein condensate is a key to reduce E_c [46]. Hence, we suggest that the quasi-one-dimensional quantum spin system Sul-Cu_2Cl_4, which was reported to undergo a ferroelectric helimagnetic state by the field-induced magnon BEC [47], can be another candidate for the "soft" nonreciprocal directional dichroism.

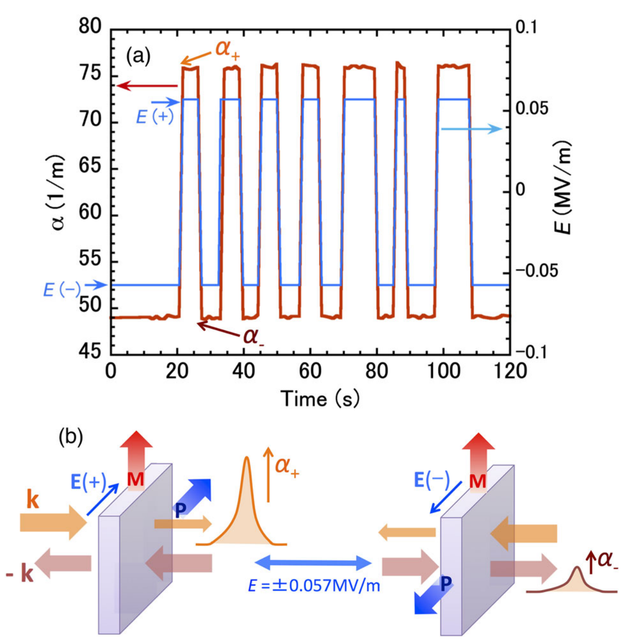

FIG. 4. (a) Change in the absorption coefficient by polarity reversal of the applied static electric field. The measurement was performed at 12.2 T, where the absorption peak due to the excitation of the Nambu-Goldstone mode is observed. (b) Switching of the nonreciprocal directional microwave response by $\boldsymbol{E}$. The propagation direction $\boldsymbol{k}$ with stronger microwave absorption is reversed by the reversal of $\boldsymbol{P}$.

As described above, this study demonstrated the high-speed directional control of microwave absorption by $\boldsymbol{E}$ in the ME multiferroic material with finite $\boldsymbol{P} \times \boldsymbol{M}$. The symmetry argument indicated that when $\boldsymbol{k} \cdot (\boldsymbol{P} \times \boldsymbol{M}) \neq 0$ is satisfied, not only electromagnetic wave absorption but also propagation of any kind of quantum particle is allowed to be nonreciprocal [1,2]. Voltage switchable directional propagation of various particles, including acoustic phonon or magnon, will open the way for developing new devices, such as an electrically controlled heat or spin current diode.

The authors thank N. Abe, K. Sakurai, and N. Watanabe for their kind help. This work was performed at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University (Project No. 19H0406) and was in part supported by the Grant-in-Aid for Scientific Research (Grants No. 17H01142, No. 17H02917, No. 17K05516, No. 19H01834) from MEXT Japan.

*shkimura@imr.tohoku.ac.jp

[1] D. Szaller, S. Bordács, and I. Kézsmárki, Phys. Rev. B **87**, 014421 (2013).
[2] Y. Tokura and N. Nagaosa, Nat. Commun. **9**, 3740 (2018).
[3] L. Feng, M. Ayache, J. Huang, Y.-L. Xu, M.-H. Lu, Y.-F. Chen, Y. Fainman, and A. Scherer, Science **333**, 729 (2011).
[4] K. Gallo, G. Assanto, K. R. Parameswaran, and M. M. Fejer, Appl. Phys. Lett. **79**, 314 (2001).
[5] T. Arima, J. Phys. Condens. Matter **20**, 434211 (2008).
[6] S. Toyoda, N. Abe, S. Kimura, Y. H. Matsuda, T. Nomura, A. Ikeda, S. Takeyama, and T. Arima, Phys. Rev. Lett. **115**, 267207 (2015).
[7] J. J. Hopfield and D. G. Thomas, Phys. Rev. Lett. **4**, 357 (1960).
[8] G. L. J. A. Rikken, C. Strohm, and P. Wyder, Phys. Rev. Lett. **89**, 133005 (2002).
[9] Y. Shimada, M. Matsubara, Y. Kaneko, J. P. He, and Y. Tokura, Appl. Phys. Lett. **89**, 101112 (2006).
[10] S. Yu, B. Gao, J. W. Kim, S.-W. Cheong, M. K. L. Man, J. Madéo, K. M. Dani, and D. Talbayev, Phys. Rev. Lett. **120**, 037601 (2018).
[11] M. Saito, K. Taniguchi, and T. Arima, J. Phys. Soc. Jpn. **77**, 013705 (2008).
[12] I. Kézsmárki, N. Kida, H. Murakawa, S. Bordács, Y. Onose, and Y. Tokura, Phys. Rev. Lett. **106**, 057403 (2011).
[13] Y. Takahashi, R. Shimano, Y. Kaneko, H. Murakawa, and Y. Tokura, Nat. Phys. **8**, 121 (2012).
[14] M. Kubota, T. Arima, Y. Kaneko, J. P. He, X. Z. Yu, and Y. Tokura, Phys. Rev. Lett. **92**, 137401 (2004).
[15] S. Bordács, I. Kézsmárki, D. Szaller, L. Demkó, N. Kida, H. Murakawa, Y. Onose, R. Shimano, T. Rõõm, U. Nagel, S. Miyahara, N. Furukawa, and Y. Tokura, Nat. Phys. **8**, 734 (2012).
[16] I. Kézsmárki, D. Szaller, S. Bordács, V. Kocsis, Y. Tokunaga, Y. Taguchi, H. Murakawa, Y. Tokura, H. Engelkamp, T. Rõõm, and U. Nagel, Nat. Commun. **5**, 3203 (2014).
[17] Y. Okamura, F. Kagawa, S. Seki, M. Kubota, M. Kawasaki, and Y. Tokura, Phys. Rev. Lett. **114**, 197202 (2015).
[18] Y. Nii, R. Sasaki, Y. Iguchi, and Y. Onose, J. Phys. Soc. Jpn. **86**, 024707 (2017).
[19] Y. Okamura, S. Seki, S. Bordács, A. Butykai, V. Tsurkan, I. Kézsmárki, and Y. Tokura, Phys. Rev. Lett. **122**, 057202 (2019).
[20] S. Miyahara and N. Furukawa, Phys. Rev. B **89**, 195145 (2014).
[21] Y. Iguchi, Y. Nii, and Y. Onose, Nat. Commun. **8**, 15252 (2017).
[22] T. Sato, N. Abe, S. Kimura, Y. Tokunaga, and T.-h. Arima, following Letter, Phys. Rev. Lett. **124**, 217402 (2020).
[23] S. Kimura, K. Kakihata, Y. Sawada, K. Watanabe, M. Matsumoto, M. Hagiwara, and H. Tanaka, Nat. Commun. **7**, 12822 (2016).
[24] S. Kimura, K. Kakihata, Y. Sawada, K. Watanabe, M. Matsumoto, M. Hagiwara, and H. Tanaka, Phys. Rev. B **95**, 184420 (2017).
[25] W. Shiramura, K. Takatsu, H. Tanaka, K. Kamishima, M. Takahashi, H. Mitamura, and T. Goto, J. Phys. Soc. Jpn. **66**, 1900 (1997).
[26] T. Nikuni, M. Oshikawa, A. Oosawa, and H. Tanaka, Phys. Rev. Lett. **84**, 5868 (2000).
[27] T. Giamarchi, C. Rüegg, and O. Tchernyshyov, Nat. Phys. **4**, 198 (2008).
[28] C. Rüegg, N. Cavadini, A. Furrer, H.-U. Güidel, K. Krämer, H. Mutka, A. Wildes, K. Habicht, and P. Vorderwisch, Nature (London) **423**, 62 (2003).
[29] M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **89**, 077203 (2002).
[30] M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. B **69**, 054423 (2004).
[31] S. Kimura, M. Matsumoto, M. Akaki, M. Hagiwara, K. Kindo, and H. Tanaka, Phys. Rev. B **97**, 140406(R) (2018).
[32] V. N. Glazkov, A. I. Smirnov, H. Tanaka, and A. Oosawa, Phys. Rev. B **69**, 184410 (2004).
[33] F. Yamada, T. Ono, H. Tanaka, G. Misguich, M. Oshikawa, and T. Sakakibara, J. Phys. Soc. Jpn. **77**, 013701 (2008).
[34] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.124.217401 for the calculations of the transition matrix elements and the electric-field dependence of the absorption coefficient, which includes Refs. [35–43].
[35] R. Shiina, H. Shiba, P. Thalmeier, A. Takahashi, and O. Sakai, J. Phys. Soc. Jpn. **72**, 1216 (2003).
[36] R. Shiina, M. Matsumoto, and M. Koga, J. Phys. Soc. Jpn. **73**, 3453 (2004).
[37] S. Sachdev and R. N. Bhatt, Phys. Rev. B **41**, 9323 (1990).
[38] T. Sommer, M. Vojta, and K. W. Becker, Eur. Phys. J. B **23**, 329 (2001).
[39] K. Penc, J. Romhányi, T. Rõõm, U. Nagel, Á. Antal, T. Fehér, A. Jánossy, H. Engelkamp, H. Murakawa, Y. Tokura, D. Szaller, S. Bordács, and I. Kézsmárki, Phys. Rev. Lett. **108**, 257203 (2012).
[40] J. Romhányi and K. Penc, Phys. Rev. B **86**, 174428 (2012).
[41] M. Matsumoto, T. Shoji, and M. Koga, J. Phys. Soc. Jpn. **77**, 074712 (2008).
[42] M. Matsumoto, J. Phys. Soc. Jpn. **83**, 084704 (2014).
[43] T. A. Kaplan and S. D. Mahanti, Phys. Rev. B **83**, 174432 (2011).
[44] H. Murakawa, Y. Onose, S. Miyahara, N. Furukawa, and Y. Tokura, Phys. Rev. Lett. **105**, 137202 (2010).
[45] V. S. Zapf, P. Sengupta, C. D. Batista, F. Nasreen, F. Wolff-Fabris, and A. Paduan-Filho, Phys. Rev. B **83**, 140405(R) (2011).
[46] K. Sakurai, S. Kimura, S. Awaji, M. Matsumoto, and H. Tanaka (to be published).
[47] F. Schrettle, S. Krohns, P. Lunkenheimer, A. Loidl, E. Wulf, T. Yankova, and A. Zheludev, Phys. Rev. B **87**, 121105(R) (2013).