Supplementary Information for "Observation of the Magnonic Dicke Superradiant Phase Transition"

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1 SRPT at finite detuning.

An anti-crossing of two polaritons occurs at the zero-detuning point ($\omega_0 = \omega_\mathrm{a}$). When the normalized coupling strength ($\eta \equiv g/\omega_0$) reaches the critical value of 0.5, the system undergoes the SRPT, and ω_- becomes zero. As shown in Fig. S1a, for $\eta = 0.5$ the SRPT occurs when $\omega_0 = \omega_\mathrm{a}$. For $\eta = 0.1$ (Fig. S1b), a situation more comparable to $ErFeO_3$, we find the phase boundary moves to the $\omega_a < \omega_0$, while the anti-crossing still occurs at the zero-detuning point. Thus one can achieve the SRPTs with a small η as long as $\nu \equiv \omega_\mathrm{a}/\omega_0$ is small enough to satisfy the inequality in Eq. (2) in main text. By contrast, when $\nu > 1$, the η_c becomes higher than 0.5.

2 THz absorption spectra at high temperatures.

Extended Data Fig. 1a shows temperature-dependent absorption spectra of the qAFM mode of Fe^{3+}. The kink occurs at 4 K which is the superradiant phase boundary at 0 T. Below this temperature, the Fe^{3+} order parameter $\langle S_y^{\mathrm{A/B}} \rangle$ becomes finite. Extended Data Fig. 1b shows magnetic field dependence of the qAFM mode of Fe^{3+} at 10 K. Only a slight change was observed at low magnetic fields without any signature of the phase transition, consistent with our phase diagram. Meanwhile, two modes that emerge at high fields are Er^{3+} EPR modes. As described in main text, $ErFeO_3$ can be modeled by the two-sublattice model. This implies we should expect four modes in total: the qFM and qAFM modes for Fe^{3+} spins, and in-phase and out-of-phase EPR modes for Er^{3+} spins. Here, we are considering the relative phase of precession of two Er^{3+} spins. A detailed derivation is in Methods and follows that in Ref.[1]. Our theory finds the lowest mode is the out-of-phase mode that is coupled to qAFM, establishing a magnon-spin system. Due to the polarization selection rule described in Fig. 2b, the qFM mode does not appear in Extended Data Fig. 1.

References

1. Bamba, M., Li, X., Marquez Peraca, N. & Kono, J. Magnonic superradiant phase transition. *Commun. Phys.* **5**, 3 (2022).

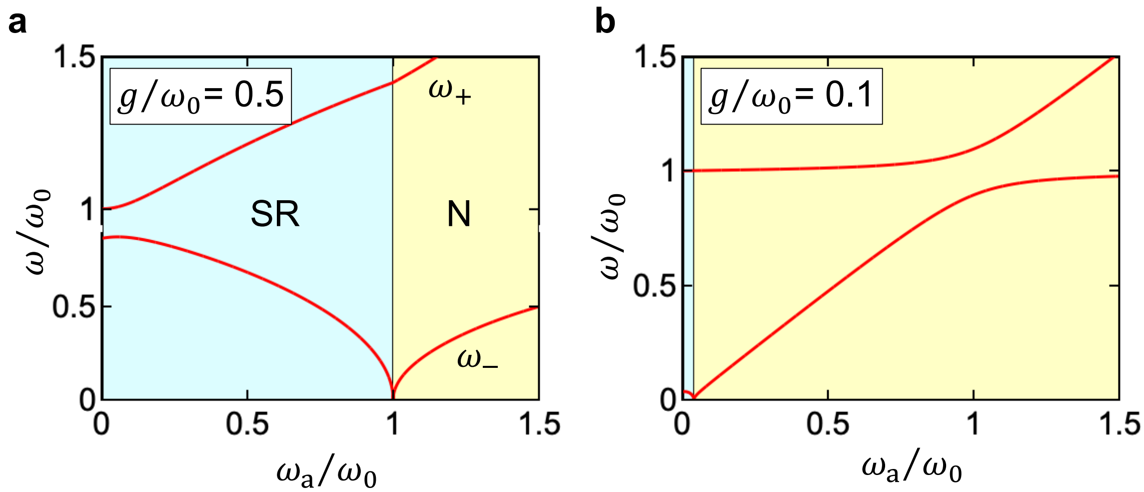

Fig. S1. Occurrence of the superradiant phase transition at finite detuning. a, b, Normalized frequencies of the upper-polariton (ω_+) and lower-polariton (ω_-) modes as a function of ω_a/ω_0 calculated using the Dicke model without the A^2 term with $g/\omega_0 = 0.5$ (**a**) and with $g/\omega_0 = 0.1$ (**b**).