Magnonic Superradiant Phase Transition

Motoaki Bamba
Department of Physics, Kyoto University, Kyoto 606-8502, Japan and
PRESTO, Japan Science and Technology Agency, Kawaguchi 332-0012, Japan

Xinwei Li
Department of Electrical and Computer Engineering, Rice University, Houston 77005, USA

Nicolas Marquez Peraca
Department of Physics and Astronomy, Rice University, Houston 77005, USA

Junichiro Kono
Department of Electrical and Computer Engineering, Rice University, Houston 77005, USA
Department of Material Science and NanoEngineering, Rice University, Houston 77005, USA and
Department of Physics and Astronomy, Rice University, Houston 77005, USA
(Dated: July 28, 2020)

We show that the low-temperature phase transition in ErFeO$_3$ that occurs at a critical temperature of $\sim 4$ K can be described as a magnonic version of the superradiant phase transition (SRPT). The role of photons in the quantum-optical SRPT is played by Fe$^{3+}$ magnons, while that of two-level atoms is played by Er$^{3+}$ spins. Our spin model, which is reduced to an extended Dicke model, takes into account the short-range, direct exchange interactions between Er$^{3+}$ spins in addition to the long-range Er$^{3+}$–Er$^{3+}$ interactions mediated by Fe$^{3+}$ magnons. By using realistic parameters determined by recent terahertz magnetospectroscopy and magnetization experiments, we demonstrate that it is the cooperative, ultrastrong coupling between Er$^{3+}$ spins and Fe$^{3+}$ magnons that causes the phase transition. This work thus proves ErFeO$_3$ to be a unique system that exhibits a SRPT in thermal equilibrium, in contrast to previous observations of laser-driven non-equilibrium SRPTs.

I. INTRODUCTION

In 1973, it was proposed [1, 2] that a static transverse electromagnetic field (a photon field) and a static polarization (a matter field) spontaneously appear in thermal equilibrium, when the photon–matter coupling strength exceeds a certain threshold, entering the so-called ultrastrong coupling regime [3–5]. This phenomenon has come to be known as the superradiant phase transition (SRPT), or the Dicke phase transition, since the Dicke model (originally developed for the phenomenon of superradiance [6]) was used in the theoretical calculations [1, 2].

While the focus of optical science has traditionally been on non-equilibrium excited-state dynamics, a unique aspect of the SRPT is that it is concerned with the thermal-equilibrium state of a light–matter coupled system. Non-equilibrium SRPTs have been demonstrated in cold atom systems driven by laser light [7, 8], but realization of the SRPT in true thermal equilibrium has been challenging. The existence of an analog of the superradiance [6] was used in the theoretical calculations [1, 2].

SRPT is impossible to realize in systems described by the minimal-coupling Hamiltonian, i.e., charged particles (without spins) interacting with electromagnetic fields. Since the classical treatment of the electromagnetic fields used in proofs of such no-go theorems can be justified only in limited situations [2, 12–16], proposals of counter-examples against the no-go theorems and criticisms against the counter-examples have been repeated in the research history of the SRPT [17–27].

One way to evade the no-go theorems is by introducing another degree of freedom, such as spin [11]. For example, it has been shown that the Rashba spin–orbit coupling can cause a paramagnetic instability in an ultrastrongly coupled system between a cyclotron resonance and a cavity photon field, implying a SRPT [27]. Another way is to utilize various types of interactions in magnetic materials, which cannot be described by the minimal-coupling Hamiltonian. Ultrastrong photon–magnon coupling has been reported [28–32], but evidence for a SRPT has not been achieved. A variety of phase transitions exist in magnetic systems, and it is conceivable that some of the known phase transitions can be understood as a realization of the SRPT. In this context, it is noteworthy that the problem of ultrastrong coupling between Er$^{3+}$ spins and Fe$^{3+}$ magnons in ErFeO$_3$ has been mapped to the Dicke model in a recent experimental study [33]. In this extraordinary situation of matter–matter ultrastrong coupling, the role of photons in the usual Dicke model is played by magnons.

In this paper, we theoretically show that the phase
transition in ErFeO$_3$ with a critical temperature ($T_c$) of $\sim 4$ K, known as the low-temperature phase transition (LTPT), is a magnonic SRPT, i.e., an analog of the SRPT where the spin-magnon coupling, compared to that obtained only by the direct Er$^{3+}$-Er$^{3+}$ exchange interactions. Also, we observed that the critical temperature $T_c$ of the LTPT is enhanced by the Er$^{3+}$-magnon coupling even in the absence of direct Er$^{3+}$-Er$^{3+}$ interactions. These results demonstrate that ErFeO$_3$ is a unique physical system in which a SRPT can be experimentally realized in thermal equilibrium.

This paper is organized as follows. We first review the SRPT in the Dicke model and the LTPT in ErFeO$_3$ in Secs. II and III respectively. Our spin model of ErFeO$_3$ is described in Sec. IV. Calculated phase diagrams are shown in Sec. V. For discussing the analogy with the SRPT, an extended version of the Dicke model is derived from the spin model in Sec. VI. The analogy is fully discussed in Sec. VII. Section VIII summarizes our findings.

Appendix A shows the details of our mean-field calculation. In Appendix B we show how the number of parameters in the spin model can be reduced by considering the low-temperature spin configuration in ErFeO$_3$. In Appendix C spin resonance frequencies are numerically calculated by the mean-field method and by the extended Dicke Hamiltonian for comparing these methods as well as for determining the parameters. In Appendix D the actual values of the parameters are listed. In Appendix E the magnon quantization procedure for the Fe$^{3+}$ subsystem is described. In Appendix F we discuss small differences of the phase diagrams between that obtained by the mean-field method and that obtained by the extended Dicke Hamiltonian.

II. SUPERRADIANT PHASE TRANSITION IN THE DICKE MODEL

The SRPT was first suggested in 1973 by Hepp and Lieb [1] and has been extensively discussed based on the Dicke model [6] expressed as

$$\hat{H}_{\text{Dicke}}(T) = \text{Tr} [e^{-\hat{H}_{\text{Dicke}}/(k_B T)}]$$

in the thermodynamic limit, $N \to \infty$, can be approximately evaluated by replacing the trace over the photonic variables with an integral over coherent states $|\sqrt{N}\hat{a}\rangle$ ($\hat{a} \in \mathbb{C}$; giving $\langle \sqrt{N}\hat{a}\rangle = \sqrt{N}\langle \sqrt{N}\hat{a}\rangle$) as

$$\bar{Z}_{\text{Dicke}}(T) = \int \frac{d^2 \hat{a}}{\pi N} \text{Tr} [e^{-\hat{H}_{\text{Dicke}}(\hat{a})/(k_B T)}]$$

where we defined an effective Hamiltonian

$$\hat{H}_{\text{Dicke}}(\hat{a}) = N \omega_{\text{ph}} |\hat{a}|^2 + \omega_{\text{ex}} \hat{S}_x + i 2g (\hat{a}^* - \hat{a}) \hat{S}_z,$$

an action

$$\bar{S}_{\text{Dicke}}(\hat{a}, T) \equiv -(k_B T \ln \text{Tr} [e^{-\hat{H}_{\text{Dicke}}(\hat{a})/(k_B T)}]),$$

and an effective Hamiltonian per atom

$$\hat{H}_{\text{Dicke}}(\hat{a}) = \frac{\omega_{\text{ex}}}{2} \hat{S}_x + ig (\hat{a}^* - \hat{a}) \hat{S}_z.$$

The normalized expectation value $\langle \hat{a} \rangle = \langle \hat{a} \rangle / \sqrt{N}$ of the annihilation operator of a photon at temperature $T$ can be determined for minimizing the action, i.e., $\partial \bar{S} / \partial \text{Re} |\hat{a}| = 0$ and $\partial \bar{S} / \partial \text{Im} |\hat{a}| = 0$. We find that $\hat{a}$ acquires a nonzero value below $T_c$ when $4g^2 > \omega_{\text{ph}} \omega_{\text{ex}}$ is satisfied ($\sqrt{N}\hat{a}$ gives a finite electric (displacement) field or vector potential even in the thermodynamic limit, $N \to \infty$, if the atomic density is fixed). The above approximation is justified if the free energy $\mathcal{F}_{\text{Dicke}}(T) \equiv - (k_B T / N) \ln \bar{Z}_{\text{Dicke}}(T)$ per atom satisfies $\hbar \omega_{\text{ph}} / N \ll |\mathcal{F}_{\text{Dicke}}(T)|$ in the thermodynamic limit [12, 13, 15, 16].

Based on the above semiclassical calculation scheme, Rzażewski et al. derived no-go theorems starting from the minimal-coupling Hamiltonian in the long-wavelength approximation in 1979 [12] and in the general case in 1981 [13]. However, since the proof had the above-mentioned limitation of validity due to the semiclassical treatment employed, the presence of the SRPT in the minimal-coupling Hamiltonian is still controversial [17, 27].
III. LOW-TEMPERATURE PHASE TRANSITION IN ErFeO$_3$

Resonance frequencies of magnons, quanta of spin waves, in magnetic materials have provided rich information on the spin configurations of materials. Softening (i.e., decrease of resonance frequency) of magnon modes has been discussed in connection with magnetic phase transitions. Magnons also provide a platform for electrodynamics studies both in the classical and quantum regimes [23, 33-35, 42].

Due to the coupling (amplitude exchange) between a magnon in magnetic materials and a photon (electromagnetic wave) in a cavity, which can be described by the last term in the Dicke model [Eq. (1)], we can observe anticrossing on their resonance frequencies. If the anticrossing frequency is higher than dephasing rates (broadening or linewidth), we can exchange the amplitude coherently between the magnon and photon modes. Such a regime is called the strong coupling regime, and it attracts much attention for coherent transfer of quantum information between different media of quanta [23, 24, 55, 38] and for magnon detection [32, 39, 42].

On the other hand, the anticrossing frequency ($2g$) can be comparable to the original resonance frequency ($\omega_{ph}$) of photons, magnons, or other material excitations ($\omega_{m}$), i.e., the ultrastrong coupling regime [3-5]. Ultrastrong photon–magnon coupling has been reported for a yttrium-iron-garnet (YIG) sphere embedded in a cavity with a resonance frequency in the gigahertz (GHz) region [25-62]. Recently, $g/\omega \sim 0.46$ has been achieved for the purpose of detecting dark matter (galactic axions) [32, 34]. Ultrastrong spin–magnon [33] and magnon–magnon [40, 41] coupling have also been observed. Among such magnetic materials with ultrastrong coupling, ErFeO$_3$ is a candidate material showing the magnonic SRPT as explained below.

As shown in Fig. 1(a) at $T_c \sim 4$ K, ErFeO$_3$ shows the LTPT [43, 44], a second-order phase transition where Er$^{3+}$ spins are ordered antiferromagnetically along the $c$ axis together with a rotation of the Fe$^{3+}$ antiferromagnetism (AFM) vector $S^A - S^B$ in the $bc$ plane due to the Er$^{3+}$–Fe$^{3+}$ exchange interactions.

In the absence of those exchange interactions, as in Fig. 1(a), Fe$^{3+}$ spins are ordered antiferromagnetically just along the $c$ axis with a slight canting to the $a$ axis in the ground state of the Fe$^{3+}$ subsystem. When we consider that the magnon excitation in this Fe$^{3+}$ subsystem corresponds to the photon excitation in the electromagnetic vacuum, the rotation of the Fe$^{3+}$ AFM vector (at $T < T_c$, as shown in Fig. 1(b)) means a spontaneous appearance of magnons, which corresponds to the appearance of photons (a static electromagnetic field) in the ordinary SRPT, in thermal equilibrium. The ordering of Er$^{3+}$ spins correspond to the spontaneous appearance of an atomic field (a polarization) in the SRPT. In this way, we can expect that there is an analogy between the LTPT in ErFeO$_3$ and the SRPT in the Dicke model.

A theoretical model for describing the LTPT was proposed by Vitebskii and Yablonskii in 1978 [15]. The ratio between the Er$^{3+}$–Er$^{3+}$ and Er$^{3+}$–Fe$^{3+}$ interaction strengths was theoretically investigated by Kadomtseva, Krynetskii, and Matveev in 1980 [46]. They also mentioned the analogy between the LTPT and the cooperative Jahn–Teller transition [47, 48]. The analogy between the cooperative Jahn–Teller transition and the SRPT was discussed by Loos in 1984 [49] and also by Larson in 2008 [50]. Loos also suggested a magnetic system consisting of coupled ferromagnetic and paramagnetic spins, such as rare-earth iron garnets, as a candidate system for observing the above analogy. However, this analogy has not yet been verified experimentally.

ErFeO$_3$ can be modeled as coupled antiferromagnetic and paramagnetic (or antiferromagnetic) spins. In the above-mentioned studies, unfortunately, the analogy between the LTPT and the SRPT was not directly drawn either theoretically or experimentally. In 2018, the $\sqrt{N}$-dependence ($N$ is the Er$^{3+}$ density) of the anticrossing frequency, or vacuum Rabi splitting ($2g$), between paramagnetic Er$^{3+}$ spins and a Fe$^{3+}$ magnon mode was confirmed experimentally at $T > T_c$ by Li et al. [32]. This $\sqrt{N}$-dependence, the Dicke cooperativity, can be taken as evidence that the coupling between the Er$^{3+}$ spin ensemble and the Fe$^{3+}$ magnon mode is cooperative, well described by the Dicke model or its extension.

As pointed out in the early studies [15, 46], it is important to take into account not only the Er$^{3+}$–magnon coupling but also the antiferromagnetic Er$^{3+}$–Er$^{3+}$ exchange interactions for discussing the LTPT in ErFeO$_3$. Therefore, we must extend the Dicke model to fully describe the LTPT, because Eq. (1) does not include the atom–atom interactions that correspond to the Er$^{3+}$–Er$^{3+}$ exchange interactions. In our experiments [33], while the Er$^{3+}$–magnon coupling was clearly observed through terahertz...
absorption spectroscopy, the influence of the Er$^{3+}$–Er$^{3+}$ interactions remained unclear.

We determined the parameters in our spin model (Sec. IV), including the Er$^{3+}$–Er$^{3+}$ interactions, through terahertz spectra that we observed previously as well as the phase diagrams obtained in a recent magnetization study. The parameter estimation method is discussed in Appendixes, and we focus on the analogy between the LTPT and SRPT in the following sections.

IV. SPIN MODEL

Each unit cell of ErFeO$_3$ contains four Er$^{3+}$ ions and four Fe$^{3+}$ ions. The four Fe$^{3+}$ spins, each of which has an angular momentum of $\hbar S = (5/2)\hbar$, are oriented in different directions with each other even in the absence of an external DC magnetic field. However, it is known that the Fe$^{3+}$ spin resonances (magon modes) are well described by considering only two spins $S^A_1/B_1$, each of which in fact consists of two real Fe$^{3+}$ spins but is usually treated as a single spin with $S = 5/2$. In such a two-sublattice model of Fe$^{3+}$, as depicted in Fig. 1(a), at $T_c < T \lesssim 90$ K, the two spins $S^A_1/B_1$ are ordered antiferromagnetically along the $c$ axis, while they are slightly canted toward the $a$ axis and show a weak magnetization (the Fe$^{3+}$ spins show the so-called spin-reorientation transition at 90 K $\lesssim T \lesssim 100$ K [34, 43, 44]). On the other hand, Er$^{3+}$ spins are paramagnetic at $T > T_c$, and they are directed along the $a$ axis by the weak Fe$^{3+}$ magnetization. This phase is called the $\Gamma_2$ phase.

At $T < T_c$, as shown in Fig. 1(b), when we use a two-sublattice model also for Er$^{3+}$ spins, they are ordered antiferromagnetically along the $a$ axis, with a canting toward the $a$ axis due to the Fe$^{3+}$ magnetization. Simultaneously, the Fe$^{3+}$ AFM vector gradually rotates in the $bc$ plane. The rotation angle measured from the $c$ axis, $\varphi$, has been estimated to be 49° at $T = 0$ K [44]. This low-temperature phase is called the $\Gamma_{12'}$ phase.

In the following, we describe our spin model for Er$_x$Y$_{1-x}$FeO$_3$ ($0 \leq x \leq 1$), which is consistent with our previous experimental study [52]. The $x$-dependence is described in more detail in Appendix C. The replacement of Er$^{3+}$ ions by non-magnetic Y$^{3+}$ ones simply reduces the density of the rare-earth (Er$^{3+}$) spins without changing the crystal structure or the magnetic configuration of Fe$^{3+}$ spins in the $\Gamma_2$ phase [33, 52].

Our Hamiltonian for the spins in Er$_x$Y$_{1-x}$FeO$_3$ consists of three parts:

$$\mathcal{H} = \mathcal{H}_{\text{Fe}} + \mathcal{H}_{\text{Er}} + \mathcal{H}_{\text{Fe-Er}},$$

where $\mathcal{H}_{\text{Fe}}$, $\mathcal{H}_{\text{Er}}$, and $\mathcal{H}_{\text{Fe-Er}}$ are the Hamiltonians of the Fe$^{3+}$ spins, Er$^{3+}$ spins, and Er$^{3+}$–Fe$^{3+}$ interactions, respectively.

As explained above, we employ the two-sublattice model for Fe$^{3+}$ spins by following Herrmann’s model [53] and our previous studies [53, 54]. The Hamiltonian of Fe$^{3+}$ spins is described as

$$\hat{H}_{\text{Fe}} = \sum_{s=A,B} \sum_{i=1}^{N_0} \mu_B \hat{S}^s_i \cdot \mathbf{g}^s \cdot B^{\text{DC}} + J_{\text{Fe}} \sum_{\text{n.n.}} \hat{S}^A_i \cdot \hat{S}^B_{i'},$$

$$- D_y^{\text{Fe}} \sum_{\text{n.n.}} \left( \hat{S}^A_{i,x} \hat{S}^B_{i',x} - \hat{S}^B_{i'} \hat{S}^A_{i,x} \right),$$

$$- \sum_{i=1}^{N_0} \left( A_x \hat{S}^A_{i,x}^2 + A_y \hat{S}^A_{i,y}^2 + A_z \hat{S}^A_{i,z} \right),$$

$$- \sum_{i=1}^{N_0} \left( A_x \hat{S}^B_{i,x}^2 + A_y \hat{S}^B_{i,y}^2 - A_z \hat{S}^B_{i,z} \right).$$

Here, $\hat{S}^{A/B}_i$ is the operator of the Fe$^{3+}$ spin with $S = 5/2$ at the $i$-th site in the A/B sublattice. $\sum_{\text{n.n.}}$ means a summation over all the nearest neighbor couplings. The number of nearest neighbors is $z_{\text{Fe}} = 6$.

$N_0$ is the number of Fe$^{3+}$ spins in each sublattice and is equal to the number of unit cells in ErFeO$_3$. Then, there are in total $2N_0$ spins representing the Fe$^{3+}$ subsystem. $\mu_B$ is the Bohr magneton, and

$$\mathbf{g}^s = \begin{pmatrix} g_{x}^s & 0 & 0 \\ 0 & g_{y}^s & 0 \\ 0 & 0 & g_{z}^s \end{pmatrix}$$

is the $g$-factor tensor for the Fe$^{3+}$ spins. In the following, the $g$-factor of free electron spin is expressed as $\mathbf{g}$. $B^{\text{DC}}$ is an external DC magnetic flux density. $J_{\text{Fe}}$ and $D_y^{\text{Fe}}$ are, respectively, the strengths of isotropic and Dzyaloshinskii-Moriya-type exchange interaction strengths between Fe$^{3+}$ spins. $A_x$, $A_y$, and $A_z$ are the energies expressing the magnetic anisotropy of Fe$^{3+}$ spins.

While we expressed the Er$^{3+}$ subsystem by a single spin lattice for the paramagnetic Er$^{3+}$ spins ($T > T_c$) in our previous studies [33, 52], we use a two-sublattice model for Er$^{3+}$ spins in this paper in order to describe the Er$^{3+}$–Er$^{3+}$ exchange interaction and the LTPT. The Hamiltonian of Er$^{3+}$ spins is expressed as

$$\hat{H}_{\text{Er}} = - \sum_{s=A,B} \sum_{i=1}^{N_0} \hat{\mu}^s_i \cdot B^{\text{DC}} + J_{\text{Er}} \sum_{\text{n.n.}} \hat{R}^A_i \cdot \hat{R}^B_{i'},$$

Here, $\hat{R}^{A/B}_i$ is the operator of rare-earth (Er$^{3+}$ or Y$^{3+}$) spin at the $i$-site in the A/B sublattice. For Er$_x$Y$_{1-x}$FeO$_3$, the rare-earth spins are represented randomly as $(s = A,B)$

$$\hat{R}^s_i = \begin{cases} \hat{\sigma}^s_{+} & \text{for Er}^{3+} \\ 0 & \text{for Y}^{3+} \end{cases}$$

We describe each Er$^{3+}$ spin by a Pauli operator $\hat{\sigma}^s_{+}$. The Y$^{3+}$ ion is nonmagnetic and $\hat{R}^s_i$ is replaced by $0$. The
first term in Eq. (11) represents the Zeeman effect, and the magnetic moment is expressed in terms of anisotropic $g$-factors, $\mu_{ij}^{Er}$, for the Er$^{3+}$ spins as

$$\hat{\mu}_i = -\frac{1}{2}\mu_B g_i^x \hat{R}_i^x + g_i^y \hat{R}_i^y + g_i^z \hat{R}_i^z = -\frac{1}{2}\mu_B g_i^{Er} \cdot \hat{R}_i^i. \quad (13)$$

The factor 1/2 is added since $(1/2)\sigma_i^z$ corresponds to a spin-$\frac{1}{2}$ operator theoretically. We defined the $g$-factor tensor for Er$^{3+}$ spins as

$$g_i^{Er} = \begin{pmatrix} g_i^x & 0 & 0 \\ 0 & g_i^y & 0 \\ 0 & 0 & g_i^z \end{pmatrix}. \quad (14)$$

The second term in Eq. (11) represents the Er$^{3+}$–Er$^{3+}$ exchange interaction with a strength of $J_i^{Er}$. Since Er$^{3+}$ ions are diluted in Er$_3$Y$_{1-x}$FeO$_3$, the number of nearest neighbor Er$^{3+}$ spins is effectively given by

$$2g_{Er} = 6x. \quad (15)$$

In a similar manner to our previous studies [33, 54], we describe the Er$^{3+}$–Fe$^{3+}$ interaction Hamiltonian as

$$\tilde{\mathcal{H}}_{Er-Fe} = \sum_{i=1}^{N_0} \sum_{s,s' = A,B} \left[ J_i^{A,B} \cdot \hat{S}_i^s + D_i^{s,s'} \cdot (\hat{R}_i^s \times \hat{S}_i^{s'}) \right]. \quad (16)$$

In our model, the Er$^{3+}$–Fe$^{3+}$ interaction is closed in each unit cell, i.e., the Er$^{3+}$ and Fe$^{3+}$ spins in the same unit cell interact with each other but do not interact with the spins in other unit cells. $J$ and $D^{s,s'}$ are the strengths of the isotropic and antisymmetric exchange interactions, respectively. Considering the spin configuration at $T < T_c$ with no external DC magnetic field (see more details in Appendix D), we assume that $D^{s,s'}$ are expressed in terms of two values $D_x$ and $D_y$ as

$$D_{A,A}^{x,y} = (D_x, D_y, 0)^t, \quad (17a)$$
$$D_{A,B}^{x,y} = (-D_x, -D_y, 0)^t, \quad (17b)$$
$$D_{B,A}^{x,y} = (-D_x, D_y, 0)^t, \quad (17c)$$
$$D_{B,B}^{x,y} = (D_x, -D_y, 0)^t. \quad (17d)$$

Note that, as explained in Appendix A, we assume that the $y$ components, $R_{i,y}^{A,B}$, of the Er$^{3+}$ spins are not influenced by the Er$^{3+}$–Fe$^{3+}$ interaction by implicitly considering a higher energy potential than the Er$^{3+}$–Fe$^{3+}$ interaction strengths $J$ and $D^{s,s'}$ along the $b$ axis. This assumption is required for properly describing the LTPT.

The actual values of the parameters that appears in our spin model are shown in Appendix D together with a description of how we determined them.

V. PHASE DIAGRAMS

In this section, we show thermal-equilibrium (averaged) values of the Er$^{3+}$ spins $\sigma_{A,B}^{1/2}$ and of the Fe$^{3+}$ spins $S_{A,B}^{1/2}$ in the zero-wavenumber (infinite-wavelength) limit by a mean-field method. Details of the mean-field method are given in Appendix A. Since we simply consider a homogeneous external DC magnetic flux density, $B_{DC}$, in this paper, $\sigma_{A,B}^{1/2}$ and $S_{A,B}^{1/2}$ are independent of the site index $i$.

Figures 2(a), (b), and (c) show calculated phase diagrams as a function of temperature, $T$, and external DC magnetic field, $B_{DC}$, applied along the $a$, $b$, and $c$ axes, respectively. We plot the difference $|\sigma_i^A - \sigma_i^B|$ of the $z$ components of the thermal-equilibrium values of Er$^{3+}$ spins mapped with red color. The bold solid lines represent the phase boundaries. The external DC magnetic field is varied from zero to positive or negative values at a fixed temperature. Since ErFeO$_3$ shows a weak magnetization along the $a$ axis, the critical field depends on whether the field is parallel or antiparallel to the magnetization in Fig. 2(a).

In Fig. 3, we plot the thermal-equilibrium values of the Er$^{3+}$ and Fe$^{3+}$ spins in the absence of an external DC magnetic field as a function of temperature. The LTPT, i.e., the antiferromagnetic ordering of Er$^{3+}$ spins along the $c$ axis and the rotation of the Fe$^{3+}$ spins in
along the $\sigma$ axis as $\sigma = \sigma^A - \sigma^B$ spontaneously appears below the critical temperature $T_c = 4.0 \text{ K}$, i.e., the $\text{Er}^{3+}$ spins are antiferromagnetically ordered along the $c$ axis. They show a magnetization along the $a$ axis as $\sigma_a = \sigma^A_a / \sigma^B_a$, due to the $\text{Er}^{3+}-\text{Fe}^{3+}$ exchange interaction with the weak $\text{Fe}^{3+}$ magnetization along the $a$ axis, while $\sigma_y = \sigma^A_y / \sigma^B_y = 0$. As shown in Fig. 3(b), above $T_c$, the $\text{Fe}^{3+}$ spins are ordered antiferromagnetically along the $c$ axis as $\tilde{S}_z = -\tilde{S}_z^A = \tilde{S}_z^B$, while they are slightly canted toward the $a$ axis as $\tilde{S}_x = \tilde{S}_x^A / \tilde{S}_x^B$, and $\tilde{S}_y = \tilde{S}_y^A / \tilde{S}_y^B = 0$. Below $T_c$, the $\text{Fe}^{3+}$ spins rotate in the $bc$ plane, and the rotation angle is $\varphi = \arctan(\tilde{S}_y / \tilde{S}_z) = 46^\circ$ at $T = 0 \text{ K}$ with our parameters.

The $bc$ plane [46] are well reproduced in our spin model. The rotation angle of the $\text{Fe}^{3+}$ AFM vector is $\varphi = 46^\circ$ at $T = 0 \text{ K}$ with our parameters. This is approximately equal to the experimentally estimated value $\varphi = 49^\circ$ [46].

VI. EXTENDED DICKE HAMILTONIAN

In the previous sections, we discussed the LTPT of $\text{ErFeO}_3$ through mean-field calculations based on our spin model. It is a standard approach for analyzing magnetic phase transitions. In this section, in order to discuss the analogy between the LTPT and the SRPT in the Dicke model, we transform the spin model, Eq. (7), into an extended version of the Dicke model, including direct $\text{Er}^{3+}-\text{Fe}^{3+}$ exchange interactions, which were not considered in our previous studies [33, 54].

We first rewrite the $\text{Fe}^{3+}$ subsystem in terms of the annihilation and creation operators of a magnon in Sec. VI.A. The $\text{Er}^{3+}$ subsystem is rewritten by large spin operators in Sec. VI.B. The $\text{Er}^{3+}-\text{Fe}^{3+}$ exchange interactions are transformed into five $\text{Er}^{3+}$-magnon couplings in Sec. VI.C. The total Hamiltonian is given in Sec. VI.D.

A. $\text{Fe}^{3+}$ subsystem

We assume that the most-stable values of the $\text{Fe}^{3+}$ spins at zero temperature, $\tilde{S}^{A/B}$, are unchanged when an external DC magnetic flux density $\mathbf{B}^{\text{DC}}$ ($\lesssim 10 \text{ T}$) is applied, as we also assumed in our previous studies [33, 54]. Under this assumption, as depicted in Fig. 1(a), the most stable state (i.e., ground state) of the $\text{Fe}^{3+}$ subsystem $\mathcal{H}_{\text{Fe}}$, Eq. (8), are expressed as

$$\tilde{S}_0^A = \begin{pmatrix} S \sin \beta_0 \\ 0 \\ -S \cos \beta_0 \end{pmatrix}, \quad \tilde{S}_0^B = \begin{pmatrix} S \sin \beta_0 \\ 0 \\ S \cos \beta_0 \end{pmatrix}. \quad (18)$$

Here, the canting angle $\beta_0$ is expressed as (see Appendix E or Refs. 33, 53, and 54)

$$\beta_0 = -\frac{1}{2} \arctan \frac{A_{xy} + z_{\text{Fe}} D_{\beta}^y}{z_{\text{Fe}} J_{\text{Fe}}^0 - A_0 + A_z}. \quad (19)$$

The magnon is the quantum of spin fluctuations from this stable state. As shown in Appendix E as well as in Refs. 33 and 54 in the long wavelength limit, the $\text{Fe}^{3+}$ Hamiltonian $\mathcal{H}_{\text{Fe}}$, Eq. (8), can be rewritten in terms of the annihilation (creation) operators $\hat{a}_K (\hat{a}_K^\dagger)$ of $\text{Fe}^{3+}$ magnons as

$$\hat{\mathcal{H}}_{\text{Fe}} \approx \sum_{K=0,\pi} \hbar \omega_K \hat{a}_K^\dagger \hat{a}_K + \text{const.} \quad (20)$$

Here, $K = 0$ and $\pi$ correspond to the quasi-ferromagnetic (qFM) and quasi-antiferromagnetic (qAFM) magnon modes [53], respectively. Their eigenfrequencies can be obtained as

$$\omega_K = \gamma \sqrt{(b \cos K - a) (d \cos K + c)} \quad (21)$$

where we defined

$$a = |S|/(g \mu_B) [-A_z - A_x - (z_{\text{Fe}} J_{\text{Fe}}^0 + A_z - A_x) \cos(2\beta_0) + (A_{xz} + z_{\text{Fe}} D_{\beta}^y) \sin(2\beta_0)], \quad (22a)$$

$$b = |S|/(g \mu_B) (z_{\text{Fe}} J_{\text{Fe}}^0), \quad (22b)$$

$$c = |S|/(g \mu_B) [(z_{\text{Fe}} J_{\text{Fe}}^0 + 2 A_z - 2 A_x) \cos(2\beta_0) + z_{\text{Fe}} D_{\beta}^y \sin(2\beta_0)], \quad (22c)$$

$$d = |S|/(g \mu_B) [-z_{\text{Fe}} J_{\text{Fe}} \cos(2\beta_0) - (2 A_{xx} + z_{\text{Fe}} D_{\beta}^y) \sin(2\beta_0)], \quad (22d)$$

The operators of the spin fluctuations $\delta \tilde{S}_i^{A/B} \equiv \tilde{S}_i^{A/B} - \tilde{S}_0^{A/B}$ are expressed as

$$\delta \tilde{S}_i^A = \sqrt{\frac{S}{2N_0}} \begin{pmatrix} (-\bar{T}_0 - \bar{T}_x) \cos \beta_0 \\ (\bar{Y}_0 - \bar{Y}_x) \\ (-\bar{T}_0 - \bar{T}_x) \sin \beta_0 \end{pmatrix}, \quad (23a)$$

$$\delta \tilde{S}_i^B = \sqrt{\frac{S}{2N_0}} \begin{pmatrix} (\bar{T}_0 + \bar{T}_x) \cos \beta_0 \\ (\bar{Y}_0 + \bar{Y}_x) \\ (-\bar{T}_0 + \bar{T}_x) \sin \beta_0 \end{pmatrix}, \quad (23b)$$
where we defined

\[
\hat{T}_K = \left( \frac{b \cos K - a}{d \cos K + c} \right)^{1/4} \left( \hat{a}_K^+ + \hat{a}_K \right),
\]
(24a)
\[
\hat{Y}_K = \left( \frac{d \cos K + c}{b \cos K - a} \right)^{1/4} \frac{i(\hat{a}_K^+ - \hat{a}_K)}{\sqrt{2}}.
\]
(24b)

For the discussion in the next subsections, we define the sum and difference of the spins as

\[
\hat{S}_i^\pm \equiv \hat{S}_i^A \pm \hat{S}_i^B.
\]
(25)

Their equilibrium (most stable) values are

\[
\hat{S}_0^+ = \hat{S}_0^A + \hat{S}_0^B = (2S \sin \beta_0, 0, 0)^t, \quad (26a)
\]
\[
\hat{S}_0^- = \hat{S}_0^A - \hat{S}_0^B = (0, 0, -2S \cos \beta_0)^t, \quad (26b)
\]

and their fluctuations are expressed as

\[
\delta \hat{S}^+ = \delta \hat{S}_1^A + \delta \hat{S}_1^B = \sqrt{\frac{2S}{N_0}} \left( \hat{T}_0 \cos \beta_0 \right),
\]
(27a)
\[
\delta \hat{S}^- = \delta \hat{S}_1^A - \delta \hat{S}_1^B = \sqrt{\frac{2S}{N_0}} \left( -\hat{T}_0 \cos \beta_0 \right).
\]
(27b)

C. Er\(^{3+}\)-Fe\(^{3+}\) interactions

In the same manner as in Refs. 43 and 54, we rewrite the Hamiltonian of the Er\(^{3+}\)-Fe\(^{3+}\) exchange interactions, Eq. (10), as

\[
\hat{H}_{Er-Fe} \approx 2J \left( \hat{\Sigma}^+ \cdot \hat{\Sigma}_0^+ + \hat{\Sigma}^+ \cdot \hat{\delta S}^+ \right)
+ \left( 0 \right) \cdot \left( \hat{\Sigma}^+ \cdot \hat{\Sigma}_0^- + \hat{\Sigma}^+ \cdot \hat{\delta S}^- \right)
+ \left( 0 \right) \cdot \left( \hat{\Sigma}^- \cdot \hat{\Sigma}_0^- + \hat{\Sigma}^- \cdot \hat{\delta S}^- \right).
\]
(32)

In each parenthesis, the first terms represent the influence of the static components (equilibrium values) \(\hat{S}_0^{A/B}\) of Fe\(^{3+}\) spins to Er\(^{3+}\) spins \(\hat{\Sigma}^\pm\), and the second terms represent the coupling between the Fe\(^{3+}\) fluctuation \(\Delta S^\pm\) and the Er\(^{3+}\) spins \(\hat{\Sigma}^\pm\). We divide these terms into the two Hamiltonians as

\[
\hat{H}^{\Sigma}_{Er-Fe} = \hat{H}^{\Sigma}_{Er-Fe} + \hat{H}^{\text{coupling}}_{Er-Fe}.
\]
(33)

The first term gives a part of the Er\(^{3+}\) spin resonance frequency, and it is expressed as

\[
\hat{H}^{\Sigma}_{Er-Fe} = E_x \hat{\Sigma}_x^z,
\]
(34)

where we used Eqs. (20) and \(E_x\) is defined as

\[
E_x = 4S(J \sin \beta_0 + D_y \cos \beta_0).
\]
(35)

Note that we neglected \((-4S D_x \cos \beta_0) \hat{\Sigma}^-\) under the assumption explained at the end of Sec. IV. The second term in Eq. (33) is rewritten in terms of the Fe\(^{3+}\) fluctuations as

\[
\hat{H}^{\text{coupling}}_{Er-Fe} = \sqrt{\frac{8S}{N_0}} \left[ (J \cos \beta_0 - D_y \sin \beta_0) \hat{T}_0 \hat{\Sigma}_x^+ \right.
+ J \hat{Y}_0 \hat{\Sigma}_y^z + (D_x \sin \beta_0) \hat{T}_x \hat{\Sigma}_y^z + D_z \hat{Y}_z \hat{\Sigma}_z^z
\]
\[
+ \left( -J \sin \beta_0 - D_y \cos \beta_0 \right) \hat{T}_0 \hat{\Sigma}_z^z \right].
\]
(36)

D. Total system

The total Hamiltonian derived from our spin model is finally expressed as

\[
\hat{H} \approx \sum_{K=0,\pi} \hbar \omega_K \hat{a}_K^+ \hat{a}_K + \hat{E}_x \hat{\Sigma}_x^x + \sum_{\xi = x,y,z} g_{Er}^\xi \mu_B D_{\xi} \hat{\Sigma}_{\xi}^x
+ \frac{8g_{Er}^x D_{\xi}}{N} \left( \hat{a}_x^+ \hat{a}_x^+ \right)
+ \frac{2h_{xy}}{\sqrt{N}} \hat{\delta \Sigma}_x^x
\]
\[
+ i \frac{2h_{xy}}{\sqrt{N}} \left( \hat{a}_x^+ - \hat{a}_x \right) \hat{\Sigma}_y^x
\]
\[
+ \frac{2h_{xy}}{\sqrt{N}} \left( \hat{a}_x^+ - \hat{a}_x \right) \hat{\Sigma}_y^x.
\]
(37)
Here, the five $\text{Er}^{3+}$-magnon coupling terms in Eq. (36) were rewritten in terms of the annihilation (creation) operators $\hat{a}_K$ ($\hat{a}_K^\dagger$) of a magnon. The five coupling strengths are defined as

$$
\begin{align*}
    h_{gx} &= \sqrt{2xS}(J \cos \beta_0 - D_y \sin \beta_0) \left(\frac{b + a}{d - c}\right)^{1/4} \\
    &= h \times \sqrt{x} \times 0.051 \text{ THz}, \\
    h_{gy} &= \sqrt{2xS}J \left(\frac{d + c}{b - a}\right)^{1/4} \\
    &= h \times \sqrt{x} \times 0.041 \text{ THz}, \\
    h_{g\eta} &= \sqrt{2xS}(D_x \sin \beta_0) \left(\frac{b + c}{d - c}\right)^{1/4} \\
    &= h \times \sqrt{x} \times 3.1 \times 10^{-5} \text{ THz}, \\
    h_{g\zeta} &= \sqrt{2xS}(D_z \cos \beta_0) \left(\frac{b - a}{d + c}\right)^{1/4} \\
    &= h \times \sqrt{x} \times (-0.040) \text{ THz}.
\end{align*}
$$

(38a)

The actual values are evaluated by the parameters shown in Appendix D. Note that, compared with the expression in our previous studies [33, 54], the above coupling strengths have additional factors: $\sqrt{2}$ and $\sqrt{S}$. The first of these factors, $\sqrt{2}$, originates from the number of $\text{Er}^{3+}$ sublattices in the present study, while a single $\text{Er}^{3+}$ lattice was considered in our previous studies [33, 54]. On the other hand, the second factor, $\sqrt{S}$, comes from the difference in the way of normalizing the $\text{Fe}^{3+}$ spins between the present and previous studies [33, 54].

VII. ANALOGY BETWEEN THE TWO PHASE TRANSITIONS

Based on the extended Dicke Hamiltonian, Eq. (37), derived in the previous section, we show in this section that the LTPT in $\text{ErFeO}_3$ is a magnonic SRPT.

In Sec. VII A we show that the $\text{Er}^{3+}$-qAFM magnon coupling with a strength of $g_x$ corresponds to the magnon coupling in the SRPT case. We also demonstrate that the thermal SRPT predicted by the extended Dicke Hamiltonian correctly reproduces the temperature-dependence of the $\text{Er}^{3+}$ and $\text{Fe}^{3+}$ spins shown in Fig. 4.

In Sec. VII B we quantitatively compare the contributions of the $\text{Er}^{3+}$-magnon coupling and the $\text{Er}^{3+}$-$\text{Er}^{3+}$ exchange interactions in the LTPT. We show that the LTPT can be caused solely by the $\text{Er}^{3+}$-magnon coupling. Furthermore, we demonstrate that the $\text{Er}^{3+}$-magnon coupling enhances the critical temperature and critical magnetic field of the phase transition, compared with the case in which the phase transition is driven by the $\text{Er}^{3+}$-$\text{Er}^{3+}$ exchange interactions alone.

A. Correspondence

In this section, by using the semiclassical method described in Sec. III with the extended Dicke Hamiltonian, Eq. (37), we calculate the thermal-equilibrium values of $\text{Er}^{3+}$ and $\text{Fe}^{3+}$ spins and magnon amplitudes as a function of temperature.

While the $\text{Er}^{3+}$ spin ensemble is described by six operators, $\hat{\Sigma}_{x,y,z}$ and $\hat{\Sigma}^{+}_{x,y,z}$ in the extended Dicke Hamiltonian, only $\hat{\Sigma}^{+}_{x}$ and $\hat{\Sigma}^{-}_{x}$ are relevant to the LTPT depicted in Fig. 1. $\hat{\Sigma}^{+}_{x}$ corresponds to the paramagnetic alignment by the $\text{Fe}^{3+}$ magnetization along the $a$ axis, and $\hat{\Sigma}^{-}_{x}$ corresponds to the antiferromagnetic ordering along the $c$ axis. Then, for analyzing the thermal-equilibrium values of the spins, we need to consider only the following two terms in the $\text{Er}^{3+}$-$\text{Er}^{3+}$ exchange interactions:

$$
\begin{align*}
    \frac{S_{2\text{Er}}}{}N \hat{\Sigma}^A B &= \frac{S_{2\text{Er}}}{N} \hat{\Sigma}^A B = \frac{2 \epsilon_{x,y,z}}{N} \sum_{i=x,y,z} \left[ (\hat{\Sigma}^{+}_{i})^2 - (\hat{\Sigma}^{-}_{i})^2 \right] \\
    \rightarrow & \frac{2 \epsilon_{x,y,z}}{N} \left[ (\hat{\Sigma}^{+}_{x})^2 - (\hat{\Sigma}^{-}_{x})^2 \right].
\end{align*}
$$

(39)

On the other hand, while $\text{Fe}^{3+}$ spins are described by the qFM and qAFM magnon modes in the extended Dicke Hamiltonian, only the qAFM mode is relevant to the LTPT. As shown in Fig. 1, $\delta S^{-}_{z}$ and $\delta S^{-}_{y}$ are required for describing the rotation of the $\text{Fe}^{3+}$-AFM vector in the $bc$ plane, and $\delta S^{-}_{x}$ is required for the possible modulation of canting along the $a$ axis. As seen in Eqs. (27), they are related to the qAFM magnon mode ($K = \pi$), and the qFM mode ($K = 0$) plays no role in the LTPT.

Consequently, among the terms in the total Hamiltonian given by Eq. (37), we only need to consider the following terms for describing the LTPT (the other terms are required for fully reproducing the THz spectra as discussed in Appendix C):

$$
\begin{align*}
    \hat{H}/h &\rightarrow \omega_x \hat{a}_x \hat{a}_x + \omega_x \hat{\Sigma}_x^z + \frac{2 \epsilon_{x,y,z}}{N h} \left( \hat{\Sigma}_x^z \right)^2 + \frac{2 g_{x}}{\sqrt{N}} (\hat{a}_x + \hat{a}_x) \hat{\Sigma}_x^z + \frac{i 2 g_{x}}{\sqrt{N}} (\hat{a}_x - \hat{a}_x) \hat{\Sigma}_x^z.
\end{align*}
$$

(40)

Here, the $\text{Er}^{3+}$ resonance frequency is defined as

$$
\omega_{\text{Er}} = \frac{|E_x + g_{x}^\text{Er} \mu_B B_{DC}|}{h}.
$$

(41)

Note that we re-wrote the large spin operators representing the $\text{Er}^{3+}$ spin ensemble as

$$
\begin{align*}
    \begin{cases}
        \hat{\Sigma}_x^z \rightarrow \hat{\Sigma}^z \\
        \hat{\Sigma}_y^z ightarrow \hat{\Sigma}_y^z \\
        \hat{\Sigma}_z^z ightarrow \hat{\Sigma}_z^z \\
    \end{cases}
\end{align*}
$$

(42)

where we re-indexed the Pauli operators representing the $\text{Er}^{3+}$ spins in the two sublattices as

$$
\begin{align*}
\hat{\sigma}_i^{A,x} &\rightarrow \hat{\sigma}_{21-1,x} \\
\hat{\sigma}_i^{A,y} &\rightarrow \hat{\sigma}_{21-1,y} \\
\hat{\sigma}_i^{A,z} &\rightarrow \hat{\sigma}_{21-1,z}
\end{align*}
$$

(43)
In Eq. (40), we assumed that the external DC magnetic field is applied along the a axis for keeping the $\Gamma_{12}$ symmetry, where either $[\vec{\sigma}_1^a - \vec{\sigma}_2^a]$ or the rotation angle $\phi$ of the Re$^{3+}$--AFM vector from the c axis can be the order parameter for the LTPT. Among the five Re$^{3+}$--magnon couplings in Eq. (37), only the $g_x$ and $g_z$ terms are required for considering the coupling between $\Sigma_{x,z}$ and the qAFM magnons. While the $g_y$ term also couples $\Sigma_y$ and qAFM magnons, its coupling strength is negligible compared with $g_{x,z}$, as shown in Eqs. (38), consistent with the experimentally observed antiferromagnetic ordering of Re$^{3+}$ spins along the c axis ($\langle \hat{\Sigma}_{y} \rangle = 0$).

Through comparison of Eq. (40) with Eq. 1 (the Dicke model), we can identify the $g_z$ term to correspond to the matter–photon coupling (transverse coupling). Additionally, the $g_y$ term represents longitudinal coupling and the $J_{Ec}$ term describes the Re$^{3+}$--Er$^{3+}$ exchange interactions in Eq. (40). The coupling strength $g_z = 2\pi \times 0.116$ THz puts the systems in the ultrastrong regime, since it is a significant fraction of the Re$^{3+}$ resonance and qAFM magnon frequencies, $E_x = h \times 0.023$ THz and $\omega_\pi = 2\pi \times 0.896$ THz. When the $g_x$ term causes a SRPT, $\langle \hat{\Sigma}_{x} \rangle = \langle \hat{\Sigma}_{z} \rangle$ spontaneously acquires a nonzero value in thermal equilibrium, corresponding to the antiferromagnetic ordering of Re$^{3+}$ spins along the c axis. As will be discussed later, the spontaneous appearance of nonzero ($i(\vec{a}_{\perp}^i - \vec{a}_\parallel)$), which is coupled with $\Sigma_{z}$ in the $g_z$ term, corresponds to the rotation of the Fe$^{3+}$--AFM vector.

Following the semiclassical treatment in Sec. III, we calculate the expectation values of the Re$^{3+}$ and Fe$^{3+}$ qAFM magnons at a finite temperature. In the thermodynamic limit, $N \to \infty$, the partition function $Z(T) \equiv \text{Tr}[e^{-\hat{H}/(k_B T)}]$ can be approximately evaluated by replacing the trace over the magnonic variables with an integral over c-numbers, $\vec{a}_r, \vec{a}_i \in \mathbb{R}$, giving $\vec{a}_\parallel \to \sqrt{N}(\vec{a}_r + i\vec{a}_i)$ as

$$
\tilde{Z}(T) = \int \frac{d\vec{a}_r d\vec{a}_i}{\pi N} \text{Tr}[e^{-\hat{H}/(k_B T)}] = \int \frac{d\vec{a}_r d\vec{a}_i}{\pi N} e^{-\hat{S}(\vec{a},T)/(k_B T)},
$$

where we defined an effective Hamiltonian

$$\hat{H}^{\text{eff}}(\vec{a}_r, \vec{a}_i)/\hbar \equiv N \omega_\pi (\vec{a}_r^2 + \vec{a}_i^2) + \omega_{Er} \hat{\Sigma}_x + \frac{4\omega_{Er}/N\hbar}{N\hbar} \left(\langle \hat{\Sigma}_x \rangle \hat{\Sigma}_x - \langle \hat{\Sigma}_x \rangle \langle \hat{\Sigma}_x \rangle \right) - \frac{2\omega_{Er}/N\hbar}{N\hbar} \left(\langle \hat{\Sigma}_y \rangle^2 - \langle \hat{\Sigma}_z \rangle^2 \right) + 4g_x \vec{a}_r \hat{\Sigma}_x + 4g_z \vec{a}_i \hat{\Sigma}_z$$

by introducing the Re$^{3+}$ components $\langle \hat{\Sigma}_{x,z} \rangle$ of the mean-fields for the Re$^{3+}$ ensemble. The action appearing in Eq. (44b) is defined as

$$\hat{S}(\vec{a}_r, \vec{a}_i, T) \equiv -k_B T \text{ln} \text{Tr}[e^{-\hat{H}/(k_B T)}]$$

$$= N \left\{\hbar \omega_\pi (\vec{a}_r^2 + \vec{a}_i^2) - \frac{2\omega_{Er}/N\hbar}{N\hbar} \left(\langle \hat{\Sigma}_x \rangle^2 - \langle \hat{\Sigma}_x \rangle^2 \right) \right\} - Nk_B T \text{ln} \text{Tr}[e^{-\hat{H}/(k_B T)}],$$

where we defined an effective Hamiltonian per Re$^{3+}$ spin as

$$\frac{\hat{\mathcal{H}}^{\text{eff}}(\vec{a}_r, \vec{a}_i)}{\hbar} \equiv \frac{\omega_{Er}}{N\hbar} \hat{\Sigma}_x + \frac{2\omega_{Er}/N\hbar}{N\hbar} \left(\langle \hat{\Sigma}_x^+ \rangle \hat{\sigma}_x - \langle \hat{\Sigma}_x^- \rangle \hat{\sigma}_z \right) + 2g_x \vec{a}_r \hat{\sigma}_x + 2g_z \vec{a}_i \hat{\sigma}_z.$$

We omitted the site index $i$ here, since all the spins are identical. The action $\mathcal{S}$ is minimized at $\partial \mathcal{S}/\partial \vec{a}_r = 0$ and $\partial \mathcal{S}/\partial \vec{a}_i = 0$, by which we get

$$\omega_\pi \vec{a}_r + g_x \vec{a}_i = 0, \quad \omega_\pi \vec{a}_i + g_x \vec{a}_r = 0,$$

where the expectation values of the Pauli operators are defined, for given $\vec{a}_r$ and $\vec{a}_i$, as

$$\vec{\sigma}_x \equiv \langle \vec{\sigma}_x \rangle \equiv \frac{\text{Tr}[\sigma_x e^{-\hat{H}/(k_B T)}]}{\text{Tr}[e^{-\hat{H}/(k_B T)}]},$$

$$\vec{\sigma}_y \equiv \langle \vec{\sigma}_y \rangle \equiv \frac{\text{Tr}[\sigma_y e^{-\hat{H}/(k_B T)}]}{\text{Tr}[e^{-\hat{H}/(k_B T)}]}.$$
In Fig. 4 we plot the thermal-equilibrium values of (a) $\sigma_z$ spins, $S_z$, (b) $S^+_{x,y,z}$, and (c) $S_3^{+}$ magnon amplitudes, as a function of temperature $T$. They were calculated by the semiclassical method with the extended Dicke Hamiltonian in the case of zero external DC magnetic field. Figures 4(a) and (b) are almost the same as Figs. 3(a) and (b), respectively, except $S_z$, which is not largely changed due to bosonization. The $S_3^{+}$ spins, $S_{x,y,z}$, were calculated by Eqs. (52) with the thermal-equilibrium value of qAFM magnon annihilation operator $\langle \hat{a} \rangle = \sqrt{\langle \hat{a}^+ \hat{a} \rangle}$ plotted in Fig. 3(c).

As seen in Eqs. (38), the transverse coupling strength, $g_{z}$, depends on $D_{x}$, and the longitudinal coupling strength, $g_{x}$, depends on $J$ and $D_{y}$. As seen in Eq. (16), the $D_{x}$ antisymmetric Er$^{3+}$–Fe$^{3+}$ exchange interaction is essential for the LTPT, because it couples $\hat{\sigma}_{x}^{A/B}$ and $\hat{S}_{y}^{A/B}$, which appear spontaneously at $T < T_c$. In contrast, the $J$ and $D_{y}$ exchange interactions are not directly related to the LTPT.

In this way, we can quantitatively reproduce the LTPT as the SRPT in the extended Dicke Hamiltonian, Eq. (37), which was derived from the spin model of ErFeO$_3$. The essential terms are extracted in Eq. (10). The $g_{z}$ term (antisymmetric Er$^{3+}$–Fe$^{3+}$ exchange interaction with $D_{x}$) corresponds to the matter–photon coupling and causes the antiferromagnetic ordering of Er$^{3+}$ spins along the $c$ axis and the $b$ component of the Fe$^{3+}$ spins through the spontaneous appearance of qAFM magnons.

**B. Er$^{3+}$—magnon coupling contribution**

Although the $g_{z}$ term causes the spontaneous appearance of both $\hat{\sigma}_{z}$ and $\hat{S}_{y}$ following the picture of the SRPT, a nonzero $\hat{\sigma}_{z}$ can spontaneously appear also by the $J_{Er}$ term (Er$^{3+}$–Er$^{3+}$ exchange interactions). While the Er$^{3+}$–magnon coupling is inevitable for the spontaneous rotation of Fe$^{3+}$ AFM vector (spontaneous appearance of $\hat{S}_{y}$), we try to evaluate quantitatively the contributions of the Er$^{3+}$–magnon coupling and Er$^{3+}$–Er$^{3+}$ exchange interactions for the LTPT in this subsection.

In Fig. 5 we plot the phase boundaries calculated by the full Hamiltonian (solid lines), in the absence of Er$^{3+}$–Fe$^{3+}$ exchange interactions (dash-dotted line; $J = D_{x} = D_{y} = g_{x} = g_{z} = 0$), and in the absence of Er$^{3+}$–Er$^{3+}$ exchange interactions (dashed line; $J_{Er} = 0$). Figures 5(a) and (b) show results by the mean-field method and by the semiclassical method with the extended Dicke Hamiltonian, respectively. The solid curve in Fig. 5(a) is equal to that in Fig. 2(a). The small differences between Figs. 5(a) and (b) are discussed in Appendix A.

As shown by the dashed lines ($J_{Er} = 0$), the phase transition occurs even in the absence of Er$^{3+}$–Er$^{3+}$ exchange interactions, and the critical temperature $T_c \sim 1.2$ K at $B^{DC} = 0$. This means that the Er$^{3+}$–magnon coupling alone can cause the LTPT. In this sense, the LTPT can be interpreted as a magnonic SRPT, because the Er$^{3+}$–magnon coupling is strong enough for the phase transition to occur.

On the other hand, in the absence of Er$^{3+}$–magnon coupling, as shown by dash-dotted lines, the critical temperature $T_c \sim 2.6$ K at $B^{DC} = 0$. This result appears to indicate that the contribution of the Er$^{3+}$–Er$^{3+}$ exchange interactions is larger than that of the Er$^{3+}$–magnon coupling. However, the real critical temperature $T_c \sim 4$ K, meaning that the Er$^{3+}$–magnon coupling enhances the critical temperature of the phase transition. In the same manner, the critical magnetic field is also en-
Further, we replace all the operators by c-numbers the phase boundaries by the full Hamiltonian, and those in Eq. (40), to the L TPT more in detail, we derive the condition for SRPT in our extended Dicke Hamiltonian, Eq. (40), not occur solely by the photon–matter coupling and their interactions, i.e., the LTPT can be caused solely by the Er–Er exchange interactions. The dashed curves are those obtained in the absence of Er–Er exchange interactions, i.e., the LTPT can be caused solely by the Er–magnon coupling and thus can be interpreted as a magnonic SRPT.

FIG. 5. Phase boundaries of the LTPT in ErFeO₃ calculated by (a) the mean-field method and (c) the semiclassical method with the extended Dicke Hamiltonian. An external DC magnetic field is applied along the a axis. The solid curves are the phase boundaries by the full Hamiltonian, and those in Fig. 5(a) and (b) are equivalent. The dash-dotted curves are the phase boundaries in the absence of Er–Er–magnon coupling (Er³⁺–Er³⁺ exchange interactions). The dashed curves are those obtained in the absence of Er–Er–Er exchange interactions, i.e., the LTPT can be caused solely by the Er–magnon coupling and thus can be interpreted as a magnonic SRPT.

We rewrite \( \tilde{\Sigma} \) by the bosonic annihilation (creation) operator \( \hat{b} \) (\( \hat{b}^\dagger \)) as

\[
\tilde{\Sigma}_x \rightarrow \hat{b}^\dagger \hat{b} - \frac{N}{2},
\]

\[
\tilde{\Sigma}_y \rightarrow \frac{\hat{b}^\dagger (N - \hat{b}^\dagger \hat{b})^{1/2} + (N - \hat{b}^\dagger \hat{b})^{1/2} \hat{b}}{2},
\]

\[
\tilde{\Sigma}_z \rightarrow \frac{\hat{b}^\dagger (N - \hat{b}^\dagger \hat{b})^{1/2} - (N - \hat{b}^\dagger \hat{b})^{1/2} \hat{b}}{2}.
\]

Further, we replace all the operators by c-numbers \( \hat{\alpha}_r, \hat{\alpha}_i, \hat{b}, \hat{b}^\dagger \in \mathbb{R} \) as

\[
\hat{\alpha} \rightarrow \sqrt{N} (\hat{\alpha}_r + i \hat{\alpha}_i),
\]

\[
\hat{b} \rightarrow i \sqrt{N} \hat{b}.
\]

Then, the Hamiltonian in Eq. (10) is transformed to

\[
\frac{\dot{\mathcal{H}}}{\hbar} \rightarrow \omega_\pi (\hat{\alpha}_r^2 + \hat{\alpha}_i^2) + \omega_{Er} \hat{b}^2 + \frac{4z_{Er} J_{Er}}{\hbar} \hat{b}^2 (\hat{b}^2 - 1) + 2g_x \hat{\alpha}_r (2\hat{b}^2 - 1) - 4g_x \hat{\alpha}_i \hat{b} \sqrt{1 - \hat{b}^2} + \text{const.}.
\]

The ground state of the system should satisfy

\[
\begin{align*}
\frac{1}{2} \hbar N \frac{\partial \mathcal{H}}{\partial \hat{\alpha}_r} &= \omega_\pi \hat{\alpha}_r + g_x (2\hat{b}^2 - 1) = 0, \\
\frac{1}{2} \hbar N \frac{\partial \mathcal{H}}{\partial \hat{\alpha}_i} &= \omega_\pi \hat{\alpha}_i - 2g_x \hat{b} \sqrt{1 - \hat{b}^2} = 0, \\
\frac{1}{2} \hbar N \frac{\partial \mathcal{H}}{\partial \hat{b}} &= \omega_{Er} \hat{b} + \frac{4z_{Er} J_{Er}}{\hbar} \hat{b} (2\hat{b}^2 - 1) + 4g_x \hat{\alpha}_r \hat{b} - 2g_x \hat{\alpha}_i \frac{1 - 2\hat{b}^2}{\sqrt{1 - \hat{b}^2}} = 0.
\end{align*}
\]

Solving the first two equations, we can express the Fe³⁺ magnon amplitudes as

\[
\begin{align*}
\hat{\alpha}_r &= -\frac{g_x}{\omega_\pi} (2\hat{b}^2 - 1), \\
\hat{\alpha}_i &= \frac{2g_x}{\omega_\pi} \hat{b} \sqrt{1 - \hat{b}^2}.
\end{align*}
\]

Substituting these into Eq. (54), we get an equation for the Er³⁺ amplitude as

\[
\begin{align*}
[\omega_{Er} - \frac{4g_x^2 - 4g_x^2}{\omega_\pi} - \frac{4z_{Er} J_{Er}}{\hbar} + \frac{8g_x^2 - 8g_x^2}{\omega_\pi} + \frac{8z_{Er} J_{Er}}{\hbar} \frac{\hbar \omega_{Er}}{\hbar} \hat{b}^2] \hat{b} = 0.
\end{align*}
\]

For a real nonzero value of \( \hat{b} \) to exist, the parameters must satisfy

\[
\frac{4g_x^2}{\omega_\pi \omega_{Er}} - \frac{4g_x^2}{\omega_\pi \omega_{Er}} + \frac{4z_{Er} J_{Er}}{\hbar \omega_{Er}} > 1.
\]

For \( J_{Er} = g_x = 0 \), this condition is reduced to \( 4g_x^2 > \omega_\pi \omega_{Er} \) for the SRPT in the Dicke model, Eq. (11).

The three terms on the left-hand side of Eq. (59) are evaluated as

\[
\begin{align*}
D_{g_x} &\equiv \frac{4g_x^2}{\omega_\pi \omega_{Er}} = 2.65, \\
D_{g_x} &\equiv -\frac{4g_x^2}{\omega_\pi \omega_{Er}} = -0.51, \\
D_{J_{Er}} &\equiv \frac{4z_{Er} J_{Er}}{\hbar \omega_{Er}} = 9.29.
\end{align*}
\]

In the following, we call them coupling depths. They are dimensionless measures of coupling strengths and are definitely determined based on the appearance of the SRPT. As seen in Eq. (59), the SRPT occurs when the sum of these coupling depths is greater than unity: \( D_{g_x} + D_{g_x} + D_{J_{Er}} > 1 \). The coupling depth \( D_{J_{Er}} \) of the \( J_{Er} \) term is the largest, which is consistent with Fig. 5. The \( g_x \) term (longitudinal coupling) gives a negative contribution for the SRPT (\( D_{g_x} < 0 \)). Among the three couplings, the contribution of the \( g_x \) term is \( D_{g_x} / (D_{g_x} + D_{g_x} + D_{J_{Er}}) = 0.23 \).
and the contribution of the total Er\(^{3+}\)-magnon coupling is \((D_{gs} + D_{gs})/(D_{gs} + D_{gs} + D_{Fe}) = 0.19\). These values are roughly equal to 1.3 K/(1.3 K + 3.4 K) = 0.28 estimated by Kadomtseva, Krynetskii, and Matveev [45], while they did not consider the longitudinal coupling \((g_x\) term), which is not included in the cooperative Jahn–Teller model [54, 50], and the parameters were determined only by the phase boundary for \(B^{DC}/a\).

From the viewpoint of the analogy between the two phase transitions, a remarkable fact is that the coupling depth of the \(g_z\) term satisfies \(D_{gs} > 1\) and \(D_{gs} + D_{gs} > 1\). This suggests that the transverse \(\text{Er}^{3+}\)-magnon coupling is much stronger than the longitudinal one (giving the negative contribution) and ultrastrong enough to cause the SRPT solely. Also in this sense, we can conclude that the LTPT in ErFeO\(_3\) is the magnonic SRPT obtained in the extended Dicke Hamiltonian with the direct atom–atom interaction and the longitudinal coupling \((g_x\) term).

VIII. SUMMARY

From a spin model of ErFeO\(_3\) that reproduces both the phase diagrams [34] and terahertz spectra [33], we derived an extended Dicke model that takes into account \(\text{Er}^{3+}\)-\(\text{Er}^{3+}\) exchange interactions as well as the cooperative coupling between \(\text{Er}^{3+}\) spins and \(\text{Fe}^{3+}\) magnon modes. We found that the LTPT in ErFeO\(_3\) can be caused solely by the \(\text{Er}^{3+}\)-magnon coupling (in the absence of \(\text{Er}^{3+}\)-\(\text{Er}^{3+}\) exchange interactions), which demonstrates that the LTPT is a magnonic SRPT in the extended Dicke model.

In the thermodynamic limit, \(N \to \infty\), the Dicke model is effectively interpreted as an infinite dimensional system [58], because the atoms interact equivalently with each other through the coupling with a single photonic mode. Such a dimensionality is reflected in critical exponents [58, 59] at phase transitions and would differentiate the LTPT in ErFeO\(_3\) from standard magnetic phase transitions caused by short-range (nearest neighbor, next-nearest-neighbor, …) exchange interactions between spins. Further, the coexistence of the direct (short-range) \(\text{Er}^{3+}\)-\(\text{Er}^{3+}\) interactions and \(\text{Er}^{3+}\)-magnon couplings (long-range retarded \(\text{Er}^{3+}\)-\(\text{Er}^{3+}\) interactions) in ErFeO\(_3\) can lead to rich physics beyond what the normal Dicke model provides.

The thermal SRPT in ErFeO\(_3\) would also give us rich physics compared with the quantum or zero-temperature SRPT that has been demonstrated by laser-driven cold atoms [7, 8]. In particular, it is known that the thermal and quantum fluctuations of photons and atoms show characteristic behaviors around the SRPT [50]. It is also known that the ground state of an ultrastrongly coupled system is a quantum squeezed vacuum even in the normal phase [7, 34, 59], and strong two-mode squeezing at the SRPT has been demonstrated numerically [66]. Our on-going terahertz magnetospectroscopy experiments of \(\text{Er}_x\text{Y}_{1-x}\text{FeO}_3\) around the LTPT [66] will experimentally examine such characteristic quantum squeezing at the thermal and quantum SRPTs.

ACKNOWLEDGMENTS

This research was supported by JST PRESTO program (grant JPMJP1767), National Science Foundation (Cooperative Agreement DMR-1720595), and U.S. Army Research Office (grant W911NF-17-1-0259). We thank Andrey Baydin, Kenji Hayashida, Chien-Lung Huang, Takuma Makihara, Atsushi Miyake, Atsuhiko Miyata, and Fuyang Tay for fruitful discussion.

Appendix A: Mean-field Calculation

Since we simply consider an homogeneous external DC magnetic flux density \(B^{DC}\) in this paper, the expectation values of \(\text{Er}^{3+}\) spins \(\sigma^{A/B} \equiv \langle \sigma_i^{A/B} \rangle\) and \(\text{Fe}^{3+}\) spins \(S^{A/B} \equiv \langle S_i^{A/B} \rangle\) are independent of the site index \(i\). The bracket represents theoretically the expectation values of operators at finite temperature in the Heisenberg picture. It also corresponds to the ensemble average of the spins in each sublattice. Their equations of motion are obtained from the Heisenberg equations derived by the Hamiltonian in Eq. (7) as \((s = A, B)\)

\[
\hbar(\partial/\partial t)\sigma^s = -\sigma^s \times \mu_B B^s_{Er} (\{\sigma^{A/B}\}, \{S^{A/B}\}), \quad (A1a)
\]

\[
\hbar(\partial/\partial t)S^s = -S^s \times \mu_B B^s_{Fe} (\{\sigma^{A/B}\}, \{S^{A/B}\}). \quad (A1b)
\]

Here, \(B^s_{Er}\) and \(B^s_{Fe}\) are the mean-fields for \(\text{Er}^{3+}\) and \(\text{Fe}^{3+}\) spins, respectively, and they are expressed as

\[
g_\mu_B B^s_{Er} (\{\sigma^{A/B}\}, \{S^{A/B}\}) = \mu_B g^s_{Er} \cdot B^{DC} + 2g_{Er} J_{Er} \sigma^B + \sum_{s=A,B} 2 \begin{pmatrix} JS^s_x - (D_{A,s}^x + S^s_x) & 0 \\ 0 & JS^s_z - (D_{A,s}^z + S^s_z) \end{pmatrix}, \quad (A2a)
\]

\[
g_\mu_B B^s_{Fe} (\{\sigma^{A/B}\}, \{S^{A/B}\}) = \mu_B g^s_{Er} \cdot B^{DC} + 2g_{Er} J_{Er} \sigma^A + \sum_{s=A,B} 2 \begin{pmatrix} JS^s_x - (D_{B,s}^x + S^s_x) & 0 \\ 0 & JS^s_z - (D_{B,s}^z + S^s_z) \end{pmatrix}, \quad (A2b)
\]
In Eqs. (A2a) and (A2b), the first, second, and third terms represent the Zeeman effect, Er$^{3+}$–Er$^{3+}$ exchange interaction, and Er$^{3+}$–Fe$^{3+}$ exchange interaction, respectively. In Eqs. (A2c) and (A2d), the first, second, and third terms represent the Zeeman effect, Er$^{3+}$–Fe$^{3+}$ exchange interaction, and Fe$^{3+}$–Fe$^{3+}$ exchange interaction, respectively. The dilution of Er$^{3+}$ spins is reflected through the factors $z_{Fe} = 6x$ and $z$, i.e., the number of neighboring Er$^{3+}$ is effectively decreased by factor $x$. Since $(1/2)\hat{\sigma}_{A/B}^±$ corresponds to the spin-$\frac{1}{2}$ operator, the factor 2 appears overall in Eqs. (A2a) and (A2b). As explained at the end of Sec. IV, the $\parallel$ component of the third term in Eqs. (A2a) and (A2b) is set to be zero by implicitly considering a high energy potential.

The free energy of the system is minimized when the thermal-equilibrium values (time-averages) of spins $\hat{\sigma}_{A/B}$ and $\hat{S}_{A/B}$ are parallel to their mean-fields $B_{Er}^\parallel = B_{Er}^\parallel(\{\hat{\sigma}_{A/B}\}, \{\hat{S}_{A/B}\})$ and $B_{Fe}^\parallel = B_{Fe}^\parallel(\{\hat{\sigma}_{A/B}\}, \{\hat{S}_{A/B}\})$ as

$$\hat{\sigma}^s = \langle \hat{\sigma}^s \rangle = \langle \hat{\sigma}^a \parallel \rangle u_{Er}^s, \quad \hat{S}^s = \langle \hat{S}^s \parallel \rangle u_{Fe}^s,$$

where we defined unit vectors of the mean-fields as

$$u_{Er}^s \equiv \frac{B_{Er}^s}{|B_{Er}^s|}, \quad u_{Fe}^s \equiv \frac{B_{Fe}^s}{|B_{Fe}^s|}.$$

The thermal-equilibrium values $\hat{\sigma}_{A/B}$ and $\hat{S}_{A/B}$ are determined as follows. For given mean-fields $B_{Er}^\parallel$ and $B_{Fe}^\parallel$, effective Hamiltonians of each Er$^{3+}$ and Fe$^{3+}$ can be defined, respectively, as

$$\hat{\mathcal{H}}_{Er}^\parallel = \frac{1}{2}g_{\mu_B}B_{Er}^\parallel \cdot \hat{\sigma}_{Er}^\parallel$$

$$\hat{\mathcal{H}}_{Fe}^\parallel = g_{\mu_B}B_{Fe}^\parallel \cdot \hat{S}_{Fe}^\parallel.$$

Then, the partition functions are expressed as

$$Z_{Er}^s \equiv \text{Tr} \left[ e^{-\hat{\mathcal{H}}_{Er}^s/(k_B T)} \right] = \sum_{m=\pm 1} e^{-m y_s},$$

$$Z_{Fe}^s \equiv \text{Tr} \left[ e^{-\hat{\mathcal{H}}_{Fe}^s/(k_B T)} \right] = \sum_{m=-S}^{S} e^{-m x_s}.$$

where we defined

$$y_s \equiv g_{\mu_B}|B_{Er}^s|/(k_B T), \quad x_s \equiv g_{\mu_B}|B_{Fe}^s|/(k_B T).$$

Since $\hat{\sigma}_{A/B}$ is not a standard spin operator with an angular momentum of $1/2$ but is a vector of the Pauli operators, the summation is performed for $m = \pm 1$. The free energies are given as $-k_B T \ln Z_{Er}^s$ and $-k_B T \ln Z_{Fe}^s$, and the thermal-equilibrium values of the spins are obtained as

$$\langle \hat{\sigma}^a \parallel \rangle = -\frac{\partial}{\partial y_s} \ln Z_{Er}^s = -\tanh(y_s),$$

$$\langle \hat{S}^a \parallel \rangle = -\frac{\partial}{\partial x_s} \ln Z_{Fe}^s = -SB_S(S x_s).$$

where $B_S(z)$ is the Brillouin function defined as

$$B_J(z) = \frac{2J+1}{2J} \coth \left( \frac{2J+1}{2J} z \right) - \frac{1}{2J} \coth \left( \frac{z}{2J} \right).$$

By consistently solving Eqs. (A2a), (A3a), and (A3b), we can determine $\hat{\sigma}_{A/B}$ and $\hat{S}_{A/B}$ at finite temperatures.

Appendix B: Reduction of number of parameters

In this appendix, we reduce the number of parameters in our spin model by considering the spin configuration in the $\Gamma_12$ phase of ErFeO$_3$ when the external DC magnetic field is zero or along the $a$ axis. In the ground
state \((T = 0)\), the equilibrium values of the spins satisfy Eqs. (A1) with \((\partial / \partial t) R^{A/B} = 0\) and \((\partial / \partial t) S^{A/B} = 0\).

Here, as depicted in Fig. 1 due to the \(\pi\)-rotational symmetry about the \(a\) axis, we represent the four spins \(R^{A/B}\) and \(S^{A/B}\) (twelve elements) by six values as

\[
\begin{align*}
R^A_x &= R^B_x \equiv R_x, \quad (B1a) \\
R^A_y &= -R^B_y \equiv R_y, \quad (B1b) \\
R^A_z &= -R^B_z \equiv R_z, \quad (B1c)
\end{align*}
\]

Using these and Eqs. (A1) and (A2), we get

\[
\begin{align*}
(\begin{array}{c}
R_x \\
R_y \\
R_z
\end{array}) &\times \left[ \begin{array}{c}
ge^{x E B}_{z} B^{D C} \\
0 \\
0
\end{array} \right] + 2 z_{E E r} \left[ \begin{array}{c}
R_x \\
-R_y \\
-R_z
\end{array} \right] + 4 \left( \begin{array}{c}
J_{A,+} S_x + D_{A,-,y} S_z + D_{A,-,z} S_y \\
J_{A,-} S_y - D_{A,+} S_x - D_{A,-,x} S_z \\
-J_{A,+} S_z - D_{A,+} S_y + D_{A,+} S_x
\end{array} \right) = 0, \quad (B2a)
\end{align*}
\]

\[
\begin{align*}
(\begin{array}{c}
S_x \\
S_y \\
S_z
\end{array}) &\times \left[ \begin{array}{c}
ge^{x E F}_{z} B^{D C} \\
0 \\
0
\end{array} \right] + 2 z_{E E r} \left[ \begin{array}{c}
S_x \\
S_y \\
S_z
\end{array} \right] + 4 \left( \begin{array}{c}
J_{B,+} S_y + D_{B,-} S_z + D_{B,-} S_y \\
J_{B,-} S_y - D_{B,+} S_x - D_{B,-} S_z \\
-J_{B,+} S_z - D_{B,+} S_y + D_{B,+} S_x
\end{array} \right) = 0, \quad (B2b)
\end{align*}
\]

\[
\begin{align*}
&\frac{1}{2} \left[ (z_{E F} D_{x}^{F} - 2 A_{z}) S_x + (z_{E F} D_{y}^{F} - A_{x}) S_y + (z_{E F} D_{y}^{F} + A_{x}) S_z \right] + \left[ (z_{E F} D_{x}^{F} - A_{x}) S_x + (z_{E F} D_{y}^{F} + 2 A_{y}) S_y \right] + \left[ (z_{E F} D_{y}^{F} + A_{x}) S_x - (z_{E F} D_{x}^{F} + 2 A_{z}) S_z \right]
&= 0, \quad (B2c)
\end{align*}
\]

\[
\begin{align*}
&\frac{1}{2} \left[ (z_{E F} D_{x}^{F} - 2 A_{x}) S_x + (z_{E F} D_{y}^{F} - A_{y}) S_y + (z_{E F} D_{y}^{F} + A_{x}) S_z \right] + \left[ (z_{E F} D_{x}^{F} - A_{y}) S_x + (z_{E F} D_{y}^{F} + 2 A_{y}) S_y \right] + \left[ (z_{E F} D_{y}^{F} + A_{x}) S_x - (z_{E F} D_{x}^{F} + 2 A_{z}) S_z \right]
&= 0. \quad (B2d)
\end{align*}
\]

where we defined

\[
\begin{align*}
J_{s,\pm} &\equiv (J_{s,A} \pm J_{s,B})/2, \quad (B3a) \\
J_{s,\pm} &\equiv (J_{s,A} \pm J_{s,B})/2, \quad (B3b) \\
D_{s,\pm} &\equiv (D_{s,A} \pm D_{s,B})/2, \quad (B3c) \\
D_{s,\pm} &\equiv (D_{s,A} \pm D_{s,B})/2. \quad (B3d)
\end{align*}
\]

For the equivalence between Eq. (B2c) and Eq. (B2d), the following equations should be satisfied for any \(R_{x,y,z};\)

\[
\begin{align*}
J_{+} R_x + D_{-,y} R_z + D_{-,z} R_y &= J_{+} B R_x + D_{-,y} R_z + D_{-,z} R_y, \quad (B4a) \\
J_{-} R_y + D_{+} R_z - D_{-,x} R_x &= -J_{-} R_y + D_{+} R_z - D_{-,x} R_x, \quad (B4b) \\
J_{-} R_z + D_{-,x} R_y - D_{+,y} R_x &= -J_{-} R_z + D_{-,x} R_y + D_{+,y} R_x. \quad (B4c)
\end{align*}
\]

Then, we get the following relations:

\[
\begin{align*}
J_{+} &= J_{+}, \quad (B5a) \\
J_{-} &= -J_{-}, \quad (B5b) \\
D_{-,x} &= -D_{-,x}, \quad (B5c) \\
D_{+,y} &= -D_{+,y}, \quad (B5d) \\
D_{-,y} &= D_{-,y}, \quad (B5e) \\
D_{+,z} &= -D_{+,z}, \quad (B5f) \\
D_{-,z} &= D_{-,z}. \quad (B5g)
\end{align*}
\]
On the other hand, for the equivalence between Eq. (B2a) and Eq. (B2b) incorporating the consistency with Eqs. (B5), the following equations should be satisfied for any $S_{x,y,z}$:

\[
\begin{align*}
J_{A,+} & = J_{B,+}, \quad (B7a) \\
J_{A,-} & = -J_{B,-}, \quad (B7b) \\
D_{A,-x} & = -D_{B,-x}, \quad (B7c) \\
D_{A,+y} & = -D_{B,+y}, \quad (B7d) \\
D_{A,-y} & = D_{B,-y}, \quad (B7e) \\
D_{A,+z} & = -D_{B,+z}, \quad (B7f) \\
D_{A,-z} & = D_{B,-z}. \quad (B7g)
\end{align*}
\]

Then, we get the following relations:

\[
\begin{align*}
J_{A,A} & = J_{B,B} = J + J', \quad (B8a) \\
J_{A,B} & = J_{B,A} = J - J'. \quad (B8b)
\end{align*}
\]

From eigenvalues $E_k$ of the $12 \times 12$ coefficient matrix for $\delta \sigma^{A/B}$ and $\delta S^{A/B}$ on the right-hand sides, we can find four positive eigenfrequencies of the spin resonances as $\nu_k = i E_k/\hbar$. Another four are negative, and the other four are zero. The temperature used for determining the equilibrium spins $\bar{\sigma}^{A/B}$ and $\bar{S}^{A/B}$ will be assumed as $T = 20 K > T_c$. While it is higher than the cryostat temperature 10 K used for measuring the THz spectrum (shown in Fig. 3), $T = 20 K$ is better suited for reproducing the experimental spectrum. The reason remains as a future problem.

We will also calculate the spin resonance frequencies from the extended Dicke Hamiltonian, Eq. (C7). We will see that the five $E_k^{3+}$ magnon couplings show a variety of frequency anti-crossings. It originates from the fact that the Fe$^{3+}$ qFM ($K = 0$) and qAFM ($K = \pi$) magnon modes and the $E_k^{3+}$ spin resonances in the A and B sublattices are all coupled in general as seen in the extended Dicke Hamiltonian.

Note that the actual Hamiltonian treated in this Appendix is

\[
\hat{H} \approx \sum_{K=0,\pi} \hbar \omega_K \hat{a}^\dagger \hat{a} + E_x \hat{\Sigma}^+_x + \sum_{\xi=x,y,z} g^E_{\xi} \mu_B D^{DC}_{\xi} \hat{\Sigma}^+_{\xi} + \frac{8 \varepsilon_{E_x} J_{E_x}}{N} \hat{\Sigma}^+_x \cdot \hat{\Sigma}^+_y + \frac{2 \hbar g_{z}}{\sqrt{N}} (\hat{a}^\dagger - \hat{a}) \delta \hat{\Sigma}^+_z \\
+ \frac{i 2 \hbar g_{y}}{\sqrt{N}} (\hat{a}^\dagger - \hat{a}) \delta \hat{\Sigma}^+_y + \frac{i 2 \hbar g_{x}}{\sqrt{N}} (\hat{a}^\dagger - \hat{a}) \delta \hat{\Sigma}^+_{\xi}.
\]

Compared with Eq. (C7), the $E_k^{3+}$ spin operators $\hat{\Sigma}^\pm_{x,y,z}$ in the coupling terms are replaced by their fluctuations $\delta \hat{\Sigma}^\pm_{x,y,z} = \hat{\Sigma}^\pm_{x,y,z} - \bar{\Sigma}^\pm_{x,y,z}$. The terms including the equi-
librium values $\Sigma_{x,y,z}^\pm$ give shifts of Er$^{3+}$ magnon frequencies. However, returning to Eq. (29), we can find that the influence of these terms is smaller by factor $N_0^{-1/2}$ than the magnon Hamiltonian $\sum_{K=0,x} h\omega_K \hat{a}_K^\dagger \hat{a}_K$. Then, the equilibrium values $\Sigma_{x,y,z}^\pm$ can be omitted in Eq. (C2).

We will calculate the eigenfrequencies of Eq. (C2). However, since we suppose the $\Gamma_2$ phase ($T > T_c$) in this Appendix, we do not consider the spontaneous ordering of Er$^{3+}$ spins nor the rotation of the Fe$^{3+}$ spins in the calculation of the eigenfrequencies. Then, the results are justified only for relatively high external DC field that makes the system in the $\Gamma_2$ phase even in the zero-temperature limit.

In the calculation based on the extended Dicke Hamiltonian, the finite temperature ($T = 20$ K) is incorporated in the following procedure. We consider the thermal excitation of the Er$^{3+}$ spins and assume that the Er$^{3+}$ density effectively depends on the temperature as

$$x = \tanh \left( \frac{E_{Er}}{2k_BT} \right), \quad (3C)$$

where the Er$^{3+}$ excitation energy $E_{Er}$ (excluding the Er$^{3+}$–Er$^{3+}$ exchange interaction) is represented as

$$E_{Er} = \sqrt{(E_x + g_{Er}^x \mu_B B_{DC}^x)^2 + \sum_{\xi = y,z} (g_{Er}^\xi \mu_B B_{DC}^\xi)^2}. \quad (C4)$$

The temperature dependence appears through this effective $x$ and $z_{Er} = 6x$.

Note that, in this Appendix, the results by the mean-field approach is more reliable than those by the extended Dicke Hamiltonian, which are derived under some approximations. However, the spin resonance frequencies and anti-crossing on them will be better clarified by the extended Dicke Hamiltonian.

In the following subsections, we discuss how the five Er$^{3+}$–magnon couplings are reflected in three configurations: $B_{DC}^x//a$ (Appendix C1), $B_{DC}^x//b$ (Appendix C2), and $B_{DC}^x//c$ (Appendix C3). We compare them with our experimental results [33] in Appendix C4

1. $B_{DC}^x//a$

If the external DC magnetic field is along the $a$ axis, the Er$^{3+}$ subsystem is most stable when the Er$^{3+}$ spins are along the $a$ axis. For calculating the spin resonance frequencies from the extended Dicke Hamiltonian in the weak excitation limit (linear optical response), we here bosonize the spin operators. By the lowest-order Holstein–Primakoff transformation, the spin-$\frac{1}{2}$ operators are transformed as $(s=A,B)$

$$\hat{S}^s_x \rightarrow \hat{b}^+_s \hat{b}_s - \frac{N}{4}, \quad (C5a)$$
$$\delta \hat{S}^s_x \rightarrow \delta \hat{b}^+_s \hat{b}_s, \quad (C5b)$$
$$\hat{S}^s_y = \delta \hat{S}^s_y \rightarrow \sqrt{\frac{N}{2}} \delta \hat{b}^+_s + \hat{b}_s, \quad (C5c)$$
$$\hat{S}^s_z = \delta \hat{S}^s_z \rightarrow \sqrt{\frac{N}{2}} \delta \hat{b}^+_s - \hat{b}_s. \quad (C5d)$$

Then, the total Hamiltonian in Eq. (C2) is transformed as

$$\hat{H} \approx \sum_{K=0,x} h\omega_K \hat{a}_K^\dagger \hat{a}_K + (E_x + g_{Er}^x \mu_B B_{DC}^x)(\hat{b}^+_s \hat{b}_s + \hat{b}^+_b \hat{b}_b)$$
$$- 4z_{Er} J_{Er} \hat{b}^+_b \hat{b}_b + h g_x (\hat{a}^+_0 - \hat{a}_0)(\hat{b}^+_s \hat{b}_s + \hat{b}^+_b \hat{b}_b)$$
$$+ i h g_y (\hat{a}^+_0 - \hat{a}_0)(\hat{b}^+_b \hat{b}_b) + h g_z (\hat{a}^+_0 - \hat{a}_0)(\hat{b}^+_b \hat{b}_b - \hat{b}^+_s \hat{b}_s - \hat{b}^+_b \hat{b}_b)$$
$$+ \text{const.} \quad (C6)$$

Here, we defined operators of the in-phase oscillation $\hat{b}_+ \equiv \hat{b}_A + \hat{b}_B / \sqrt{2}$. In the weak excitation limit, the $g_x$ term can be neglected, since it is involved with the number of Er$^{3+}$ excitations $\hat{b}^+_A \hat{b}_A$. Then, the Hamiltonian can be divided into two parts as

$$\hat{H} \approx \hat{H}_{A+} + \hat{H}_{A-} + \text{const.} \quad (C7)$$

The first term consists of the Fe$^{3+}$ qFM magnon mode and Er$^{3+}$ in-phase mode, and it is expressed as

$$\hat{H}_{A+} = h \omega_0 \hat{a}^+_0 \hat{a}_0 + (E_x + g_{Er}^x \mu_B B_{DC}^x)(\hat{b}^+_s \hat{b}_s + \hat{b}^+_b \hat{b}_b) + i h g_y (\hat{a}^+_0 - \hat{a}_0)(\hat{b}^+_b \hat{b}_b)$$
$$- i h g_z (\hat{a}^+_0 - \hat{a}_0) \times \begin{cases} \left( \frac{\hat{b}^+_b - \hat{b}_b}{\hat{b}_b - \hat{b}_b} \right), & g_{Er}^x \mu_B B_{DC}^x > -E_x \\ \left( \frac{\hat{b}^+_s - \hat{b}_s}{\hat{b}_s - \hat{b}_s} \right), & g_{Er}^x \mu_B B_{DC}^x < -E_x \end{cases} \quad (C9)$$

If the coefficient $(E_x + g_{Er}^x \mu_B B_{DC}^x)$ of the second term in Eq. (C9) is negative for negative $B_{DC}^x$, the roles of the annihilation operator $\hat{b}_+$ and creation one $\hat{b}^+_s$ are flipped. As a result of it, the sign of the last term in Eq. (C9) was flipped. On the other hand, the second term in Eq. (C8) consists of the Fe$^{3+}$ qAFM magnon mode and Er$^{3+}$
out-of-phase mode, and it is expressed as

\[ \hat{H}_{\pi-} = \hbar \omega_0 \hat{a}_x \hat{a}_x + \hbar g_0' (\hat{a}_0^\dagger + \hat{a}_0) (\hat{b}_-^\dagger + \hat{b}_-) \]

\[ + \left( (E_x + g_x^E \mu_B B_{DC} - 4\varepsilon_{Er}J_{Er}) \hat{b}_-^\dagger \hat{b}_- + \hbar g_z (\hat{a}_0^\dagger - \hat{a}_0)(\hat{b}_-^\dagger - \hat{b}_-) \right) \]

\[ \delta \hat{\Sigma}_y \rightarrow \Delta \hat{\Sigma}_y \rightarrow B_0 \hat{b}_x - N/4, \]

\[ \delta \hat{\Sigma}_y \rightarrow B_0 \hat{b}_x, \]

\[ \hat{\Sigma}_z = \Delta \hat{\Sigma}_z \rightarrow \sqrt{\frac{N}{2}} \frac{\hat{b}_-^\dagger + \hat{b}_-}{\hat{b}_-^\dagger - \hat{b}_-}. \]

Then, in the weak excitation limit, the total Hamiltonian in Eq. (C2) is transformed to

\[ \hat{H} \approx \sum_{K=0,\pi} \hbar \omega_K \hat{a}_K \hat{a}_K + |g_y^E \mu_B B_{DC}| (\hat{b}_+^\dagger \hat{b}_+ - \hat{b}_-^\dagger \hat{b}_-) \]

\[ - 4\varepsilon_{Er}J_{Er} \hat{b}_-^\dagger \hat{b}_- - i\hbar g_z (\hat{a}_0^\dagger + \hat{a}_0)(\hat{b}_-^\dagger - \hat{b}_-) \]

\[ + i\hbar g_z (\hat{a}_0^\dagger - \hat{a}_0)(\hat{b}_-^\dagger + \hat{b}_-) + \hbar g_z (\hat{a}_0^\dagger + \hat{a}_0)(\hat{b}_+^\dagger + \hat{b}_+) + \text{const.} \]

This Hamiltonian can be used for \(|g_y^E \mu_B B_{DC}^E| > 4\varepsilon_{Er}J_{Er}\) similarly as the previous subsection. In this configuration, the two \(Fe^{3+}\) magnon modes and two \(Er^{3+}\) modes are all coupled in general. However, when we focus around the \(Fe^{3+}\) qFM magnon frequency, the Hamiltonian can be simplified as

\[ \hat{H} \approx \hbar \omega_0 \hat{a}_0^\dagger \hat{a}_0 + |g_y^E \mu_B B_{DC}| (\hat{b}_+^\dagger \hat{b}_+ + \hat{b}_-^\dagger \hat{b}_-) \]

\[ + \hbar g_z (\hat{a}_0^\dagger + \hat{a}_0)(\hat{b}_+^\dagger + \hat{b}_+) + \text{const.} \]

In this way, the \(Fe^{3+}\) qFM mode shows anti-crossing with the \(Er^{3+}\) in-phase mode. On the other hand, when we focus on the \(Fe^{3+}\) qFM magnon frequency, the Hamiltonian is simplified as

\[ \hat{H}_{\pi-} \approx \hbar \omega_0 \hat{a}_x \hat{a}_x + |g_y^E \mu_B B_{DC}^E| - 4\varepsilon_{Er}J_{Er} \hat{b}_-^\dagger \hat{b}_- \]

\[ + |g_y^E \mu_B B_{DC}^E| (\hat{b}_+^\dagger \hat{b}_+ + \hat{b}_-^\dagger \hat{b}_-) \]

\[ + i\hbar g_z (\hat{a}_0^\dagger - \hat{a}_0)(\hat{b}_+^\dagger + \hat{b}_-) + \text{const.} \]

In this way, the \(Fe^{3+}\) qFM mode shows anti-crossing with both \(Er^{3+}\) in-phase and out-of-phase modes.

In Figs. (a) and (b), we plot the spin resonance frequencies calculated by the mean-field approach, Eqs. (C1), and by Eq. (C12), respectively. The \(Er^{3+}\) in-phase and \(Fe^{3+}\) qFM modes show frequency anti-crossing around \(B_y = 12\) T. The \(Fe^{3+}\) qFM mode shows anti-crossing with the two \(Er^{3+}\) modes around \(B_y = 20\) T. The two approaches show almost the same resonance frequencies in the present case.
Finally, when the external DC magnetic field along the $c$ axis is large enough ($|g_{z}^{\text{Er}}\mu_{B}B_{z}^{\text{DC}}| \gg E_{a}$), the $E_{a}\Sigma_{z}^{+}$ term in Eq. (C2) can be neglected. In the same manner as the previous subsections, we transform the $\text{Er}^{3+}$ spins as

$$
\hat{\Sigma}_{z}^{+} = \hat{b}_{+}^{\dagger}\hat{b}_{-} - \frac{N}{4},
$$
(C14a)

$$
\delta\hat{\Sigma}_{z}^{+} = \hat{b}_{+}^{\dagger}\hat{b}_{-},
$$
(C14b)

$$
\hat{\Sigma}_{z}^{-} = \delta\hat{\Sigma}_{z}^{+} = \sqrt{\frac{N}{2}}(\hat{b}_{+}^{\dagger} + \hat{b}_{-}),
$$
(C14c)

$$
\hat{\Sigma}_{y}^{+} = \delta\hat{\Sigma}_{y}^{+} = \frac{\sqrt{N}}{2}(\hat{b}_{+}^{\dagger} - \hat{b}_{-}).
$$
(C14d)

In the weak excitation limit, the total Hamiltonian in Eq. (C2) is transformed to

$$
\hat{H} \approx \sum_{K=0,\pi} h\omega_{K}\hat{a}_{K}^{\dagger}\hat{a}_{K} + |g_{z}^{\text{Er}}\mu_{B}B_{z}^{\text{DC}}|(\hat{b}_{+}^{\dagger}\hat{b}_{-} + \hat{b}_{+}^{\dagger}\hat{b}_{-})
$$

$$
- 4z_{\text{Er}}J_{\text{Er}}\hat{b}_{-}^{\dagger}\hat{b}_{-}
$$

$$
+ h_{g_{x}}(\hat{a}_{x}^{\dagger} + \hat{a}_{x})(\hat{b}_{+}^{\dagger} + \hat{b}_{+}) + h_{g_{y}}(\hat{a}_{y}^{\dagger} - \hat{a}_{y})(\hat{b}_{+}^{\dagger} - \hat{b}_{+})
$$

$$
- i\hbar g_{y}^{\prime}(\hat{a}_{x}^{\dagger} + \hat{a}_{x})(\hat{b}_{+}^{\dagger} - \hat{b}_{-}) + \text{const}. \quad (C15)
$$
The first term consists of the two $^{3+}$Fe magnon modes and the $^{3+}$Er in-phase mode as
\[
\hat{H}_{0\pi\pi} = \sum_{K=0,\pi} \hbar \omega_K \hat{a}_K^\dagger \hat{a}_K + |\tilde{g}_{\text{Er}}^\text{EF} \mu_B B_{DC}^z| \hat{b}_z^\dagger \hat{b}_z \\
+ \hbar g_y (\hat{a}_b^\dagger - \hat{a}_b) (\hat{b}_+ - \hat{b}_-) \\
+ \hbar g_x (\hat{a}_r^\dagger + \hat{a}_r) (\hat{b}_+ + \hat{b}_-). \tag{C17}
\]
In this way, the $^{3+}$Er in-phase mode shows anti-crossing with both the two $^{3+}$Fe magnon modes. The second term in Eq. (C16) represents only the $^{3+}$Er out-of-phase mode as
\[
\hat{H}_- = (|\tilde{g}_{\text{Er}}^\text{EF} \mu_B B_{DC}^z| - 4z_{\text{Er}} J_{\text{Er}}) \hat{b}_z^\dagger \hat{b}_-. \tag{C18}
\]
This mode is coupled only with the qAFM mode by the strength of $g_y' \ll g_x, g_y$ under the approximation used for deriving the extended Dicke Hamiltonian.

In Figs. 8(a) and (b), we plot the spin resonance frequencies calculated by the mean-field approach, Eqs. (C1), and by the extended Dicke Hamiltonian, Eqs. (C17) (solid lines) and (C18) (dashed lines), respectively. As shown by solid lines in Fig. 8(b), obeying Eq. (C17), the $^{3+}$Er in-phase mode shows frequency anti-crossing with $^{3+}$Fe qFM mode around $B_z = 4$ T and with qAFM mode around $B_z = 7$ T. The frequency shifts of the $^{3+}$Fe magnon modes at large $B_z$ are not reproduced in Fig. 8(b) due to the approximations explained at the end of Appendix C 1.

As shown in Fig. 8(a), the $^{3+}$Er out-of-phase mode shows frequency anti-crossing with the $^{3+}$Fe qFM mode around $B_z = 4.5$ T and with the qAFM mode around $B_z = 8.5$ T. They are not obtained by the present calculation with the extended Dicke Hamiltonian as shown in Fig. 8(b). Such an inconsistency does not appear in the previous cases ($B_{DC}^y//a, b$). We checked that the inconsistency cannot be resolved even by considering the equilibrium contribution $\hat{X}_{x,y,z}^a$ in the $^{3+}$Er –magnon couplings in Eq. (C2). The $g_y'$ term also cannot resolve it, since it induces only the coupling between the $^{3+}$Er out-of-phase and $^{3+}$Fe qAFM modes.

This inconsistency originates from the fact that we did not properly consider the change of the equilibrium values of $^{3+}$Er and $^{3+}$Fe spins by the presence of the external DC field $B_{DC}$ in the derivation of the extended Dicke Hamiltonian. In fact, in the presence of $B_{DC}^y/c$, we can find by the mean-field method that the $^{3+}$Fe spins become strongly asymmetric about the $ab$ plane due to the large $z$ component of the macroscopic $^{3+}$Er spins induced by $B_{DC}^y/c$. Such an asymmetry causes the coupling between the $^{3+}$Er out-of-phase mode and the two $^{3+}$Fe magnon modes. Then, the anti-crossing appears in Fig. 8(a).

The reproduction of these anti-crossing by the extended Dicke Hamiltonian is beyond the scope of the present paper and it remains as a future task.

### 4. Comparison with experimental results

Since the maximum external DC magnetic flux density was limited by around 10 T in our previous study 32, the $^{3+}$Er –magnon anti-crossing was experimentally observed mainly for $B_{DC}^y//c$. The anti-crossing around $B_{DC}^z = 4$ T (7 T) was clearly (slightly) observed. If we apply the external DC field in the anti-parallel direction to the magnetization along the $a$ axis, we could observe anti-crossing around $B_{DC}^z = -7$ T as shown in Fig. 8. If we can apply a stronger DC magnetic field and the linewidth is narrow enough, we could observe the anti-crossing around $B_{DC}^y = 20$ T for $B_{DC}^y//b$ as shown in Fig. 7. In our previous study 32, the anti-crossing was slightly observed around $B_{DC}^y = 7$ T. It corresponds to the one around $B_{DC}^y = 12$ T in Fig. 7. The difference between the theoretical and experimental external DC fields is due to the red-shift of $^{3+}$Fe qFM mode caused by the DC-field-induced structural change, which is not considered in the present calculation. For $B_{DC}^y//a$, in order to observe the large anti-crossing around $B_{DC}^y = 13$ T and...
They were determined for fitting the spin resonance frequencies in Figs. [6]7 and 8 to their absorption peak positions observed in our experiments [32]. They are basically multiplied by factor 2 from the values estimated in our previous study [32] due to the additional factor 1/2 in Eq. (13).

The anisotropic g-factors for Fe$^{3+}$ spins were assumed to be

$$g^e_{Fe} = 2,$$  \hspace{1cm} (D3a)
$$g^o_{Fe} = 2,$$  \hspace{1cm} (D3b)
$$g^z_{Fe} = 0.6.$$

Here, $g^e_{Fe}$ was determined for reproducing the critical magnetic flux density $B_C^{DC}$ \(\sim 20 \ T\) \cite{34} of the transition between the $\Gamma_3$ phase and the $\Gamma_4$ one, where the Fe$^{3+}$ spins are ordered antiferromagnetically along the $a$ axis with a slight canting to the $c$ axis, in the case of $B^{DC}/c$. It occurs around $B_C^{DC} \sim 20 \ T$ \cite{34}. However, the temperature-induced $\Gamma_2-\Gamma_4$ spin-reorientation phase transition around $90 \ K \lesssim T \lesssim 100 \ K$ \cite{34, 43, 44} cannot be reproduced in the present model. Furthermore, there are at least four parameters, $J_{Er}$, $J$, $D_x$, and $D_y$, even if we reduce the number of parameters by the analysis in Appendix D. Further, the anisotropic $g$-factors $g^e_{Er}$, $g^o_{Er}$, and $g^z_{Er}$ of Er$^{3+}$ spins were also free parameters, and they can easily change the critical DC fields. The critical temperature and the three critical DC fields obtained by the magnetization measurements were not enough for determining the above parameters.

In order to determine all of them, the spin resonance frequencies are informative. Especially, as we discussed in Appendix C by the extended Dicke Hamiltonian, the Er$^{3+}$–Er$^{3+}$ exchange interaction strength $J_{Er}$ clearly appears as the frequency splitting between the Er$^{3+}$ in-phase and out-of-phase resonances. The out-of-phase mode cannot be excited by the THz wave unless it couples with the Fe$^{3+}$ magnon modes. In that sense, the

Appendix D: Parameters

Following our previous study \cite{32}, we used the following values for the Fe$^{3+}$ subsystem in our numerical calculations, except $A_x$, which was determined for fitting the spin resonance frequencies in Fig. 8 to the corresponding THz absorption spectrum in our experiments: \cite{32}

$$J_{Fe} = 4.96 \ meV,$$  \hspace{1cm} (D1a)
$$D^e_y = -0.107 \ meV,$$  \hspace{1cm} (D1b)
$$A_x = 0.0073 \ meV,$$  \hspace{1cm} (D1c)
$$A_z = 0.0150 \ meV,$$  \hspace{1cm} (D1d)
$$A_{xz} = 0.$$

The anisotropic $g$-factors for Er$^{3+}$ spins were assumed to be

$$g^e_{Er} = 6,$$  \hspace{1cm} (D2a)
$$g^o_{Er} = 3.4,$$  \hspace{1cm} (D2b)
$$g^z_{Er} = 9.6.$$  \hspace{1cm} (D2c)

They were roughly determined for fitting the Er$^{3+}$ spin resonance frequencies in Figs. 6, 7, and 8 to their absorption peak positions observed in our experiments \cite{32}. They are

The anisotropic $g$-factors for Fe$^{3+}$ spins were assumed to be

$$g^e_{Fe} = 2,$$  \hspace{1cm} (D3a)
$$g^o_{Fe} = 2,$$  \hspace{1cm} (D3b)
$$g^z_{Fe} = 0.6.$$  \hspace{1cm} (D3c)
anti-crossing between the Er$^{3+}$ in-phase, out-of-phase resonances, and the Fe$^{3+}$ qFM magnon mode around $B_2^{xyc} \sim 4$ T in Fig. 5 gave the most fruitful information for determining $J_{Er}$ and other parameters.

**Appendix E: Magnon quantization**

Here, we rewrite the Hamiltonian of Fe$^{3+}$ spins described by $\mathcal{H}_{Fe}$ in Eq. (5) in terms of the annihilation and creation operators of a magnon. As shown in Fig. 5, we define the modulations $\{\delta \hat{S}_{\ell,T}, \delta \hat{S}_{\ell,Y}\}$ of Fe$^{3+}$ spins from their most stable values $\hat{S}_{A/B}^\ell$ in its subsystem. The index $\ell = 2i - 1$ and $2i$ correspond to the spins at the $i$-th site in the A and B sublattices, respectively. The spin configurations are changed homogeneously in space (we set the same assumption in the mean-field calculation), and the fluctuations are approximated as the free electron $g$-factor $g$ and the Bohr magneton $\mu_B$. The coefficients $a$, $b$, $c$, and $d$ are defined in Eqs. (22) [53]. Then, the Hamiltonian of the Fe$^{3+}$ spins is approximated (bosonized) as

$$\mathcal{H}_{Fe} \approx \hbar \gamma \sum_{\ell=1}^{2N_z} \left( -\frac{a}{2} \delta \hat{S}_{\ell,Y}^2 + \frac{c}{2} \delta \hat{S}_{\ell,T}^2 + \frac{b}{2} \delta \hat{S}_{\ell,Y} \delta \hat{S}_{\ell+1,Y} + \frac{c}{2} \delta \hat{S}_{\ell+1,Y} \delta \hat{S}_{\ell,T} \right) + \text{const.}$$

(E4)

Here, $N_z$ and $2N_z$ are the number of unit cells and of Fe$^{3+}$ spins, respectively, in the $z$ direction. In terms of the annihilation operator $\hat{a}_K$ of a magnon with a dimensionless wavenumber $K$, satisfying $[\hat{a}_K, \hat{a}_K^\dagger] = \delta_{KK'}$, the modulation operators are expressed as

$$\delta \hat{S}_{\ell,T} = \frac{1}{\sqrt{2N_z}} \sum_{K=-\pi}^{\pi} e^{iK\ell} \hat{T}_K,$$

(E5a)

$$\delta \hat{S}_{\ell,Y} = \frac{1}{\sqrt{2N_z}} \sum_{K=-\pi}^{\pi} e^{iK\ell} \hat{Y}_K,$$

(E5b)

$$\hat{T}_K = \left( \frac{b \cos K - a}{d \cos K + c} \right)^{1/4} \left( \hat{a}_K^\dagger + \hat{a}_K \right),$$

(E6a)

$$\hat{Y}_K = \left( \frac{d \cos K + c}{b \cos K - a} \right)^{1/4} i(\hat{a}_K^\dagger - \hat{a}_K).$$

(E6b)

The Hamiltonian in Eq. (24) is rewritten as

$$\mathcal{H}_{Fe} \approx \hbar \omega_K \left( \hat{a}_K^\dagger + 1 \bigg| \frac{1}{2} \right) + \text{const.}$$

(E7)

Since we want to discuss a phase transition where spin configurations are changed homogeneously in space (we set the same assumption in the mean-field calculation), we focus on only the two modes with $K = 0$ and $\pi$. Then, the above Hamiltonian is approximated to Eq. (20). The fluctuations are approximated as

$$\delta \hat{S}_{2\ell-1,T} \approx \frac{1}{\sqrt{2N_z}} (\hat{T}_0 - \hat{T}_\pi),$$

(E8a)

$$\delta \hat{S}_{2\ell-1,Y} \approx \frac{1}{\sqrt{2N_z}} (\hat{Y}_0 - \hat{Y}_\pi),$$

(E8b)

$$\delta \hat{S}_{2\ell,T} \approx \frac{1}{\sqrt{2N_z}} (\hat{T}_0 + \hat{T}_\pi),$$

(E8c)

$$\delta \hat{S}_{2\ell,Y} \approx \frac{1}{\sqrt{2N_z}} (\hat{Y}_0 + \hat{Y}_\pi).$$

(E8d)

Under this approximation, the fluctuations do not depend on the index $\ell$ of unit cell. In the original $xyz$-axes shown in Fig. 5, the fluctuation vectors are expressed in Eqs. (25).
Appendix F: Aspects of phase boundaries

In Fig. 5, the phase boundaries obtained by the two approaches show small differences. The dash-dotted curves (phase transition only by the Er$^{3+}$–Er$^{3+}$ exchange interaction) are almost the same. However, the solid and dashed curves by the extended Dicke Hamiltonian are shifted to the positive side from those obtained by the mean-field approach. These shifts of the critical magnetic fields are mainly due to the neglect of $B^{DC}$-dependence of Fe$^{3+}$ spins in the derivation of the extended Dicke Hamiltonian. Then, a more sophisticated derivation of the extended Dicke Hamiltonian will resolve these differences, while it is beyond the scope of the present paper.

Note also that, in both approaches, the absolute values of the negative critical fields are larger than the positive ones for the solid and dash-dotted curves, while they are almost the same (symmetric about the origin) for the dashed curves. The symmetric phase boundary is obtained because the Er$^{3+}$ spins are not influenced by the weak magnetization of Fe$^{3+}$ spins in the absence of the Er$^{3+}$–Fe$^{3+}$ exchange interactions (Er$^{3+}$–magnon couplings). In contrast, the phase boundaries become asymmetric about the origin in the presence of the Er$^{3+}$–Fe$^{3+}$ exchange interactions (Er$^{3+}$–magnon couplings). It is for compensating the magnetization along the $a$ axis.

[1] K. Hepp and E. H. Lieb, On the superradiant phase transition for molecules in a quantized radiation field: the dicke maser model, Ann. Phys. (N. Y.) 76, 360 (1973)
[2] Y. K. Wang and F. T. Hioe, Phase transition in the dicke model of superradiance, Phys. Rev. A 7, 831 (1973)
[3] C. Ciuti, G. Bastard, and I. Carusotto, Quantum vacuum properties of the intersubband cavity polariton field, Phys. Rev. B 72, 115303 (2005)
[4] P. Forn-Díaz, L. Lamata, E. Rico, J. Kono, and E. Solano, Ultrastrong coupling regimes of light-matter interaction, Rev. Mod. Phys. 91, 025005 (2019)
[5] A. Frisk Kockum, A. Miranowicz, S. De Liberato, M. Bamba, K. Inomata, and Y. Nakamura, Superradiant phase transition without dipole approximation, Phys. Rev. Lett. 117, 173601 (2016).
[6] K. Rzążewski, K. Wódkiewicz, and W. Zakowicz, Phase Transitions, Two-Level Atoms, and the $A^2S$ Term, Phys. Rev. Lett. 35, 432 (1975)
[7] J. M. Knight, Y. Aharonov, and G. T. C. Hsieh, Are super-radiant phase transitions possible?, Phys. Rev. A 17, 1454 (1978).
[8] I. Białynicki-Birula and K. Rzążewski, No-go theorem concerning the superradiant phase transition in atomic systems, Phys. Rev. A 19, 301 (1979).
[9] K. Gawedzki and K. Rzążewski, No-go theorem for the superradiant phase transition without dipole approximation, Phys. Rev. A 23, 2134 (1981).
[10] K. Hepp, E. H. Lieb, R. Field, and K. Etudes, Equilibrium Statistical Mechanics of Matter Interacting with the Quantized Radiation Field, Phys. Rev. A 8, 2517 (1973).
[11] J. L. van Hemmen and K. Rzążewski, On the thermodynamic equivalence of the Dicke maser model and a certain spin system, Phys. Lett. A 77, 211 (1980).
[12] M. Bamba and N. Imoto, Circuit configurations which may or may not show superradiant phase transitions, Phys. Rev. A 96, 053857 (2017).
[13] J. Keeling, Coulomb interactions, gauge invariance, and phase transitions of the Dicke model, J. Phys. Condens. Matter 19, 295213 (2007).
[14] A. Vukics and P. Domokos, Adequacy of the Dicke model in cavity QED: A counter-no-go statement, Phys. Rev. A 86, 53807 (2012).
[15] A. Vukics, T. Grießer, and P. Domokos, Elimination of the A-square problem from cavity QED, Phys. Rev. Lett. 112, 136001 (2014).
[16] M. Bamba and T. Ogawa, Stability of polarizable materials against superradiant phase transition, Phys. Rev. A 90, 063825 (2014).
[17] A. Vukics, T. Grießer, and P. Domokos, Fundamental limitation of ultrastrong coupling between light and atoms, Phys. Rev. A 92, 43835 (2015).
[18] T. Grießer, A. Vukics, and P. Domokos, Depolarization shift of the superradiant phase transition, Phys. Rev. A 94, 033815 (2016).
[19] D. Hagenmüller and C. Ciuti, Cavity QED of the graphene cyclotron transition, Phys. Rev. Lett. 108, 267403 (2012).
[20] L. Chirolli, M. Polini, V. Giovannetti, and A. H. MacDonald, Drude weight, cyclotron resonance, and the dicke model of graphene cavity QED, Phys. Rev. Lett. 109, 267404 (2012).
[21] G. Mazza and A. Georges, Superradiant Quantum Materials, Phys. Rev. Lett. 122, 017401 (2019).
[22] G. M. Andolina, F. M. D. Pellegrino, V. Giovannetti, A. H. MacDonald, and M. Polini, Cavity quantum electrodynamics of strongly correlated electron systems: A no-go theorem for photon condensation, Phys. Rev. B 100, 121409 (2019).
[23] P. Nataf, T. Champel, G. Blatter, and D. M. Basko, Rashba Cavity QED: A Route Towards the Superradiant Quantum Phase Transition, Phys. Rev. Lett. 123, 207402 (2019).
[24] X. Zhang, C. L. Zou, L. Jiang, and H. X. Tang, Strongly coupled magnons and cavity microwave photons, Phys. Rev. Lett. 113, 156401 (2014).
[25] M. Goryachev, W. G. Farr, D. L. Creedon, Y. Fan,
M. Kostylev, and M. E. Tobar, High-cooperativity cavity QED with magnons at microwave frequencies, Phys. Rev. Appl. 2, 54002 (2014).

J. Bourhill, N. Kostylev, M. Goryachev, D. L. Creedon, and M. E. Tobar, Ultrahigh cooperativity interactions between magnons and resonant photons in a YIG sphere, Phys. Rev. B 93, 1 (2016).

N. Kostylev, M. Goryachev, and M. E. Tobar, Superstrong coupling of a microwave cavity to yttrium iron garnet magnons, Appl. Phys. Lett. 108 (2016).

G. Flower, M. Goryachev, J. Bourhill, and M. E. Tobar, Experimental implementations of cavity-magnon systems: From ultra strong coupling to applications in precision measurement, New J. Phys. 21, 095004 (2019).

X. Li, M. Bamba, N. Yuan, Q. Zhang, Y. Zhao, M. Xiang, K. Xu, Z. Jin, W. Ren, G. Ma, S. Cao, D. Turchinovich, and J. Kono, Observation of Dicke cooperativity in magnetic interactions, Science (80-. ). 361, 794 (2018).

Y. Tabuchi, S. Ishino, T. Ishikawa, K. Usami, and Y. Nakamura, Hybridizing ferromagnetic magnons and microwave photons in the quantum limit, Phys. Rev. Lett. 113, 83603 (2014).

Y. Tabuchi, S. Ishino, A. Noguchi, T. Ishikawa, R. Yamazaki, K. Usami, and Y. Nakamura, Coherent coupling between a ferromagnetic magnon and a superconducting qubit, Science (80-. ). 349, 405 (2015).

Y. Tabuchi, S. Ishino, A. Noguchi, T. Ishikawa, R. Yamazaki, K. Usami, and Y. Nakamura, La magnonique des quanta: Le magnon rencontre le qubit supraconducteur, Comptes Rendus Phys. 17, 729 (2016).

R. G. Morris, A. F. Van Loo, S. Kosen, and A. D. Karenowska, Strong coupling of magnons in a YIG sphere to photons in a planar superconducting resonator in the quantum limit, Sci. Rep. 7, 1 (2017).

G. Flower, J. Bourhill, M. Goryachev, and M. E. Tobar, Broadening frequency range of a ferromagnetic axion haloscope with strongly coupled cavity-magnon polaritons, Phys. Dark Universe 25, 100306 (2019).

D. Macnill, J. T. Hou, D. R. Klein, P. Zhang, P. Jarillo-Herrero, and L. Liu, Gigahertz Frequency Antiferromagnetic Resonance and Strong Magnon-Magnon Coupling in the Layered Crystal CrCl3, Phys. Rev. Lett. 123, 47204 (2019).

L. Liensberger, A. Kummer, H. Maiert-Flag, S. Geprägs, A. Erb, S. T. B. Goennenwein, R. Gross, W. Belzig, H. Huebl, and M. Weiler, Exchange-enhanced Ultrastrong Magnon-Magnon Coupling in a Compensated Ferrimagnet, Phys. Rev. Lett. 123, 117204 (2019).

D. Lachance-Quirion, S. P. Wolski, Y. Tabuchi, S. Kono, K. Usami, and Y. Nakamura, Entanglement-based single-shot detection of a single magnon with a superconducting qubit, Science (80-. ). 367, 425 (2020).

G. Gorodetskiy, R. M. Hornreich, T. Yaeger, H. Pinto, G. Schachar, and H. Shaked, Magnetic Structure of ErFeO3 below 4.5 K, Phys. Rev. B 8, 3398 (1973).

V. A. Klochan, N. M. Kvitun, and V. M. Khimara, Low-temperature spin configuration of iron ions in erbium orthoferrite, Zh. Eksp. Teor. Fiz. 68, 721 (1975).

M. Artoni and J. L. Birman, Polariton squeezing: theory and proposed experiment, Quantum Opt. J. Eur. Opt. Soc. Part B 1, 91 (1989).

P. Schwendimann and A. Quattropani, Nonclassical Properties of Polariton States, Europhys. Lett. 17, 355 (1992).

P. Schwendimann and A. Quattropani, Nonclassical Properties of Polariton States, Europhys. Lett. 18, 281 (1992).

A. Quattropani and P. Schwendimann, Polariton squeezing in microcavities, Phys. status solidi 242, 2302 (2005).

T. Makihara, K. Hayashida, G. T. Noe II, X. Li, N. M. Peraca, X. Ma, Z. Jin, W. Ren, G. Ma, I. Katayama, J. Takeda, H. Nojiri, D. Turchinovich, S. Cao, M. Bamba, and J. Kono, under reviewing (2020).

N. Marquez Peraca, X. Li, M. Bamba, C.-L. Huang, N. Yuan, X. Ma, G. T. Noe II, E. Morosan, S. Cao,
and J. Kono, Terahertz Magnon Spectroscopy Mapping of the Low-Temperature Phases of Er$_x$Y$_{1-x}$FeO$_3$, Proceedings of 2020 Conference on Lasers and Electro-Optics (CLEO), FM4D.5.

[67] J. R. Shane, Resonance frequencies of the orthoferrites in the spin reorientation region, Phys. Rev. Lett. 20, 728 (1968).

[68] L. M. Levinson, M. Luban, and S. Shtrikman, Microscopic model for reorientation of the easy axis of magnetization, Phys. Rev. 187, 715 (1969).

[69] T. Yamaguchi, Theory of spin reorientation in rare-earth orthochromites and orthoferrites, J. Phys. Chem. Solids 35, 479 (1974).

[70] A. M. Balbashov, G. V. Kozlov, A. A. Mukhin, and A. S. Prokhorov, Submillimeter Spectroscopy of Antiferromagnetic Dielectrics: Rare-Earth Orthoferrites, in High Freq. Process. Magn. Mater. (World Scientific, 1995), pp. 56–98.

[71] E. E. Zubov, V. Markovich, I. Fita, A. Wisniewski, and R. Puzniak, Magnetic order in ErFeO$_3$ single crystals studied by mean-field theory, Phys. Rev. B 99, 1 (2019).

[72] C. H. Tsang, R. L. White, and R. M. White, Spin-wave damping of domain walls in YFeO$_3$, J. Appl. Phys. 49, 6063 (1978).