

Field dispersion and strong coupling of nuclear-electron spin excitation in MnCO_3 Takahiko Makiuchi^{1,2,*}, Takashi Kikkawa^{2,3}, Thanaporn Sichanugrist^{1,2,4}, Junki Numata^{1,2}, Masaki Imai³, Hiroyuki Chudo³, Saburo Takahashi⁵, and Eiji Saitoh^{1,2,3,5,6}¹*RIKEN Center for Emergent Matter Science (CEMS), Wako 351-0198, Japan*²*Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan*³*Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan*⁴*Department of Physics, The University of Tokyo, Tokyo 113-8656, Japan*⁵*Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*⁶*Institute for AI and Beyond, The University of Tokyo, Tokyo 113-8656, Japan*

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Hybridized nuclear and electron spin excitation in a MnCO_3 crystal, a weakly anisotropic antiferromagnet, has been investigated. In this material, the hyperfine interaction is strong enough to form a nuclear spin wave. We measure the microwave absorption induced by magnetic resonance in a bulk MnCO_3 and observe the dispersion representing strong frequency repulsion between electron and nuclear modes due to their hybridization, the signature of nuclear spin wave. Additionally, we observe that the nuclear spin resonance enters a nonlinear regime above a certain excitation power, attributed to the excitation of finite-wavenumber nuclear spin waves.

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I. INTRODUCTION

In condensed matter, nuclear spins exist in isolation and interact with neighboring electron spins through the hyperfine interaction. The hyperfine interaction has played a pivotal role in exploring electron spin states through nuclear magnetic resonance (NMR) [1]. Recently, the field of spintronics has been extended into the realm of the nucleus, a concept termed nuclear spintronics [2–7]. At the core of nuclear spintronics is the nuclear spin wave enabled by the Suhl-Nakamura interaction—an indirect interaction among nuclear spins facilitated by the exchange interaction between electron spins and the hyperfine interaction [8–10]. The nuclear spin wave operates within a megahertz frequency range [2], and its fluctuation persists even at very low temperatures [3], which distinguishes it from the electron counterpart. The conversion of nuclear spin waves into electron spin and charge currents may offer unique advantages such as extension of the frequency range and high-coherence regime.

Materials suitable for nuclear spintronics include weakly anisotropic antiferromagnets with strong hyperfine interactions [11], such as manganese carbonate MnCO_3 [2,3]. The angular momenta in such materials can travel as electron- and nuclear spin waves. The low-frequency, coupled nuclear-electron spin-resonance branches have been studied in various contexts, including the increased resonance gap for the

electron spin waves due to the hyperfine interaction with nuclear spins [12,13], the pulling effect [10,14,15], parametric excitation of finite-wavenumber nuclear spin wave [16,17], Bose-Einstein condensation of nuclear magnons [18], and coupling between nuclear spin excitation and a cavity [19,20]. However, the properties of linear and nonlinear excitations of coupled electron and nuclear spins have yet to be comprehensively studied within the same sample system, and such an investigation should be essential for the future development of nuclear spintronics.

In this study, we clarify the field dispersion for the hybridized nuclear and electron spin excitations in MnCO_3 as a function of temperature by using broadband microwave spectroscopy. The observed dispersions of electron and nuclear spins show the impact of the hybridization and a high cooperativity. Examining the temperature dependence in these dispersions, we determine various material parameters of our MnCO_3 sample. We also find a nonlinear regime in the nuclear spin-wave dispersion, indicating a formation of finite-wavenumber nuclear spin waves.

II. METHODS

MnCO_3 is an insulator whose crystalline structure belongs to the trigonal crystal system [21–23]. The rhombohedral unit cell is displayed in Fig. 1(a). A total electron spin $S = 5/2$ and a nuclear spin $I = 5/2$ (natural isotopic abundance of ^{55}Mn is 100%) are located at each Mn site and couple together through the on-site hyperfine interaction. The electron spins order into the antiferromagnetic state with a finite-canting angle due to the Dzyaloshinskii-Moriya (DM) interaction [24,25]. The electron spin orientations at different sublattices can be discussed in terms of various internal fields: the exchange field H_E , the DM field H_D , the hard-axis anisotropy

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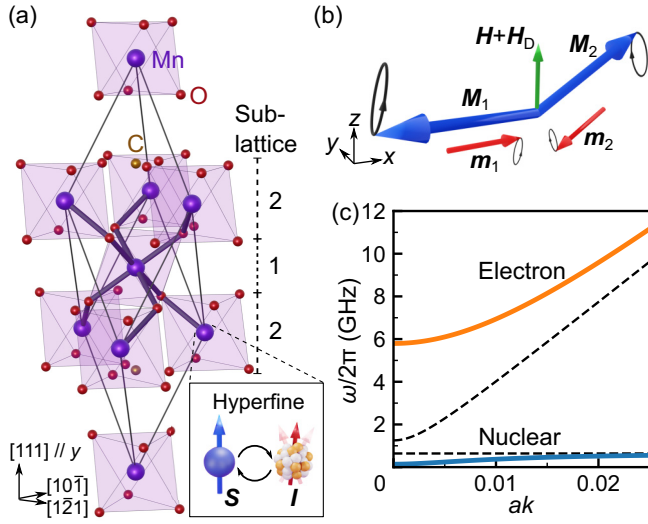


FIG. 1. (a) Crystal structure of MnCO_3 . (b) Schematic illustration of the hybridized electron and nuclear spin resonance. (c) Dispersion relations of the electron and nuclear spin excitations in MnCO_3 [Eqs. (1)–(4)]. The colored curves show hybridized dispersions, while the dashed curves show dispersions without hybridization.

field H_K ([111] direction), the easy-axis anisotropy field $H_{K'}$ ([$\bar{1}\bar{2}1$] direction), and the hyperfine (hf) field from the nuclear spin H_{hf} [14]. Meanwhile, the nuclear spins are paramagnetically polarized along the electron spin direction by the strong on-site hyperfine field of $\mu_0 H_n \approx 60$ T, where μ_0 is the vacuum permeability. The orientations of electron and nuclear magnetic moments, $\mathbf{M}_i \propto -\mathbf{S}_i$ and $\mathbf{m}_i \propto \mathbf{I}_i$, are as shown in Fig. 1(b). The subscripts $i = 1, 2$ represent the sublattice numbers. Here, we define the coordinate as $x \parallel [1\bar{2}1]$ (easy axis), $y \parallel [111]$ (hard axis), and $z \parallel [10\bar{1}]$. We apply an external field H along the z direction. Large H_E and small $H_{K'}$ orient $\mathbf{M}_i$ along the x axis, and $H + H_D$ deflects $\mathbf{M}_i$ toward the z direction. The canting angle of $\mathbf{M}_i$ from the x axis is $\psi \approx \arcsin(H + H_D)/(2H_E + H_{K'})$ under the approximation of $H_E \gg$ (other fields).

Two types of antiferromagnetic electron-spin resonance modes can be excited [4,26]. One is a precession of the net magnetization called the weak-ferromagnetic resonance (in-phase mode) and the other is a precession of the Néel vector (out-of-phase mode). We focus on the weak-ferromagnetic resonance, which significantly hybridizes with the nuclear spin excitation due to its smaller resonance gap. The coupled electron and nuclear spin excitation modes were derived from a simplified model of MnCO_3 and experimentally verified in a previous work [14]. We extend the derivations to include the finite-wavenumber k of magnon following the model. The unhybridized electron and nuclear spin excitation modes are

$$\omega_{e0}^2(k) \simeq \omega_H(\omega_H + \omega_D) + 2\omega_E(\omega_{K'} + \omega_{hf}) + \Gamma^2(k), \quad (1)$$

$$\omega_{n0} \simeq \mu_0 \gamma_n H_n, \quad (2)$$

where $\omega_H = \mu_0 \gamma_e H$, $\omega_D = \mu_0 \gamma_e H_D$, $\omega_E = \mu_0 \gamma_e H_E$, $\omega_{K'} = \mu_0 \gamma_e H_{K'}$, $\omega_{hf} = \mu_0 \gamma_e H_{hf}$, γ_e is the gyromagnetic ratio of electron, and $\gamma_n = 2\pi \times 10.553$ MHz/T is the gyromagnetic ratio

of ^{55}Mn nucleus in MnCO_3 [27]. $\Gamma^2(k) = (1 - \gamma_k)(\mu_0 \gamma_e H_E)^2$ is a k -dependent term with the form factor $\gamma_k = \frac{1}{z} \sum_{\delta} e^{i\delta \cdot \mathbf{k}} \simeq 1 - (ak)^2/6$, the location of nearest neighbors δ , and the lattice constant $a = 0.4768$ nm [22,23]. The hybridized electron and nuclear spin excitation modes read

$$\omega_e^2(k) \simeq \omega_{e0}^2(k) + G^2, \quad (3)$$

$$\omega_n^2(k) \simeq \omega_{n0}^2 - G^2, \quad (4)$$

where

$$G = \sqrt{\frac{2\omega_E \omega_{hf} \omega_{n0}^2}{\omega_{e0}^2(k) - \omega_{n0}^2}} \quad (5)$$

is a detuning parameter due to the hybridization by the transverse dynamical components of $\mathbf{S}$ and $\mathbf{I}$. The coupling diminishes as ω_{hf} approaches zero ($G \rightarrow 0$). These unhybridized/hybridized electron and nuclear spin excitation modes, described by Eqs. (1)–(4), are plotted in Fig. 1(c) using the material parameters obtained in this study (as shown later). For smaller k , the unhybridized $\omega_{e0}(k)$ approaches ω_{n0} , and $\omega_e(k)$ and $\omega_n(k)$ repel each other due to the hybridization.

We experimentally obtain the hybridized nuclear and electron spin dispersions in MnCO_3 using a broadband microwave spectroscopy technique. A bulk MnCO_3 crystal ($3 \times 3 \times 0.5$ mm³) is set on a coplanar waveguide. See Ref. [3] for the details of the characterization of the crystallographic and static magnetic properties of the sample. The sample is located in a cryostat with a superconducting magnet to apply H to the $z \parallel [10\bar{1}]$ direction. By using a network analyzer [28], we irradiate a microwave at the angular frequency ω and the input power $P_{\text{in}} = -5$ dBm to the coplanar waveguide to induce a microwave magnetic field parallel to the $x \parallel [1\bar{2}1]$ direction and measure the ratio of absorbed and input microwave powers $P_{\text{abs}}/P_{\text{in}}$. We designed two coplanar waveguides. One is used for most of the measurements and the other improved one is used for the main data in Fig. 2(a).

III. RESULTS AND DISCUSSION

A. Magnetic resonance in MnCO_3

Figures 2(a) and 2(b) show the observed $P_{\text{abs}}/P_{\text{in}}$ representing magnetic resonance in MnCO_3 at 1.8 K, well below the Néel temperature $T_N \approx 34$ K of MnCO_3 . The absorption above 6 GHz is due to the hybridized electron spin excitation mode, being almost linearly proportional to the external field H . The multiple lines can be attributed to magnetostatic standing-wave modes in the thickness direction of the sample [29,30]. The hybridized nuclear spin excitation mode appears in the low-frequency region in Fig. 2(a). Figure 2(b) is a magnified view around the low-frequency region of the spectrum shown in Fig. 2(a). The strong frequency drop near $H = 0$ is the signature of the nuclear spin-wave formation known as the frequency-pulling effect [10,14]. The nuclear branch approaches the unhybridized value $\omega_{n0}/2\pi \approx 640$ MHz at large H as the hybridization weakens due to the increasing frequency difference between nuclear and electron spin excitations. The linewidth is small at large H , whereas it broadens

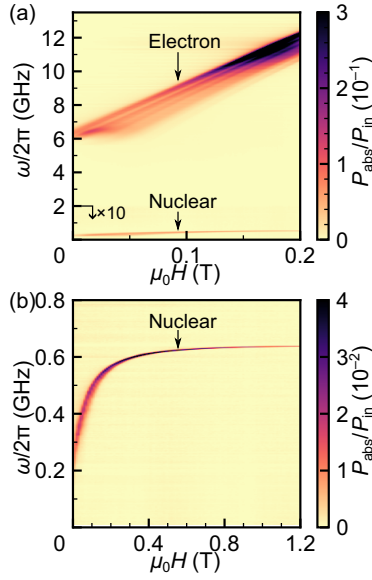


FIG. 2. (a) Observed microwave absorption by MnCO_3 at 1.8 K. The displayed P_{abs} below 2 GHz is multiplied by 10 for clarity. (b) Observed microwave absorption at 1.8 K in the low-frequency region.

at small H . The increase of linewidth at $H \approx 0$ is attributed to the significant hybridization with the dissipative electron spin excitation. Usually, the nuclear spin excitation is invisible in broadband microwave spectroscopy due to the small nuclear magnetic moment. The large absorption of the nuclear branch can be attributed to the strong hybridization with a large electron magnetic moment.

We estimate the damping coefficients, coupling constant, and cooperativity for the coupling between nuclear and electron spins from the data in Fig. 2. Figure 3 shows the main data of color-mapped microwave absorption (identical to Fig. 2) and its slice along the magnetic field and frequency axes. We assume that the four magnetostatic modes are equally separated and have the same linewidth. The electron and nuclear spin branches plotted along the angular frequency ω are fitted

to the sum of four Lorentzian functions,

$$f(\omega) = \sum_{n=0}^3 \frac{a_n \omega \kappa (\omega_0 - n \Delta \omega)^2}{[(\omega_0 - n \Delta \omega)^2 - \omega^2]^2 + (\omega_0 - n \Delta \omega)^2 \kappa^2}, \quad (6)$$

where a_n is the amplitude of the n th Lorentzian, κ is the linewidth, $\omega_0 = \omega_e$, ω_n is the resonance angular frequency, and $\Delta \omega$ is the separation. The linewidths at $H \approx 0$ are 200 MHz for the electron spin excitation mode and 100 MHz for the nuclear spin excitation mode, where the broadening of the nuclear branch can be attributed to the hybridization. The damping coefficients of $\kappa_e/2\pi \approx 200$ MHz for the electron spin excitation mode and $\kappa_n/2\pi \approx 1$ MHz for the nuclear spin excitation mode are taken from the asymptotic linewidths at large H .

We estimate the coupling constant g from a model Hamiltonian $\mathcal{H}/\hbar = \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger b + ab^\dagger)$, where ω_a , $a^\dagger$, and a are the unhybridized (uncoupled) frequency, creation, and annihilation operators for the electron spin excitation, ω_b , $b^\dagger$, and b are those for the nuclear spin excitation, and $\hbar$ is the reduced Planck constant. Diagonalizing $\mathcal{H}$ gives

$$\omega_e, \omega_n = \frac{1}{2}(\omega_a + \omega_b) \pm \frac{1}{2}\sqrt{(\omega_a - \omega_b)^2 + 4g^2}. \quad (7)$$

Using Eqs. (1)–(4), $\omega_a = \omega_{e0}$, and $\omega_b = \omega_{n0}$, we have $g/2\pi = 1.2$ GHz at 1.7 K and 0 T. As a result, the electron and nuclear spins in MnCO_3 are in the strong-coupling regime ($g > \kappa_e, \kappa_n$) [31] with the cooperativity $C = g^2/\kappa_e \kappa_n \approx 7000$. This value is greater than those of magnon-magnon ($C \sim 10$) [32,33] and magnon-phonon ($C \sim 3000$) [34] couplings, but is comparable to those of millimeter-sized cavity-magnon systems [31,35–40].

B. Temperature dependence

Figures 4(a) and 4(b) show the microwave absorption spectra at different temperatures T . The electron branch shifts toward lower frequencies as T increases. The frequency pulling and absorption intensity of the hybridized nuclear spin excitation mode weaken as T increases. The dark spot at 6 GHz and horizontal lines are attributed to the effect of microwave standing waves in the coplanar waveguide.

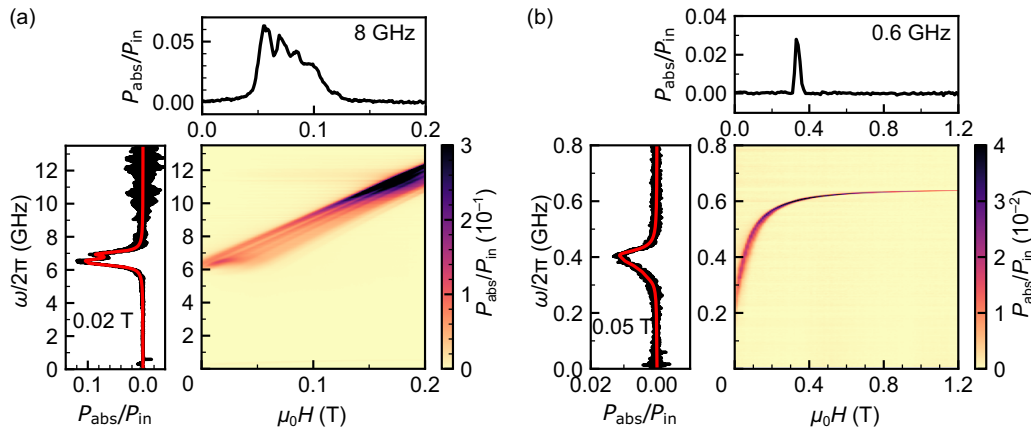


FIG. 3. (a) Two-dimensional colormap (center), frequency- (left) and field-slice (top) of the microwave absorption induced by magnetic resonances in MnCO_3 at 1.8 K. The red curve shows the multi-Lorentzian fit, Eq. (6), for the electron branch. (b) The microwave absorption in the narrow-frequency, wide-field region. The red curve shows the multi-Lorentzian fit for the nuclear branch.

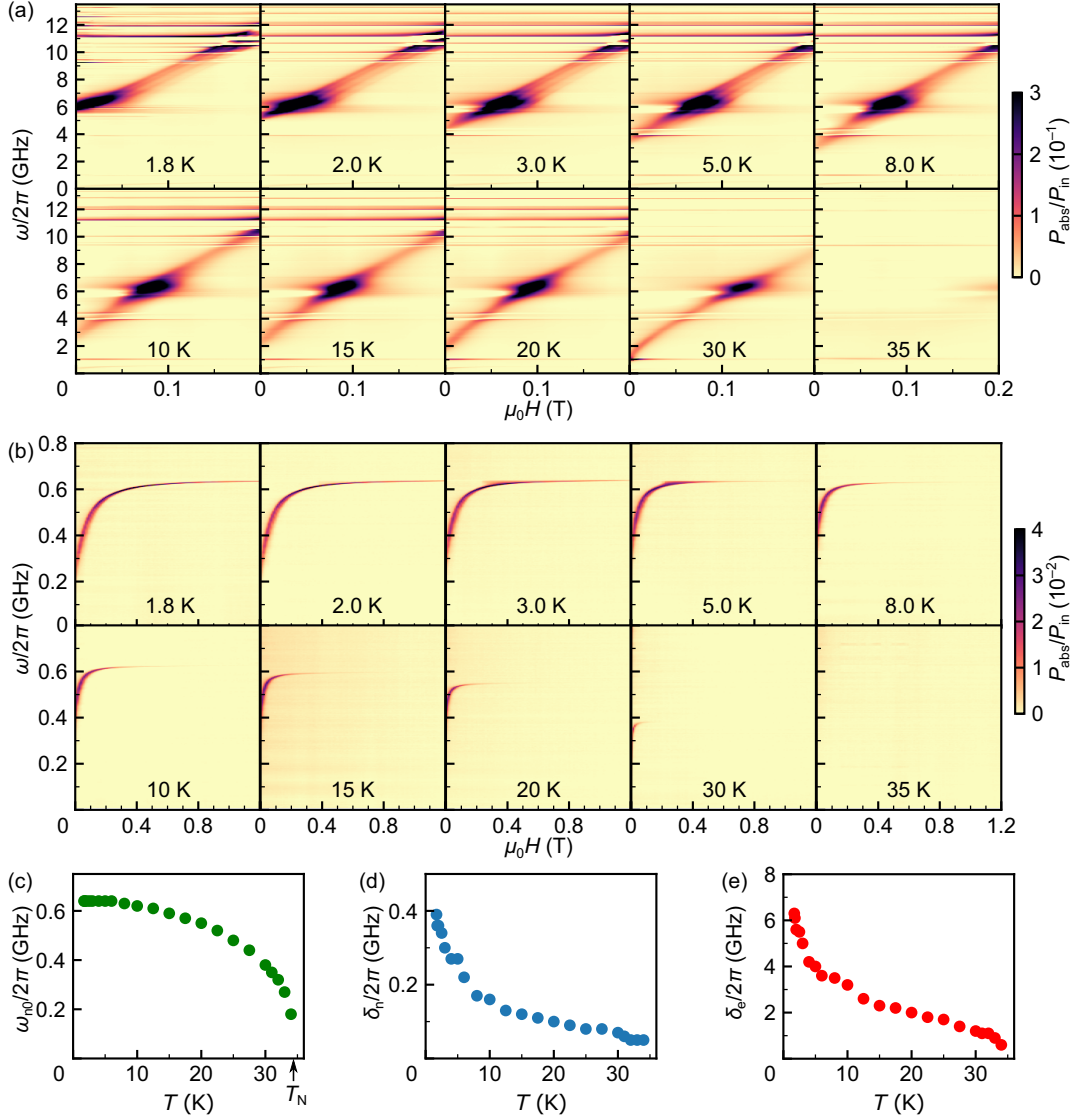


FIG. 4. (a) Microwave absorptions at different temperatures for MnCO_3 . (b) Magnified view of the microwave absorptions in the low-frequency region. (c) Temperature dependence of the nuclear spin excitation frequency at a large magnetic field (1.2 T), i.e., $\omega_{n0} \approx \lim_{H \rightarrow \infty} \omega_n$. The arrow indicates the Néel temperature $T_N = 34.4$ K. (d) Temperature dependence of the pulled frequency of the nuclear branch $\delta_n \equiv \omega_{n0} - \omega_n(H = 0)$. (e) Temperature dependence of the electron spin excitation gap $\delta_e \equiv \omega_e(H = 0)$.

The nuclear spin excitation frequency ω_{n0} in Fig. 4(c) directly gives the electron magnetic order parameter $\langle S_z \rangle$ through $\omega_{n0} = A \langle S_z \rangle / \hbar$, where A is the hyperfine coefficient. A fitting of ω_{n0} according to $\langle S_z \rangle \propto [1 - T/T_N]^\beta$ gives the Néel temperature $T_N = 34.4$ K and the critical exponent $\beta = 0.310(3)$ for the antiferromagnetic ordering. The relatively high T_N implies a good quality of the sample, compared with the reported values $T_N = 31.5\text{--}34.5$ K in Refs. [23,41–47]. The critical exponent agrees with values from experiments on weakly anisotropic antiferromagnets [23,45,48].

The pulling frequency $\delta_n \equiv \omega_{n0} - \omega_n(H = 0)$ represents the bandwidth on the nuclear spin-wave dispersion. The temperature dependence of δ_n , plotted in Fig. 4(d), represents that the hybridization rapidly increases below ~ 10 K, driven by the growing nuclear spin polarization $\langle I_z \rangle \propto 1/T$. The gap of the electron mode δ_e accordingly increases in Fig. 4(e). Although the nuclear spin polarization is $\langle I_z \rangle / I \approx 2\%$ at

$T = 1.8$ K, the pulled frequency of the nuclear branch $\delta_n/2\pi \equiv [\omega_{n0} - \omega_n(H = 0)]/2\pi \approx 400$ MHz is about 60% of the unhybridized nuclear spin excitation frequency $\omega_{n0}/2\pi = 640$ MHz. This large shift implies strong hybridization due to the exchange amplification in weakly anisotropic antiferromagnet [11,19,49,50]. This effect is described in the detuning parameter $G \propto \sqrt{2\omega_E\omega_{hf}}$, with large $\omega_E/\pi = 1.87$ THz and $\omega_{hf} \propto \langle I_z \rangle$. The full T -dependence data give the internal field values: $\mu_0 H_E = 33.4$ T, $\mu_0 H_D = 0.461$ T, $\mu_0 H_{K'} = 0.03$ mT, $\mu_0 H_{hf} = (1.1 \text{ mT})/(T/\text{K})$, and $\mu_0 H_n = 60.65$ T.

C. Nonlinear microwave absorption

We found a nonlinear regime of the hybridized nuclear spin excitation mode under the large input microwave power P_{in} . Figure 5(a) depicts the microwave absorption at different

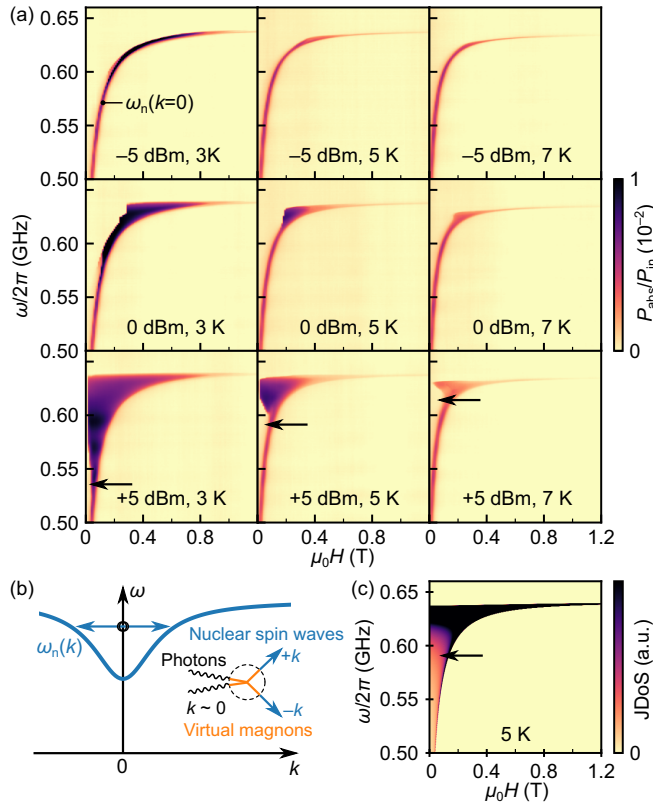


FIG. 5. (a) Observed nonlinear microwave absorption at different input microwave powers and temperatures. The arrow indicates the zero-absorption pocket (5 dBm). (b) Dispersion relation of the nuclear spin wave. The diagram shows a process for generating the finite-wavenumber nuclear spin wave. (c) Calculated joint density of state. The arrow indicates the absence of joint density of state (JDoS).

P_{in} and T . With $P_{\text{in}} = -5$ dBm and less, the nuclear branch obeys $\omega_n(k=0)$ in Eq. (4). By contrast, with $P_{\text{in}} = 0$ and +5 dBm, the absorption spread toward higher frequencies. The spread appears to be bounded by the upper limit at $\omega_{n0}/2\pi = 640$ MHz and the lower limit of $\omega_n(k=0)$. The nonlinear absorption is prominent at lower T 's, at which the hybridization is strong. Note that we swept the frequency from 0.66 to 0.50 GHz with a long dwell time to prevent history effects.

Several nonlinear responses of hybridized nuclear spin excitation mode in MnCO_3 have already been reported [14,20,50]. One possible explanation is an increase of the nuclear spin temperature T_n from the lattice and electron temperature T [14,50]. In this case, the absorption shifts toward a higher frequency. However, the absorptions in Fig. 5(a) keep the low-frequency edge at $\omega_n(k=0, T)$, meaning that $T_n \approx T$. Another possibility is Duffing (Kerr) nonlinearity of the uniform mode ($k=0$) [20]. In this case, the absorption shows a single triangular peak in the frequency sweep. However, the absence of the absorption (zero-absorption pocket) in the middle-frequency region [indicated by the arrow in Fig. 5(a) (5 dBm)] contradicts this single-peak scenario.

We attribute the observed nonlinear absorption to scattering into finite-wavenumber nuclear spin waves. Several factors can be considered to enhance the nonlinearity of nuclear spin wave excitation. (1) Strong hybridization contributes to

increased nonlinearity through virtual electron-magnon scattering processes, particularly at low temperatures and on the low-field side of the nuclear branch where the frequency pulling is significant. (2) A longer excitation lifetime also enhances nonlinearity, especially on the high-field side of the nuclear branch. (3) A high density of final states leads to stronger nonlinear effects, which occur in the high-frequency region of the nuclear branch.

Figure 5(b) illustrates a virtual electron-magnon scattering process by which finite-wavenumber nuclear spin waves are created. The rate of this process is proportional to the joint density of states (JDoS) of the hybridized nuclear spin-wave dispersion,

$$J(\omega) = \sum_{\mathbf{k}} \delta(\omega_n(\mathbf{k}) - \omega). \quad (8)$$

Figure 5(c) shows the calculated JDoS, which exhibits a dense, extended region below the upper-frequency boundary ω_{n0} , a sharp peak at the lower-frequency boundary $\omega_n(k=0)$, and a depleted region in between, as indicated by the arrow.

The experimental results at 5 and 7 K show nonlinear absorption except in the zero-absorption pocket, where the JDoS is small, suggesting that factors (2) and (3) are reflected in the observed data. However, factor (1) is essential for the emergence of nonlinearity and becomes dominant at lower temperatures. Indeed, at 3 K, the zero-absorption pocket becomes smaller, indicating that strong hybridization governs the nonlinear behavior across a broad frequency-field range.

IV. CONCLUSIONS

We measured the field dispersion of the hybridized nuclear and electron spin excitation in MnCO_3 by using the broadband microwave spectroscopy technique. We quantified the internal hyperfine coupling fields, the cooperativity, the critical exponent, and the Néel temperature from the observed dispersions. The nonlinearity in the hybridized nuclear spin excitation mode implies the generation of the finite-wavenumber nuclear spin waves with relatively small microwave fields. The high cooperativity in MnCO_3 may be useful for applications such as electric control of nuclear spin dynamics.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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