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Rapid Communications

Electric dipole spin resonance in a quantum spin dimer system driven by magnetoelectric coupling

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In this Rapid Communication, we propose a mechanism for electric dipole active spin resonance caused by spin-dependent electric polarization in a quantum spin gapped system. This proposal was successfully confirmed by high-frequency electron spin resonance (ESR) measurements of the quantum spin dimer system $KCuCl_3$. ESR measurements by an illuminating linearly polarized electromagnetic wave reveal that the optical transition between the singlet and triplet states in $KCuCl_3$ is driven by an ac electric field. The selection rule of the observed transition agrees with the calculation by taking into account spin-dependent electric polarization. We suggest that spin-dependent electric polarization is effective in achieving fast control of quantum spins by an ac electric field.

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Dynamical coupling between spin and electric fields has attracted much attention in the diverse field of condensed matter physics. For instance, fast spin control by ac electric fields is a primary goal of spintronics [1] because an electric field rather than a magnetic field is easier to handle, and it is possible to address a confined area with a shorter length scale. The electromagnon, which drives the collective motion of spins by electric dipole active spin resonance in magnetoelectric multiferroic materials, is a striking manifestation of the coupling of spin to ac electric fields [2,3]. Magnetoelectric coupling, which arises from spin-dependent electric polarization, plays a key role in this phenomenon. An electromagon occurs because the normal oscillation of the ordered magnetic moments due to spin wave excitation causes the oscillation of the electric polarization via magnetoelectric coupling. Thus, an electromagnon has been observed in the long-range magnetic ordered state of multiferroic materials. In contrast, we noticed here that if spin-dependent electric polarization works properly, electric dipole spin resonance can occur even in a quantum spin gapped system, which has a nonmagnetic spin singlet ground state with no order. In this Rapid Communication, the observation of electric dipole spin resonance from the spin singlet to the triplet state in a spin gapped system, caused by spin-dependent electric polarization, is reported. Based on our observation, we suggest that spin-dependent electric polarization permits electric dipole spin resonance not only for the collective motion of a large number of spins, as found in mutiferroics, but also for an isolated spin monomer or dimer, which is strongly desired for fast spin manipulation necessary for spin-based quantum computation.

The transition between the spin singlet and triplet states is, in principle, forbidden in magnetic dipole transitions. Nonetheless, the transition was seen in many spin gapped systems, such as $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP), $CuGeO_3$, or $SrCu_2(BO_3)_2$, by means of high-frequency electron spin resonance (ESR) measurements, and its observation has been established as an advantageous probe to study these systems [4–8]. It was indicated that the mixing of the singlet and triplet states by an antisymmetric Dzyaloshinskii-Moriya interaction or staggered magnetic fields gives rise to a finite probability of magnetic dipole transitions between these states [9,10]. On the other hand, a phonon-assisted mechanism for the electric dipole transition was also suggested [11,12]. However, the detailed mechanism of the singlet-triplet transition is not yet understood. Thus, by taking into account spin-dependent electric polarization, we propose a mechanism for an electric dipole transition in spin gapped systems. Recent studies of spin-driven ferroelectricity in magnetoelectric mutiferroic materials revealed that an outer product of neighboring spins $\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j}$, namely, the vector spin chirality, can produce electric polarization $\boldsymbol{P}$ under the influence of a relativistic spin-orbit

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interaction [13–17]. The most general form of $\boldsymbol{P}$, which depends on $\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j}$, is expressed as

$$\boldsymbol{P} = \tilde{C}(\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j}), \tag{1}$$

where $\tilde{C}$ is a 3×3 matrix [17]. An important point is that the coupling between this $\boldsymbol{P}$ and the ac electric field $\boldsymbol{E}^\omega = \boldsymbol{E}e^{-i\omega t}$ can be regarded as a dynamical Dzyaloshinskii-Moriya interaction as follows,

$$\mathcal{H}_\mathrm{PE} = \boldsymbol{P} \cdot \boldsymbol{E}e^{-i\omega t} = \boldsymbol{d} \cdot (\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j})e^{-i\omega t}, \tag{2}$$

where $\boldsymbol{d}$ is

$$\boldsymbol{d} = \sum_{\nu,\mu} E_\nu C_{\nu\mu} \quad (\nu,\mu = x,y,z). \tag{3}$$

The Dzyaloshinskii-Moriya interaction $\mathcal{H}_\mathrm{DM} = \boldsymbol{d} \cdot (\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j})$ has its matrix elements between the singlet and triplet states. Therefore, according to Fermi's golden rule, $\mathcal{H}_\mathrm{PE}$ in Eq. (2) results in a finite probability of the electric dipole transition when $\hbar\omega$ equals the energy difference between these states, where $\hbar$ is the Dirac constant. In the following, from ESR measurements of the quantum spin dimer system $\mathrm{KCuCl_3}$ in an illuminating polarized electromagnetic wave, we will show that the singlet-triplet transitions are driven by ac electric fields of the electromagnetic wave, and the selection rule of the observed transitions agrees with the electric dipole active spin resonance that arises because of the spin-dependent electric polarization given in Eq. (1).

In $\mathrm{KCuCl_3}$, which crystallizes in the monoclinic space group $P2_1/c$ [18], antiferromagnetic dimers composed of $\mathrm{Cu^{2+}}$ ions with spin $S = 1/2$ are coupled by weak interdimer exchange interactions, forming a three-dimensional dimer network. Reflecting the dimer structure, the ground state of this compound is a spin singlet state with no order [19]. Figure 1 shows the frequency-field plot of the ESR resonance fields of $\mathrm{KCuCl_3}$ observed in our previous measurements in the magnetic field H parallel to [010] at 4.2 K [20,21]. Two sets of singlet-triplet transitions A and B were observed in $\mathrm{KCuCl_3}$. This is because two kinds of crystallographically different dimers I and II are involved in this compound [18], as shown in the inset of Fig. 2. In a weakly coupled dimer system such as $\mathrm{KCuCl_3}$, a spin triplet excited on a dimer propagates through the dimer network via the interdimer exchange interaction, forming an energy band with finite dispersion. In the case of $\mathrm{KCuCl_3}$, two bands, which correspond to the acoustic and optical modes of the triplet excitation, are superimposed because of the presence of two kinds of dimers [22,23]. Figure 2 shows the energy dispersion of the triplet excitation calculated by a bond operator theory with the exchange parameters listed in Refs. [21,22]. The experimentally observed dispersion was well reproduced by this calculation [22,23]. The singlet-triplet transitions A and B observed by the ESR measurements correspond to the excitations of the acoustic and optical modes, respectively, at the Γ point. As shown in Fig. 1, the resonance points in the disordered phase below $H_\mathrm{c} = 22$ T agree well with the Zeeman splitting of the triplet excitation, obtained by the theoretical calculation with $g = 2.05$. It should be mentioned that in $\mathrm{TlCuCl_3}$, which is isomorphous with $\mathrm{KCuCl_3}$, a spontaneous electric polarization, whose absolute value is proportional to $|\langle \boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j} \rangle|$ in the ground state, appears owing to the field-induced Bose-Einstein condensation of the triplet excitation [24,25]. Thus, we can expect the action of $\boldsymbol{P} = \tilde{C}(\boldsymbol{S}_\mathrm{i} \times \boldsymbol{S}_\mathrm{j})$ in $\mathrm{KCuCl_3}$.

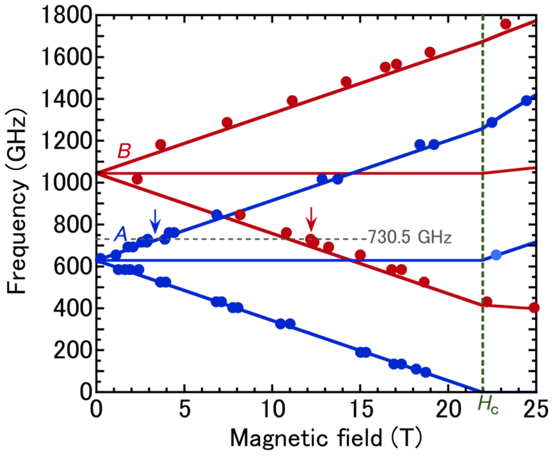

FIG. 1. Frequency-field plot of the ESR resonance fields observed in $\mathrm{KCuCl_3}$ for $H \parallel [010]$. Two sets of the singlet-triplet transition A and B are observed.

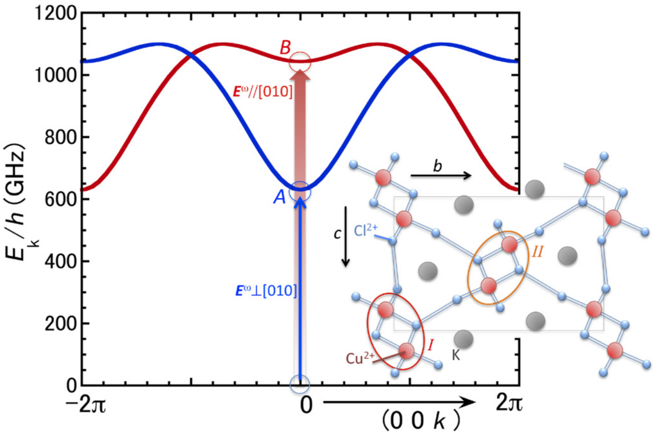

FIG. 2. Dispersion curves of triplet excitation in $\mathrm{KCuCl_3}$ calculated by the bond operator theory. The inset shows the crystal structure of $\mathrm{KCuCl_3}$. Two sets of triplet excitation modes are superimposed because of the presence of two kinds of crystallographically different dimers I and II in this compound. The singlet-triplet transitions A and B observed by the ESR measurements correspond to the excitation of these two modes from the spin singlet ground state at the Γ point. Our experiments reveal that the A and B modes are excited by the ac electric field for $\boldsymbol{E}^\omega \perp [010]$ and for $\boldsymbol{E}^\omega \parallel [010]$, respectively. The inset shows the crystal structure of $\mathrm{KCuCl_3}$.

High-field ESR measurements of $\mathrm{KCuCl_3}$ were performed at a frequency of 730.5 GHz at 4.2 K by utilizing a far infrared laser and a pulse magnet. The external magnetic field was applied along the [010] axis of a single crystal of $\mathrm{KCuCl_3}$. The ESR measurements were performed in both the Faraday and Voigt configurations. In the Voigt configuration, a wire grid polarizer was placed in front of the sample to polarize the incident electromagnetic wave. The single crystal of $\mathrm{KCuCl_3}$ was grown by the Bridgman method.

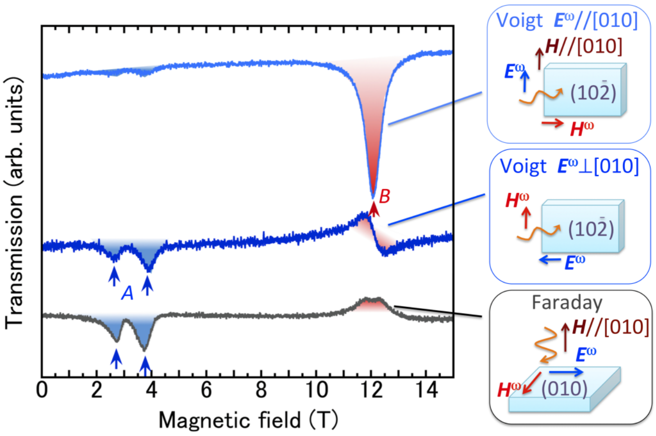

FIG. 3. ESR spectra observed in KCuCl$_3$ for $H \parallel [010]$. The upper and middle curves are the ESR spectra observed in the Voigt configuration with the ac electric fields $\boldsymbol{E}^{\omega} \parallel [010]$ and $\boldsymbol{E}^{\omega} \perp [010]$, respectively. The lower curve is the spectrum observed in the Faraday configuration with no fixed polarization direction of the incident electromagnetic wave. The spectra are normalized by the transmission at zero magnetic field.

Figure 3 shows the ESR spectra of KCuCl$_3$ for $H \parallel [010]$ observed at 730.5 GHz. The ESR signal of the A mode with a magnetic quantum number $m = -1$ and that of the B mode with $m = +1$ were observed at around 3 and 12 T, respectively. The ESR signal of the A mode splits into two peaks, though its origin is not clear at this moment. A strong ESR signal of the A mode is observed, whereas the signal of the B mode is scarcely seen in the Faraday configuration, in which the propagating vector $\boldsymbol{k}$ of the incident electromagnetic wave is parallel to the direction of the external static magnetic field. This result indicates that the A mode couples with the ac electric field $\boldsymbol{E}^{\omega}$ or magnetic field $\boldsymbol{H}^{\omega}$ oscillating perpendicular to [010]. Then, to clarify the selection rule of the observed ESR signals, we measured the polarization dependence of the ESR spectrum in the Voigt configuration with $\boldsymbol{k} \perp [010]$ by an illuminating linearly polarized electromagnetic wave. Figure 3 shows that a strong ESR signal of the A mode is observed for $\boldsymbol{E}^{\omega} \perp [010]$, whereas the signal almost disappears for $\boldsymbol{H}^{\omega} \perp [010]$ in the Voigt configuration. In contrast, the strong ESR signal of the B mode is only observed for $\boldsymbol{E}^{\omega} \parallel [010]$ in the Voigt configuration. These results clearly exhibit that both the A and B modes are excited by the ac electric field of the electromagnetic wave. Furthermore, the directions of the ac electric fields, which couple with the A and B modes, are orthogonal to each other. Namely, the A mode is excited for $\boldsymbol{E}^{\omega} \perp [010]$, whereas the B mode is excited for $\boldsymbol{E}^{\omega} \parallel [010]$. As described in the following, this selection rule is consistent with the electric dipole resonance due to spin-dependent electric polarization.

To calculate the probability of the electric dipole transition due to the spin-dependent electric polarization given in Eq. (1), we defined a Cartesian coordinate with the x, y, and z axes as follows: The x and z are parallel to [201] and [010], respectively, and the y is perpendicular to $(10\bar{2})$ in KCuCl$_3$. The recent theory indicated that the form of the matrix $\tilde{C}$ in Eq. (1) is restricted by a local crystallographic symmetry between the two spin sites [17]. However, in the case of the Cu^{2+} dimer in KCuCl$_3$, which possesses only an inversion center between the spins, the matrix $\tilde{C}$ can be an arbitral tensor of second rank [17]. The two spin dimers I and II in KCuCl$_3$ are transformed into each other by 2_1 helical symmetry operation along [010] (z) [18]. Therefore, the matrix $\tilde{C}_{\mathrm{II}}$ for the dimer II is given as

$$\tilde{C}_{\mathrm{II}} = \tilde{U}_z(\pi)\tilde{C}_{\mathrm{I}}\tilde{U}_z(\pi)^{-1} = \begin{bmatrix} C_{xx} & C_{xy} & -C_{xz} \\ C_{yx} & C_{yy} & -C_{yz} \\ -C_{zx} & -C_{zy} & C_{zz} \end{bmatrix}. \quad (4)$$

Here, $\tilde{C}_{\mathrm{I}}$ is the matrix for dimer I and $\tilde{U}_z(\pi)$ is the transformation matrix. These are expressed as

$$\tilde{C}_{\mathrm{I}} = \begin{bmatrix} C_{xx} & C_{xy} & C_{xz} \\ C_{yx} & C_{yy} & C_{yz} \\ C_{zx} & C_{zy} & C_{zz} \end{bmatrix} \quad (5)$$

and

$$\tilde{U}_z(\pi) = \begin{bmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (6)$$

The electric dipole transition probability W between the spin singlet and triplet states in KCuCl$_3$ is approximately given as

$$W \propto |\langle s|(\boldsymbol{P}_{\mathrm{I}} + \boldsymbol{P}_{\mathrm{II}})\boldsymbol{E}|t_{\mathrm{m}}\rangle|^2. \quad (7)$$

Here, $|s\rangle$ represents a wave function of the singlet state and $|t_{\mathrm{m}}\rangle$ is that of the triplet state of the system with $m = \pm 1$ or 0, whereas $\boldsymbol{P}_{\mathrm{I}}$ and $\boldsymbol{P}_{\mathrm{II}}$ are quantum mechanical operators of the electric polarization on the I and II dimers, respectively. In the case of the acoustic A mode, the matrix element $\langle s|(\boldsymbol{P}_{\mathrm{I}} + \boldsymbol{P}_{\mathrm{II}})\boldsymbol{E}|t_{\mathrm{m}}\rangle$ can be rewritten as

$$\langle s|(\boldsymbol{P}_{\mathrm{I}} + \boldsymbol{P}_{\mathrm{II}})\boldsymbol{E}|t_{\mathrm{m}}\rangle \propto \langle 0,0|(\tilde{C}_{\mathrm{I}} + \tilde{C}_{\mathrm{II}})(\boldsymbol{S}_1 \times \boldsymbol{S}_2)\boldsymbol{E}|1,m\rangle, \quad (8)$$

where $|0,0\rangle$ is the singlet and $|1,m\rangle$ is the triplet state of a dimer. From Eqs. (4) and (5), the matrix element for $m = -1$ is calculated to be

$$\begin{aligned} &\langle 0,0|(\tilde{C}_{\mathrm{I}} + \tilde{C}_{\mathrm{II}})(\boldsymbol{S}_1 \times \boldsymbol{S}_2)\boldsymbol{E}|1,-1\rangle \\ &\quad = \{(iC_{xx} + C_{xy})E_x + (iC_{yx} + C_{yy})E_y\}/\sqrt{2}. \end{aligned} \quad (9)$$

This result indicates that the A mode with $m = -1$ is excited by the electric fields E_x and E_y, namely, by $\boldsymbol{E}^{\omega}$ perpendicular to [010], thus showing agreement with the experimental results. On the other hand, in the case of the optical B mode, a transfer of the triplet excitation from the dimer I to II causes a change in the phase factor of its wave function by π [26]. Therefore, the matrix element $\langle s|(\boldsymbol{P}_{\mathrm{I}} + \boldsymbol{P}_{\mathrm{II}})\boldsymbol{E}|t_{\mathrm{m}}\rangle$ for the B mode is written as

$$\langle s|(\boldsymbol{P}_{\mathrm{I}} + \boldsymbol{P}_{\mathrm{II}})\boldsymbol{E}|t_{\mathrm{m}}\rangle \propto \langle 0,0|(\tilde{C}_{\mathrm{I}} - \tilde{C}_{\mathrm{II}})(\boldsymbol{S}_1 \times \boldsymbol{S}_2)\boldsymbol{E}|1,m\rangle. \quad (10)$$

In the case of $m = +1$, the matrix element is calculated as

$$\langle 0,0|(\tilde{C}_{\mathrm{I}} - \tilde{C}_{\mathrm{II}})(\boldsymbol{S}_1 \times \boldsymbol{S}_2)\boldsymbol{E}|1,+1\rangle = (iC_{zx} + C_{zy})E_z/\sqrt{2}. \quad (11)$$

Therefore, the B mode with $m = +1$ should be excited by the electric field E_z, namely, by $\boldsymbol{E}^{\omega}$ parallel to [010]. This result is also consistent with the experimental observation. It should be mentioned that for the static magnetic field applied perpendicular to [010], which breaks 2_1 symmetry, the situation becomes rather complex. In this case, a calculation similar to the above shows that the ac electric fields, which couple to the A and B modes, are not orthogonal to each other. From the angular dependence of the static electric polarization, which is expected to appear above H_c for KCuCl$_3$, one can determine the parameters of the matrix $\tilde{C}$. Then, a more quantitative evaluation of our mechanism will be achieved by a comparison between the calculation with these parameters and the experimental results of angular dependence measurements in the ESR intensity for the singlet-triplet transition.

From the above discussion, we have shown that the singlet-triplet transitions in KCuCl$_3$ are driven by ac electric fields, and the observed electric dipole transitions can be explained by spin-dependent electric polarization. Based on symmetry considerations, it was suggested that the electric polarization, which depends on $\boldsymbol{S}_i \times \boldsymbol{S}_j$, can appear regardless of the local structural symmetry between the two spin sites [17]. Therefore, electric dipole spin resonance by spin-dependent electric polarization is universal for spin gapped systems. It is possible that the singlet-triplet transitions that were previously observed in various spin gapped materials are induced by this polarization mechanism. Our results suggest that spin resonance by spin-dependent electric polarization can be implemented to achieve fast manipulation of highly entangled spin states in a magnetic dimer by ac electric fields. This kind of a spin resonance technique is also applicable for the manipulation of a single spin. A spin located at a noncentrosymmetric environment can generate an electric polarization expressed as follows [27],

$$p_s^{\alpha} = \sum_{\beta,\gamma} K_{\beta\gamma}^{\alpha} S^{\beta} S^{\gamma}. \tag{12}$$

Here, p_s^{α} and S^{α} ($\alpha = x,y,z$) are the α components of the electric dipole and spin operators, respectively, and K^{α} is a coefficient of the tensor. As described in Ref. [27], the electric dipole given in Eq. (12) can cause a spin resonance by the ac electric fields for a spin $S \geqslant 1$ because p_s^{α} has finite matrix elements between the spin multiples via the electric quadrupoles defined by the product of spin operators. The spin resonance by the ac electric fields observed in manganese defects in polar ZnO [28] and in the silicon carbide color center [29] can be understood in terms of electric dipole spin resonance by the electric polarization p_s^{α}. Electric dipole spin resonance by spin-dependent electric polarization will open a different way for the fast manipulation of quantum spins by ac electric fields.

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[26] The dimers I and II in $\mathrm{KCuCl_3}$ can be transformed into each other not only by the 2_1 symmetry but also by the c-glide symmetry operation. In Refs. [22, 23], the latter is adopted for the transformation between the dimers. In this case, the A and B modes correspond to the triplet excitation at $Q = (010)$ and (000), respectively, in the extended zone scheme, whereas in the case of the former, which is adopted in this Rapid Communication, the correspondences are vice versa. However, the matrix elements for the transition probabilities of the A and B modes, given in Eqs. (8) and (11), are the same for the former and the latter because the signs of the matrix $\tilde{C}_{\mathrm{II}}$ in these two cases are opposite to each other owing to its axial nature.

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