MAGNETISM

Observation of icke cooperativity in magnetic interactions

Xinwei Li[1] Motoaki Bamba[2 3] Ning Yuan[4] Qi Zhang[5] Yage Zhao[6] Maolin Xiang[4] Kai Xu[4] Zuanming Jin[4] Wei Ren[4] Guohong Ma[4] Shixun Cao[4*] Dmitry Turchinovich[7 8] Junichiro Kono[1 9 10*]

The interaction of two-level atoms with a single-mode light field is an extensively studied many-body problem in quantum optics, first analyzed by Dicke in the context of superradiance. A characteristic of such systems is the cooperative enhancement of the coupling strength by a factor of $\sqrt{\ }$. In this study, we extended this cooperatively enhanced coupling to a solid-state system, demonstrating that it also occurs in a magnetic solid in the form of matter-matter interaction. Specifically, the exchange interaction of paramagnetic erbium III) Er^{3+}) spins with an iron III) Fe^{3+}) magnon field in erbium orthoferrite ErFeO$_3$) exhibits a vacuum Rabi splitting whose magnitude is proportional to $\sqrt{\ }$. Our results provide a route for understanding, controlling, and predicting novel phases of condensed matter using concepts and tools available in quantum optics.

hen an ensemble of two-level atoms interacts with a single-mode long-wavelength light field, coherence can develop within the ensemble through photon exchange; the interaction becomes cooperative. This phenomenon, captured by the Dicke superradiance model 1), has profound consequences in cavity quantum electrodynamics QED) research. In a photonic cavity, the coupling rate Λ between N dipoles and a quantized vacuum photon field is cooperatively enhanced by a factor of $\sqrt{N}$ 2). The ground state of the atom ensemble is predicted to be unstable against a phase transition, known as the superradiant phase transition SRPT), when Λ reaches a critical value 3). This possibility has stimulated much recent interest in condensed matter cavity QED systems consisting of N dipoles with very large moments 4–7).

The role light plays in the Dicke model can be performed by any fundamental excitation—such as lattice waves phonons) and spin waves magnons)—that can be bosonized and quantized in the same way as photons. This concept of Dicke physics without light is crucial for understanding phase transitions in condensed matter. The most relevant example is the cooperative Jahn-Teller JT) effect, which describes the dynamics of an ensemble of pseudospins with degenerate electronic levels cooperatively coupled with a phonon mode in the same material. The cooperative coupling leads to a phase transition that distorts the lattice and breaks the degeneracy of the pseudospin energy levels. Theoretically, the transition is believed to be analogous to the SRPT 8); the displacive lattice distortion is comparable to the appearance of a static electromagnetic field in the photon SRPT. Although these theories can explain JT and JT-like transitions phenomenologically, unambiguous evidence for the cooperative coupling of two matter subsystems in one material system is still lacking.

Here we report cooperative exchange coupling of a spin ensemble with a vacuum magnon field within a solid. We used Y^{3+}-doped single-crystal ErFeO$_3$ samples—namely, Er$_x$Y$_{1\ x}$FeO$_3$—and systematically studied the doping, temperature, and magnetic field dependence of their terahertz THz) absorption spectra. The Er^{3+} electron paramagnetic resonance EPR) in an external magnetic field strongly coupled with a vacuum magnon mode of the ordered Fe^{3+} spins. The situation is analogous to a standard N-atom cavity QED experiment, in which an ensemble of N two-level atoms couples with a vacuum photon field in an optical cavity. The Fe^{3+}-Er^{3+} coupling rate showed a characteristic scaling behavior with the density of Er^{3+} ions, evidencing Dicke cooperativity. By analyzing this scaling behavior with our microscopic theoretical model, we determined the Fe^{3+}-Er^{3+} exchange coupling constants. These constants are important for understanding the widely discussed 3d-4f magnetic coupling that is responsible for many exotic phenomena in a variety of compounds: examples include novel magnetic phase transitions 9), magnetoelectric effects 1 , 11), electromagnons 12), nonlinear spin excitations 13), and heavy fermions 14).

ErFeO$_3$ crystallizes in an orthorhombic perovskite structure Fig. 1A) that can be described by the space group D^{16}_{2h}-$Pbnm$. The Fe^{3+} spins order antiferromagnetically below 650 K. Many magnetic phases—such as the Γ_4, Γ_{24}, Γ_2, and Γ_{12} phases—can appear as a function of temperature 15). In the temperature range 4.5 K < T < 85 K, the crystal is in the Γ_2 phase, in which the spins in the two Fe^{3+} sublattices, $\mathbf{S}_1$ and $\mathbf{S}_2$, are antiparallel along the c axis but cant toward the a axis by a small angle β, owing to the antisymmetric Dzyaloshinskii-Moriya interaction Fig. 1, A and B). The sum of the two spins $\mathbf{S}_+ = \mathbf{S}_1 + \mathbf{S}_2$ induces $\mathbf{M}_{Fe} \parallel a$, where $\mathbf{M}_{Fe}$ is the macroscopic magnetization vector of the Fe^{3+} subsystem. The quasi-ferromagnetic qFM) magnon mode of the Fe^{3+} subsystem can be selectively excited by using linearly polarized THz radiation with $\mathbf{H}_{THz} \perp a$ 16, 17), where $\mathbf{H}_{THz}$ is the magnetic component of the THz electromagnetic field. Figure 1B shows how the Fe^{3+} spins oscillate in the qFM mode. $\mathbf{S}_1$ and $\mathbf{S}_2$ oscillate in phase while the angle between them remains constant, so the model can be reduced to the precession of the combined spin $\mathbf{S}_+$ about the a axis.

On the other hand, the Er^{3+} ions 4f^{11}) occupy low-symmetry sites in the crystal. The crystal field forms Kramers doublets; each doublet

[1]Department of Electrical and Computer Engineering, Rice University, Houston, TX 77005, USA. [2]Department of Materials Engineering Science, Osaka University, Osaka 560-8531, Japan. [3]PRESTO, Japan Science and Technology Agency, Saitama 332-0012, Japan. [4]Department of Physics, International Center of Quantum and Molecular Structures, and Materials Genome Institute, Shanghai University, Shanghai 200444, China. [5]Argonne National Laboratories, Lemont, IL 60439, USA. [6]School of Physics, Peking University, Beijing 100871, China. [7]Fakultät für Physik, Universität Duisburg-Essen, Lotharstr. 1, Duisburg 47057, Germany. [8]Max Planck Institute for Polymer Research, Ackermannweg 10, Mainz 55128, Germany. [9]Department of Materials Science and NanoEngineering, Rice University, Houston, TX 77005, USA. [10]Department of Physics and Astronomy, Rice University, Houston, TX 77005, USA.
*Corresponding author. Email: kono@rice.edu J.K.); sxcao@shu.edu.cn S.C.)

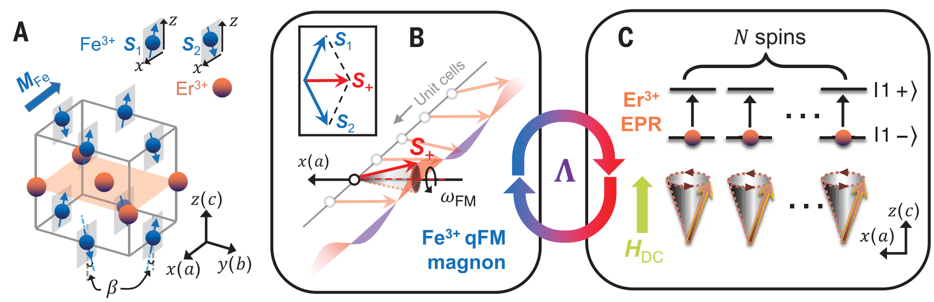

Fig. 1. Cooperatively coupled -spin magnon system as an analog of an -atom cavity QED system. A) Crystal and magnetic structure of ErFeO$_3$ in the Γ_2 phase. **B**) Oscillations of spins in the qFM magnon mode of the Fe^{3+} subsystem in the Γ_2 phase. **C**) The EPR of the Er^{3+} spin ensemble is the transition between the lowest Kramers-doublet states. The two matter subsystems, illustrated in B) and C), are resonantly coupled with the coupling constant Λ.

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