

Paramagnetic electron-nuclear spin entanglement in $\text{HoCo}_2\text{Zn}_{20}$ Takafumi Kitazawa^{1,*}, Yasuyuki Shimura,² Takahiro Onimaru², Shun Tsuchida,³ Katsunori Kubo¹, Yoshinori Haga¹, Hironori Sakai¹, Yoshifumi Tokiwa¹, Shinsaku Kambe¹, and Yo Tokunaga¹¹Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan²Department of Quantum Matter, Graduate School of Advanced Science and Engineering, Hiroshima University, Higashi-Hiroshima 739-8530, Japan³Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan

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We investigated electron-nuclear spin entanglement in the paramagnetic ground state of the Ho-based cubic compound $\text{HoCo}_2\text{Zn}_{20}$. From analyses of magnetization and specific heat data, we determined the cubic crystalline electric field (CEF) parameters, the magnetic exchange constant, and the hyperfine coupling constant between the $4f$ magnetic moment and the ^{165}Ho nuclear spin. Our results show that the Γ_5 CEF ground state is split by the hyperfine coupling, with an energy width of 1.3 K at 0 T, and that the true paramagnetic ground state is a quasisextet arising primarily from entanglement between the f -electron effective spin $S = 1$ and the ^{165}Ho nuclear spin $I = 7/2$. We further demonstrate that, depending on the CEF parameters, the paramagnetic ground state can switch to an electron-nuclear coupled dectet. These findings underscore the importance of accurately identifying the electron-nuclear level scheme for understanding the low-temperature properties of rare-earth compounds containing spin-active nuclei.

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I. INTRODUCTION

In studies of rare-earth-based compounds, the energy-level scheme at rare-earth sites—particularly the nature of the ground state, and in some cases the first excited state—provides essential information for understanding exotic phenomena that emerge at low temperatures. When discussing the level scheme at rare-earth sites, we typically begin with the ground J multiplet within the LS -coupling scheme, since the Coulomb interaction among the $4f$ electrons is much stronger than their spin-orbit coupling [1]. In crystals, the ground J multiplet is further split by the crystalline electric field (CEF). The CEF parameters have been determined using a variety of experimental probes, including specific heat, magnetization, the elastocaloric effect [2], Raman scattering [3,4], inelastic neutron scattering [5–10], and x-ray spectroscopic techniques [11–13].

Although most electronic properties of rare-earth-based compounds can be understood in terms of CEF splitting alone, it should be emphasized that, when a nuclear spin is present at the rare-earth site, the CEF states are no longer exact eigenstates even in zero magnetic field. This arises from the hyperfine coupling between the magnetic dipole

moment of the f electrons, $\mathbf{J}$, and the nuclear spin $\mathbf{I}$ at the rare-earth site, expressed as $A_{\text{HF}} \mathbf{I} \cdot \mathbf{J}$, where A_{HF} is the hyperfine coupling constant. As a result, the CEF eigenlevels are split, and each eigenstate becomes a coupled electron-nuclear state. This splitting can often be neglected in magnetically ordered rare-earth-based compounds, since their ordering temperatures are typically much higher than the energy scale of the hyperfine interaction. In contrast, the hyperfine coupling becomes significant in certain exotic materials, such as spin ices [14,15] and quantum critical materials [16–23], where localized electrons remain in the paramagnetic state even at very low temperatures. In such systems, determining the paramagnetic level scheme while accounting for the hyperfine interaction is crucial for understanding quantum phenomena.

An important material-dependent parameter in this context is the hyperfine coupling constant A_{HF} . However, experimental techniques for determining A_{HF} at rare-earth sites have so far been limited to specific microscopic probes [24,25]. Here, we demonstrate that specific heat can serve as an alternative probe for estimating A_{HF} and that the hyperfine electron-nuclear coupled wave function in the paramagnetic state can be extracted solely from macroscopic thermodynamic measurements.

As a candidate material for investigating the paramagnetic electron-nuclear state, we focused on the holmium-based compound $\text{HoTr}_2\text{Zn}_{20}$ (Tr = transition metal) for the following three reasons. (1) The ^{165}Ho isotope, which is 100% naturally abundant, has the largest nuclear spin, $I = 7/2$, among the lanthanides excluding the nonmagnetic elements La and Lu [26]. Therefore, a Schottky-like specific heat anomaly arising from large hyperfine splitting is expected to appear at relatively high temperatures. (2) The $\text{RTr}_2\text{Zn}_{20}$

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(R = rare-earth element) family crystallizes in the $\text{CeCr}_2\text{Al}_{20}$ -type structure, in which the nearest-neighbor distance between rare-earth ions is as large as 6 Å [27], implying a very low ordering temperature due to weak dipole and/or multipole interactions between rare-earth ions. In fact, although the CEF ground state is not a singlet, f -electron ordering temperatures below 1 K have been reported in several $\text{RTr}_2\text{Zn}_{20}$ compounds with Co-group elements (Tr = Co, Rh, and Ir) [28–31]. (3) The rare-earth site has cubic point symmetry T_d , indicating the absence of a nuclear electric quadrupole interaction [32]. Thus, the nuclear quadrupole effect does not need to be considered in calculations of thermodynamic quantities.

Among the $\text{HoTr}_2\text{Zn}_{20}$ compounds, $\text{HoCo}_2\text{Zn}_{20}$ is the only one for which no phase transition has been reported, although its physical properties above 1.85 K have been studied [33,34]. Thus, $\text{HoCo}_2\text{Zn}_{20}$ serves as a suitable candidate for examining the electron-nuclear state in the paramagnetic regime. In this paper, we first provide a brief overview of the experimental methods in Sec. II, and then present macroscopic measurements down to approximately 0.3 K on single crystals of $\text{HoCo}_2\text{Zn}_{20}$ in Sec. III. In Sec. IV, we introduce a model Hamiltonian for the Ho sites and determine its free parameters from the macroscopic experimental data. We then describe the electron-nuclear states at zero magnetic field in $\text{HoCo}_2\text{Zn}_{20}$. Finally, in Sec. V, we discuss the electron-nuclear level scheme in general cubic Ho compounds and the multichannel Kondo effect in realistic materials containing nuclear spins, and conclude the paper in Sec. VI.

II. EXPERIMENTAL METHODS

Single crystals of $\text{HoCo}_2\text{Zn}_{20}$ and the nonmagnetic isostructural compound $\text{LuCo}_2\text{Zn}_{20}$ were grown using the Zn self-flux method. Pure elements were weighed in the nominal ratio of R:Co:Zn = 1:2:47 (R = Ho, Lu), following Ref. [34], and placed into an aluminum crucible. Each batch of $\text{RCo}_2\text{Zn}_{20}$ was sealed in an evacuated quartz tube and heated to 1100 °C. After maintaining this temperature for 3 h, the samples were slowly cooled to 700 °C at a rate of -2 °C/h. The cubic lattice constant of $\text{HoCo}_2\text{Zn}_{20}$, determined by single-crystal x-ray diffraction using Mo K_α radiation and an R-Axis RAPID diffractometer (Rigaku), was found to be $a = 14.0344(5)$ Å. This value is consistent with $a = 14.028$ Å, extracted from Fig. 2 in Ref. [34], which shows the cubic lattice constants of $\text{RTr}_2\text{Zn}_{20}$ (R = rare earth) obtained by powder x-ray diffraction.

Magnetization measurements in the temperature range from 1.9 to 300 K were performed using a commercial dc superconducting quantum interference device magnetometer (MPMS XL7, Quantum Design). Additional magnetization measurements down to 0.27 K were carried out using a capacitive Faraday magnetometer [35] mounted on a ^3He refrigerator (Heliox 2^{VL}, Oxford Instruments). Electrical resistivity and specific heat were measured using a physical property measurement system equipped with a ^3He cooling option (PPMS DynaCool-9T, Quantum Design). Resistivity was measured using the standard four-probe method with the electrical transport option, while specific heat was measured using the thermal relaxation method.

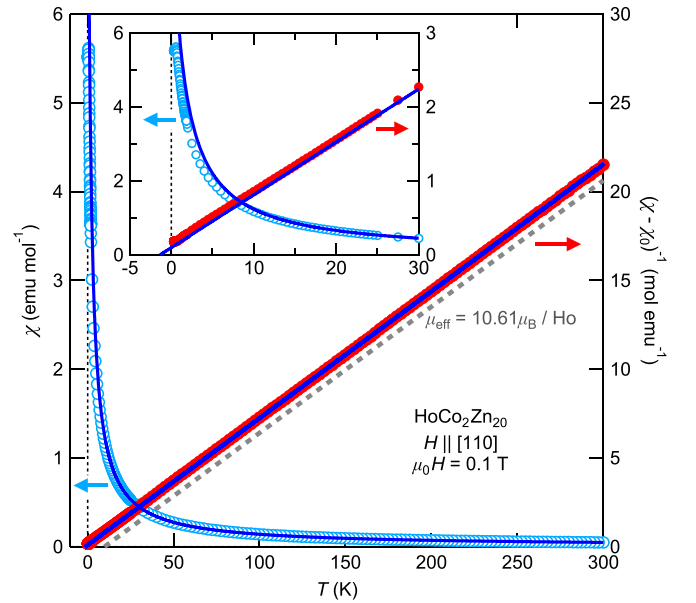


FIG. 1. Temperature dependence of the magnetization divided by the applied magnetic field $\mu_0 H = 0.1$ T, denoted as χ (left axis), and the inverse one, $(\chi - \chi_0)^{-1}$ (right axis), obtained after subtracting a temperature-independent term χ_0 , in $\text{HoCo}_2\text{Zn}_{20}$ for $H \parallel [110]$. Data for $H \parallel [100]$ and $[111]$ are presented in Fig. 9. The blue solid line represents a fit to the modified Curie-Weiss law, and the gray dashed line indicates the expected slope of the inverse susceptibility for free Ho^{3+} ions. The inset shows an enlarged view of χ and $(\chi - \chi_0)^{-1}$ below 30 K.

III. EXPERIMENTAL RESULTS

A. Overview of $\text{HoCo}_2\text{Zn}_{20}$

Figure 1 shows the temperature dependence of the magnetization divided by the applied magnetic field $\mu_0 H = 0.1$ T in

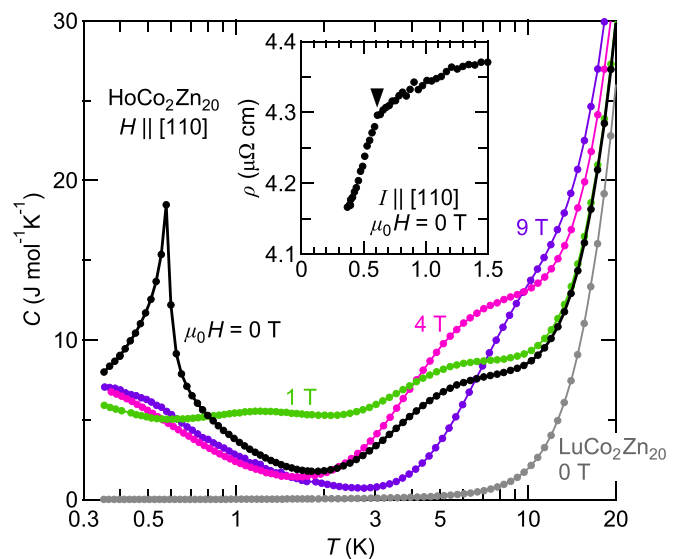


FIG. 2. Temperature dependence of the specific heat for $\text{HoCo}_2\text{Zn}_{20}$ at 0, 1, 4, and 9 T applied along the $[110]$ direction, and for $\text{LuCo}_2\text{Zn}_{20}$ at 0 T. The inset shows the electrical resistivity of $\text{HoCo}_2\text{Zn}_{20}$ at 0 T.

TABLE I. Effective magnetic moment μ_{eff} , Curie-Weiss temperature θ_C , and temperature-independent susceptibility χ_0 in $\text{HoCo}_2\text{Zn}_{20}$, obtained from fits to the magnetic susceptibility $\chi(T)$ using the modified Curie-Weiss law in the temperature range 100–300 K.

	μ_{eff} (μ_B/Ho)	θ_C (K)	χ_0 ($10^{-3} \text{ emu mol}^{-1}$)
$H \parallel [100]$	10.59(2)	−1.9(3)	2.3(1)
$[110]$	10.58(2)	−1.4(3)	2.1(1)
$[111]$	10.58(2)	−1.7(3)	5.1(1)

$\text{HoCo}_2\text{Zn}_{20}$. In this paper, we define χ as M/H for $\mu_0 H = 0.1$ T, and omit the data for $H \parallel [100]$ and $[111]$ from Fig. 1, since χ measured by MPMS is nearly isotropic as shown in Fig. 9 (Appendix A). The effective magnetic moment μ_{eff} and the Curie-Weiss temperature θ_C were evaluated by fitting the modified Curie-Weiss law, $\chi = W_C/(T - \theta_C) + \chi_0$, where W_C is the Curie constant and χ_0 is the temperature-independent magnetic susceptibility. The fitting results in the temperature range 100–300 K are summarized in Table I. The dashed line in Fig. 1, which runs parallel to the $(\chi - \chi_0)^{-1}$ data (red plot), clearly indicates that the μ_{eff} of $\text{HoCo}_2\text{Zn}_{20}$ is very close to $10.61 \mu_B/\text{Ho}$, the value expected for the ground J multiplet ($J = 8$) of the $4f^{10}$ configuration at Ho sites. This result confirms that $\text{HoCo}_2\text{Zn}_{20}$ hosts a localized magnetic moment at each Ho site. In addition to the value of μ_{eff} , the small magnitude of θ_C , which suggests a low magnetic transition temperature, is consistent with previously reported values ($\mu_{\text{eff}} = 10.7 \mu_B/\text{Ho}$, $\theta_C = 1.4$ K) [34].

Next, we present the temperature dependence of the specific heat $C(T)$ and resistivity $\rho(T)$ of $\text{HoCo}_2\text{Zn}_{20}$, shown in Fig. 2. At zero field, $C(T)$ and $\rho(T)$ exhibit a sharp peak and a kink, respectively, around 0.6 K, indicating a phase transition. Details of this transition are discussed in Sec. III B.

In addition to the phase transition, several other features appear in $C(T)$. First, $C(T)$ at 0, 1, and 4 T shows a shoulder-like anomaly near 5 K. As revealed in the CEF level-scheme analysis of Sec. IV B, this anomaly arises from thermal excitation between CEF levels. Second, $C(T)$ at 1 T does not fall below $5 \text{ J mol}^{-1} \text{ K}^{-1}$ even at the lowest measured temperature of 0.35 K, and $C(T)$ at 4 and 9 T exhibits an enhancement below about 2 K. These anomalies can be attributed to the nuclear specific heat from nuclear spins. As listed in Table II, $\text{HoCo}_2\text{Zn}_{20}$ contains nuclear spins not only in Ho

TABLE II. Isotopes with nuclear spins for Ho, Lu, Co, and Zn. Here, n_a , I , and g_N denote the natural abundance, nuclear spin, and nuclear g factor, respectively. Values of n_a and I are taken from Ref. [26], while g_N is calculated as $g_N = \mu_n/I$, where μ_n is the nuclear magnetic moment given in units of the nuclear magneton μ_N in Ref. [26].

Isotope	n_a	I	g_N
^{165}Ho	1	7/2	1.192
^{175}Lu	0.974 01	7/2	0.637 91
^{176}Lu	0.025 99	7	0.4527
^{59}Co	1	7/2	1.322
^{67}Zn	0.0404	5/2	0.350 082

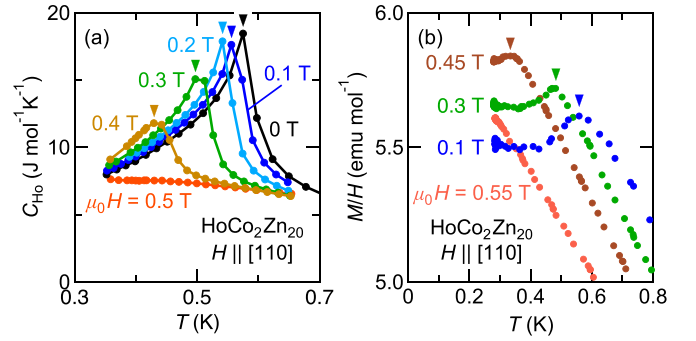


FIG. 3. Phase transition in $\text{HoCo}_2\text{Zn}_{20}$ observed in (a) the specific heat contribution from Ho sites, C_{Ho} , and (b) the magnetization divided by the applied magnetic field, M/H . The transition temperatures, indicated by downward-pointing triangles, are defined as the temperatures at which $C_{\text{Ho}}(T)$ or $M(T)/H$ exhibits a maximum.

but also in Co and Zn. However, based on the specific heat of the nonmagnetic reference compound $\text{LuCo}_2\text{Zn}_{20}$ (Fig. 11), the contributions from Co and Zn, $C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$, are clearly negligible compared with that from the Ho site. Details of the $\text{LuCo}_2\text{Zn}_{20}$ measurements are provided in Appendix B 1. To isolate the contribution of the $4f$ electrons and Ho nuclear spins, denoted by C_{Ho} , we subtracted the electronic and phonon contribution of $\text{LuCo}_2\text{Zn}_{20}$, C_{nonmag} , together with $C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$, from the total specific heat C :

$$C_{\text{Ho}} = C - (C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}). \quad (1)$$

The resulting $C_{\text{Ho}}(T)$ at 0, 1, 4, and 9 T for $H \parallel [110]$ is shown in Fig. 7(a) and is used for the level-scheme analysis in Sec. IV. The procedure for estimating $C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$ from $\text{LuCo}_2\text{Zn}_{20}$ is described in Appendix B 2.

B. Phase transition of $\text{HoCo}_2\text{Zn}_{20}$

To investigate the nature of the low-temperature phase below 0.6 K, we performed specific heat and magnetization measurements under weak magnetic fields. Figure 3(a) shows $C_{\text{Ho}}(T)$ for $H \parallel [110]$ in steps of 0.1 T. The sharp peak observed at zero field shifts to lower temperatures and broadens as the magnetic field increases. At 0.5 T, the peak in $C_{\text{Ho}}(T)$ is no longer observed down to the base temperature of 0.35 K. In the temperature dependence of the magnetization $M(T)$ for $H \parallel [110]$ at 0.1 and 0.3 T [Fig. 3(b)], a broad maximum appears at the same temperature where $C_{\text{Ho}}(T)$ shows a peak. This maximum in $M(T)$, also observed at 0.45 T, suggests the onset of antiferromagnetic (AFM) order. At 0.55 T, $M(T)$ increases monotonically upon cooling down to the base temperature of 0.28 K. We also measured the field dependence of the magnetization $M(H)$ along the $[110]$ direction at 0.29 and 2 K, as presented in Fig. 4, via the capacitive Faraday method [35]. While the differential magnetization dM/dH at 2 K decreases monotonically, dM/dH at 0.29 K gradually increases and exhibits a peak near 0.47 T (inset of Fig. 4), suggesting a weak metamagnetic transition from the AFM to the paramagnetic state. The magnetic field-temperature (H - T) phase diagram for $H \parallel [110]$, constructed from $\rho(T)$, $C_{\text{Ho}}(T)$, $M(T)$, and $M(H)$ data, is shown in Fig. 5.

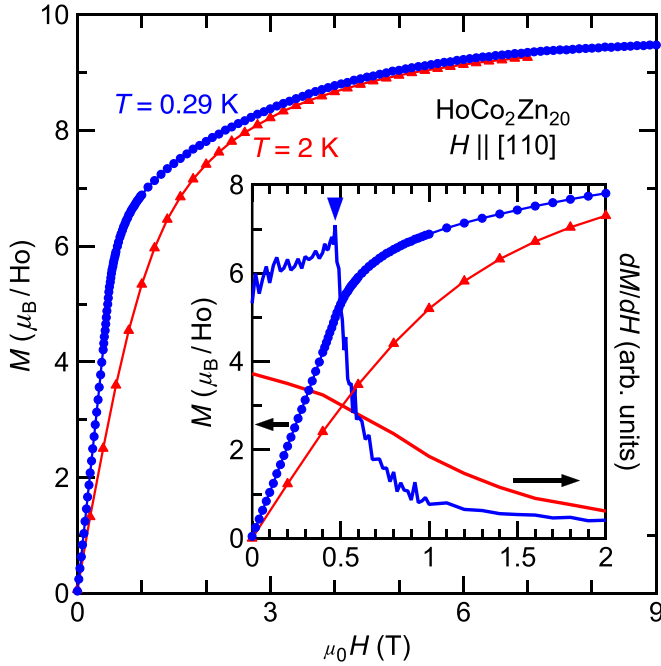


FIG. 4. Magnetization curves $M(H)$ of $\text{HoCo}_2\text{Zn}_{20}$ measured at 0.29 and 2 K for $H \parallel [110]$. The inset shows an enlarged view of $M(H)$ (lines with symbols, left axis) and the differential magnetization dM/dH (lines without symbols, right axis).

Based on Figs. 1–5 and Table I, we can conclude that the low-temperature phase of $\text{HoCo}_2\text{Zn}_{20}$ below 0.6 K at zero field is an AFM phase for the following two main reasons. First, $M(T)$ exhibits features consistent with AFM behavior across the entire measured temperature range. The

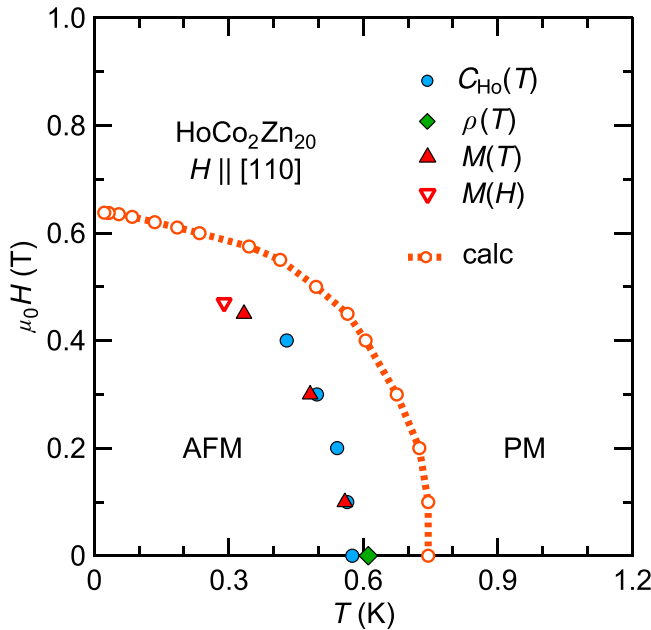


FIG. 5. H - T phase diagram of $\text{HoCo}_2\text{Zn}_{20}$ for $H \parallel [110]$. AFM and PM denote the antiferromagnetic and paramagnetic states, respectively. Open circles indicate transition temperatures calculated from Eq. (4) using the refined parameters given in Secs. IV B and IV C.

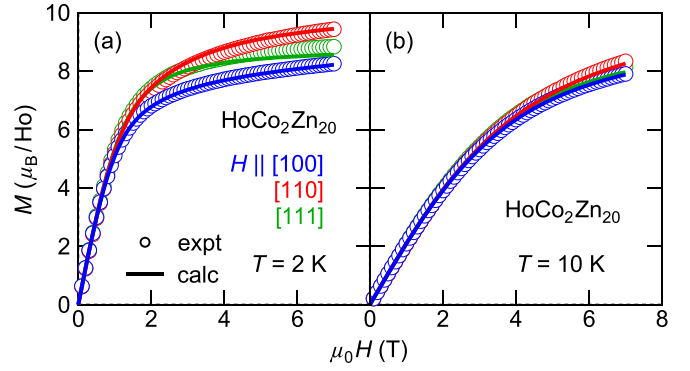


FIG. 6. Magnetization curves of $\text{HoCo}_2\text{Zn}_{20}$ at (a) 2 K and (b) 10 K. Open circles represent the experimental data, and solid lines indicate the calculated results based on the Hamiltonian excluding nuclear spin terms (see Sec. IV B for details).

Curie-Weiss temperature θ_C is negative (Table I), and $M(T)$ decreases with decreasing temperature below the transition temperature [Fig. 3(b)]. In addition, as shown in the inset of Fig. 1, $\chi(T)$ below approximately 10 K is slightly suppressed compared to the Curie-Weiss fit performed between 100 and 300 K, suggesting the development of antiferromagnetic correlations among Ho moments upon cooling. Second, the basic physical properties of $\text{HoCo}_2\text{Zn}_{20}$ reported in this section (Sec. III) closely resemble those of the antiferromagnet $\text{NdCo}_2\text{Zn}_{20}$. $\text{NdCo}_2\text{Zn}_{20}$, an isostructural compound of $\text{HoCo}_2\text{Zn}_{20}$, exhibits AFM order with a Néel temperature of $T_N = 0.53$ K [31], as confirmed microscopically by neutron scattering experiments [36]. In fact, the magnetic specific heat $C_m(T)$, resistivity $\rho(T)$, and magnetic susceptibility $\chi(T)$ near T_N in $\text{NdCo}_2\text{Zn}_{20}$ [see Figs. 5(a), 6(a), and 7 in Ref. [31], respectively] exhibit qualitatively similar behavior to $C_{\text{Ho}}(T)$, $\rho(T)$, and $M(T)/H$ in $\text{HoCo}_2\text{Zn}_{20}$. Moreover, the weak metamagnetic transition (inset of Fig. 4) and the H - T phase diagram (Fig. 5) observed in $\text{HoCo}_2\text{Zn}_{20}$ closely resemble those in $\text{NdCo}_2\text{Zn}_{20}$ [see the inset of Fig. 7 and 5(a) in Ref. [31], respectively]. Taken together, the phase transition of $\text{HoCo}_2\text{Zn}_{20}$ is understood as an AFM transition of the Ho magnetic moments.

IV. ANALYSIS OF THE LOCAL ELECTRON-NUCLEAR STATE

In this section, we determine the electron-nuclear level scheme at the Ho sites based on the experimental results of magnetization and specific heat.

A. Model

Based on recent studies of other Ho-based magnetic materials [15,37], we construct the following Hamiltonian to calculate thermodynamic quantities and determine the electron-nuclear level scheme at the Ho sites:

$$\begin{aligned} \mathcal{H} = & \sum_i \mathcal{H}_{\text{CEF}}^{\text{cubic}} + \mu_0 \mu_B g_J \mathbf{H} \cdot \sum_i \mathbf{J}_i + \mu_0 \mu_N g_N \mathbf{H} \cdot \sum_i \mathbf{I}_i \\ & + A_{\text{HF}} \sum_i \mathbf{I}_i \cdot \mathbf{J}_i - \mathcal{J} \sum_{(i,j)} \mathbf{J}_i \cdot \mathbf{J}_j. \end{aligned} \quad (2)$$

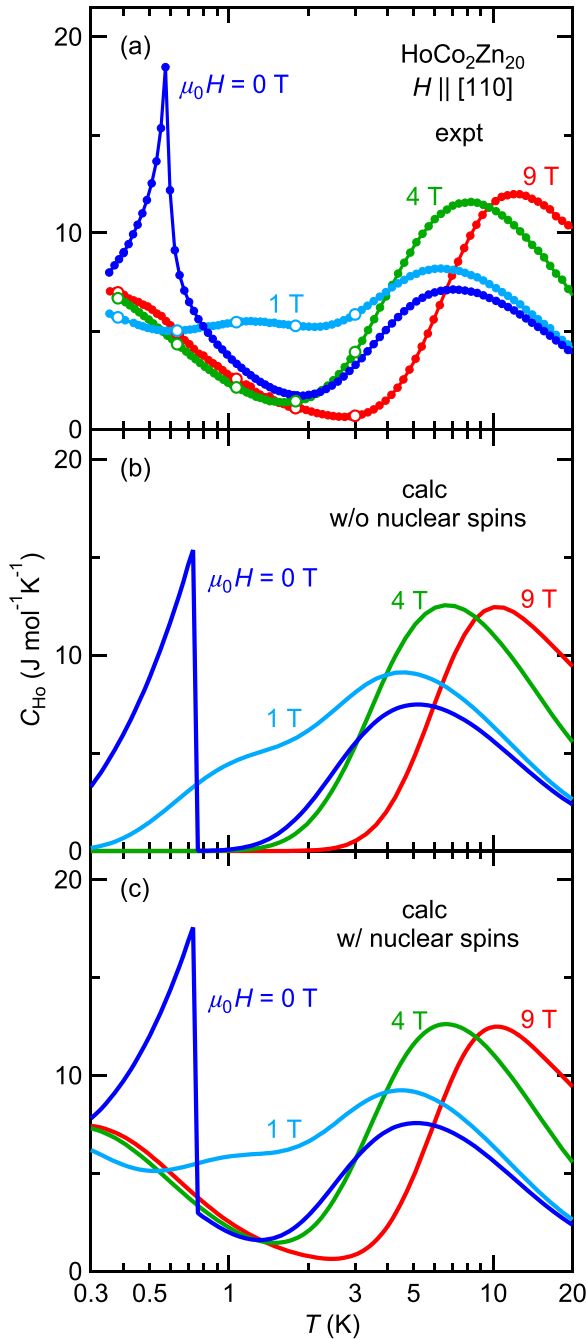


FIG. 7. Temperature dependence of the specific heat contribution from Ho ions, C_{Ho} , in $\text{HoCo}_2\text{Zn}_{20}$. Panels (a), (b), and (c) show the experimental data, the calculated results without nuclear spins, and the calculated results including nuclear spins, respectively. Open circles in panel (a) indicate the data points used to refine the hyperfine coupling constant A_{HF} (see Appendix C2 for details).

Here, the indices i label Ho sites and $\langle i, j \rangle$ denotes nearest-neighbor pairs. The five terms on the right-hand side of Eq. (2) correspond, respectively, to the crystalline electric field (CEF) term, the Zeeman term for $4f$ electrons, the nuclear Zeeman term, the hyperfine interaction, and the intersite exchange interaction. Details of each term are given below.

The term $\mathcal{H}_{\text{CEF}}^{\text{cubic}}$ represents the CEF Hamiltonian for the Ho^{3+} ion in cubic symmetry and is expressed as [38]

$$\mathcal{H}_{\text{CEF}}^{\text{cubic}} = W \left\{ x \frac{O_4^0 + 5O_4^4}{60} + (1 - |x|) \frac{O_6^0 - 21O_6^4}{13860} \right\}, \quad (3)$$

where W and x are CEF parameters, and O_n^m are Stevens operators [39,40]. The second and third terms describe the Zeeman interactions of the $4f$ electrons and the ^{165}Ho nuclear spins, respectively. Here, μ_B is the Bohr magneton, $g_J = 5/4$ is the Landé g factor for the $4f^{10}$ configuration, μ_N is the nuclear magneton, and $g_N = 1.192$ is the nuclear g factor for ^{165}Ho (Table II). The fourth term represents the hyperfine interaction between the electronic angular momentum $\mathbf{J}$ and the nuclear spin $\mathbf{I}$ at each Ho site, with A_{HF} denoting the hyperfine coupling constant. As noted in Sec. I, nuclear electric quadrupole interactions are neglected due to the cubic symmetry of the Ho site [32]. Finally, the fifth term describes an isotropic Heisenberg exchange interaction between nearest-neighbor Ho magnetic moments with the exchange constant $\mathcal{J}$.

By applying the mean-field approximation $\mathbf{J}_i \cdot \mathbf{J}_j = \mathbf{J}_i \cdot \langle \mathbf{J}_j \rangle + \langle \mathbf{J}_i \rangle \cdot \mathbf{J}_j - \langle \mathbf{J}_i \rangle \cdot \langle \mathbf{J}_j \rangle$, where $\langle \mathbf{J}_i \rangle$ denotes the canonical thermal average of the operator $\mathbf{J}_i$, the total Hamiltonian $\mathcal{H}$ for $\mathcal{J} < 0$ can be written as

$$\mathcal{H} = \sum_{i \in A} \mathcal{H}_A + \sum_{i \in B} \mathcal{H}_B, \quad (4)$$

$$\mathcal{H}_\alpha = \mathcal{H}_{\text{CEF}}^{\text{cubic}} + \mu_0(g_J\mu_B\mathbf{J}_\alpha + g_N\mu_N\mathbf{I}_\alpha) \cdot \mathbf{H} + A_{\text{HF}}\mathbf{I}_\alpha \cdot \mathbf{J}_\alpha - J_{\text{ex}}\mathbf{J}_\alpha \cdot \langle \mathbf{J}_\beta \rangle + \frac{J_{\text{ex}}}{2} \langle \mathbf{J}_\alpha \rangle \cdot \langle \mathbf{J}_\beta \rangle, \quad (5)$$

where A and B denote the two sublattices of the Ho sites, (α, β) refers to either (A, B) or (B, A) , and $J_{\text{ex}} = z\mathcal{J}$, with z being the number of nearest-neighbor sites. Since the Ho sites in $\text{HoCo}_2\text{Zn}_{20}$ form a diamond lattice, we set $z = 4$. In the case of $\mathcal{J} > 0$ ($J_{\text{ex}} > 0$), the term $\sum_{i \in B} \mathcal{H}_B$ in Eq. (4) is omitted because the system consists of a single sublattice. Accordingly, (α, β) in Eq. (5) becomes (A, A) . In this section, we employ the mean-field Hamiltonian given by Eqs. (4) and (5), using four free parameters, W , x , J_{ex} , and A_{HF} , to refine the quantum states at the Ho sites.

B. Refinement of CEF parameters and the magnetic exchange constant

We first determine the CEF parameters W and x , as well as the magnetic exchange parameter J_{ex} , based on the isothermal magnetization data at 2 and 10 K shown in Fig. 6. Since the enhancement of C_{Ho} due to nuclear spin contributions becomes significant below approximately 2 K [Fig. 7(a)], the Hamiltonian excluding nuclear spin terms is expected to be valid above this temperature. Therefore, W , x , and J_{ex} were determined using the total Hamiltonian $\mathcal{H}$ with the nuclear Zeeman term $\mu_0 g_N \mu_N \mathbf{I}_\alpha \cdot \mathbf{H}$ and the hyperfine interaction term $A_{\text{HF}} \mathbf{I}_\alpha \cdot \mathbf{J}_\alpha$ omitted from Eq. (5). Details of the analysis method are provided in Appendix C1. The best-fit parameters obtained from the analysis of $M(H)$ at 2 and 10 K are $W = 0.0443$ K, $x = -0.0640$, and $J_{\text{ex}} = -0.0511$ K. The corresponding calculated $M(H)$ curves are shown as solid lines in Fig. 6. Although J_{ex} was treated as a free parameter that could take either positive or negative values, a negative value

was obtained, consistent with the antiferromagnetic nature of $\text{HoCo}_2\text{Zn}_{20}$.

To evaluate the validity of these parameters, we calculated $C_{\text{Ho}}(T)$ using the values of W , x , and J_{ex} determined above, as shown in Fig. 7(b). Although these parameters were optimized using the experimental data at 2 and 10 K, where $\text{HoCo}_2\text{Zn}_{20}$ is in the paramagnetic state, the calculated $C_{\text{Ho}}(T)$ exhibits an antiferromagnetic transition at nearly the same temperature as observed experimentally. In addition, the Curie-Weiss temperature θ_C estimated from the mean-field exchange constant J_{ex} via $\theta_C = J(J+1)J_{\text{ex}}/3$ is -1.23 K, which is close to the experimental value obtained from $\chi(T)$ (Table I). The agreement between the calculated and experimental values of T_N and θ_C supports the validity of J_{ex} . Furthermore, the calculated $C_{\text{Ho}}(T)$ in Fig. 7(b) semiquantitatively reproduces the experimental data in Fig. 7(a) above approximately 2 K, both at zero field and under magnetic fields, confirming the reliability of the CEF parameters. The discrepancy in C_{Ho} below 2 K can be attributed to the neglect of nuclear spin effects. Therefore, in the next step, we include the nuclear Zeeman and hyperfine interaction terms, $\mu_0 g_N \mu_N \mathbf{I}_\alpha \cdot \mathbf{H}$ and $A_{\text{HF}} \mathbf{I}_\alpha \cdot \mathbf{J}_\alpha$, and refine the hyperfine coupling constant A_{HF} to reproduce the experimental $C_{\text{Ho}}(T)$ down to the lowest temperature of 0.35 K.

C. Refinement of the hyperfine coupling constant

A_{HF} was refined using the full Hamiltonian $\mathcal{H}$ in Eq. (4), including all terms in Eq. (5). In general, the specific heat calculated within the mean-field approximation exhibits a discontinuous jump at the transition temperature, as seen in $C_{\text{Ho}}(T)$ at 0 T in Fig. 7(b), due to the neglect of magnetic fluctuations. In contrast, the experimentally observed $C_{\text{Ho}}(T)$ at 0 T varies continuously, indicating the presence of magnetic fluctuations. Therefore, while the 0 T data were excluded from the refinement, $C_{\text{Ho}}(T)$ data below 3 K at 1, 4, and 9 T were used to refine A_{HF} . Since these data at finite magnetic fields are well separated from the phase boundary in the experimental H - T phase diagram (Fig. 5), magnetic fluctuations are expected to be negligible. Therefore, the mean-field treatment is sufficient for estimating A_{HF} . Details of the analysis method are provided in Appendix C2. As a result of the refinement, the hyperfine coupling constant was determined to be $A_{\text{HF}} = 0.0355$ K [41]. This value is close to the expected value of $A_{\text{HF}} = 0.0390$ K for an isolated Ho^{3+} ion [42]. The calculated $C_{\text{Ho}}(T)$ using all refined parameters, W , x , J_{ex} , and A_{HF} , is shown in Fig. 7(c), and it reproduces the experimental $C_{\text{Ho}}(T)$ well over the entire temperature range. We also note that not only the specific heat but also the magnetization (Fig. 10) and the H - T phase diagram (Fig. 5) are well reproduced using the refined parameters.

D. Level scheme of Ho sites at 0 T

Since all free parameters in Eq. (4) have been determined, we calculate the energy-level scheme of the Ho sites at 0 T in the paramagnetic state by diagonalizing the single-site Hamiltonian at 0 T, given in Eq. (5) as $\mathcal{H}^{0\text{T}} = \mathcal{H}_{\text{CEF}}^{\text{cubic}} + A_{\text{HF}} \mathbf{I} \cdot \mathbf{J}$. The resulting level scheme is shown in Fig. 8(a). In the absence of both CEF effects and hyperfine coupling, the ground state

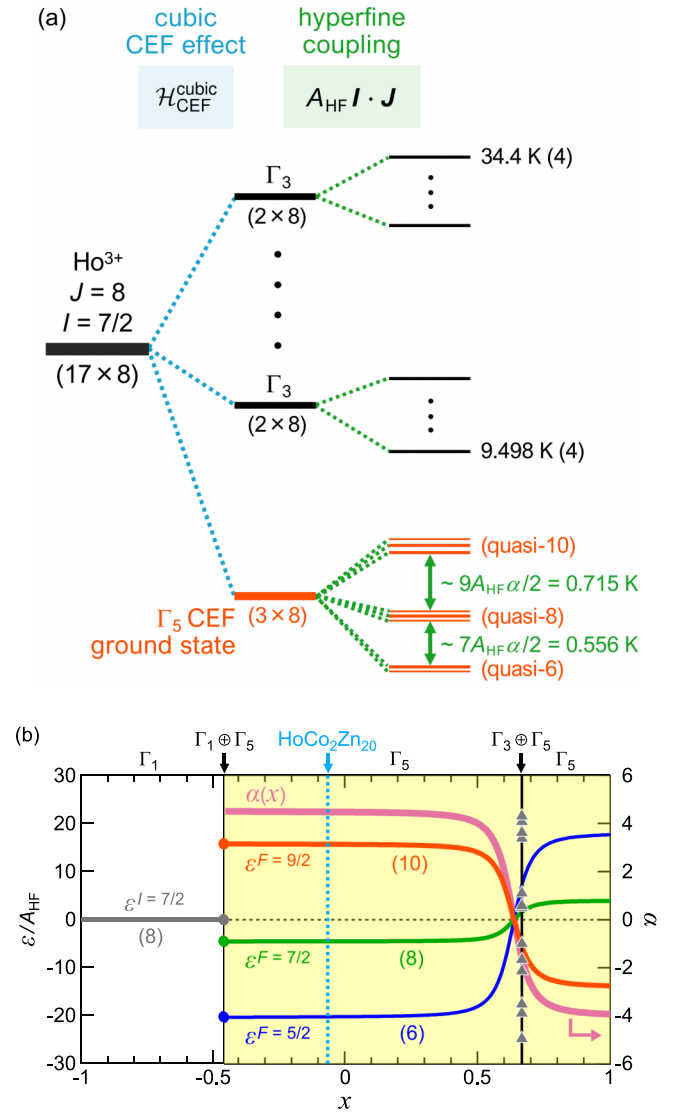


FIG. 8. (a) Schematic energy-level diagram of the Ho sites in $\text{HoCo}_2\text{Zn}_{20}$. Numbers in parentheses indicate the degeneracy of each level; the prefix “quasi” denotes quasidegenerate levels. (b) Eigenenergy diagram as a function of x for $W > 0$, restricted to the CEF ground-state subspace. Eigenenergies ε (left axis) are normalized by the hyperfine coupling constant A_{HF} . Degeneracies are again indicated in parentheses. The light-yellow background indicates the region where the CEF ground state is Γ_5 , within which $\alpha(x)$ (light-purple line, right axis) is defined. The triangle at $x = 2/3$ denotes the eigenenergy in the direct-sum space $\Gamma_3 \oplus \Gamma_5$, where the degeneracy is 2 or 4 (see Appendix D). The light-blue vertical line indicates the value of x determined for $\text{HoCo}_2\text{Zn}_{20}$ ($x = -0.0640$).

of the $4f^{10}$ configuration is a $J = 8$ multiplet, and the ^{165}Ho nuclear spin $I = 7/2$ remains degenerate. Thus, the ground state consists of $17 \times 8 = 136$ degenerate eigenstates.

Under the cubic CEF, this multiplet in the J space splits into a Γ_1 singlet, two Γ_3 doublets, two Γ_4 triplets, and two Γ_5 triplets [38]. In $\text{HoCo}_2\text{Zn}_{20}$, the CEF ground state corresponds to the irreducible representation Γ_5 . Since the CEF Hamiltonian does not include the nuclear spin operator $\mathbf{I}$, nuclear spin degeneracy is preserved, resulting in $3 \times 8 = 24$ degenerate

eigenstates in the CEF ground state. It is noteworthy that the energy separation between the Γ_5 ground state and the highest CEF level is only 33.1 K. Such a narrow level spacing is a characteristic feature of the $\text{CeCr}_2\text{Al}_{20}$ -type structure, where the rare-earth ion is enclosed in an almost spherical cage formed by 16 Zn atoms [27]. Indeed, the CEF splitting Δ_{CEF} confirmed by inelastic neutron scattering experiments is approximately 80 K in $\text{NdCo}_2\text{Zn}_{20}$ [5] and 30 K in $\text{YbCo}_2\text{Zn}_{20}$ [43].

The 24-fold degenerate CEF ground state is further split by the hyperfine coupling into four quartets and four doublets. The corresponding eigenenergies are 0 K (2), 0.83 mK (4), 0.543 K (2), 0.546 K (4), 0.551 K (2), 1.256 K (4), 1.263 K (4), and 1.265 K (2), where the numbers in parentheses denote degeneracy. One can identify quasisextet, quasioctet, and quasidectet structures near 0, 0.55, and 1.26 K, respectively. We use the prefix “quasi” to emphasize that these structures are not perfectly degenerate multiplets but are slightly split into doublets and quartets on the millikelvin or submillikelvin energy scale. The resulting doublet and quartet states are eigenstates of $\mathcal{H}^{\text{OT}}$. The origin of these quasimultiplets can be understood from the hyperfine coupling within the Γ_5 CEF ground-state subspace, as explained below. We note that a similar situation was discussed for a cubic Pr compound in Ref. [44].

Since the total hyperfine splitting within the Γ_5 ground state (1.265 K) is much smaller than the energy gap between the ground pseudosextet and the lowest multiplet split from the first excited CEF level [9.498 K; see Fig. 8(a)], it is justified to restrict the discussion to the Γ_5 subspace. As detailed in Appendix D, the angular momentum operator in this subspace can be expressed as $\mathbf{J} = \alpha(x)\mathbf{S}$, where $\alpha(x)$ depends on the cubic CEF parameter x , and $\mathbf{S}$ is an effective spin operator with $S = 1$. Accordingly, the hyperfine coupling becomes $A_{\text{HF}}\mathbf{I} \cdot \mathbf{J} = A_{\text{HF}}\alpha(x)\mathbf{I} \cdot \mathbf{S}$. By defining the total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{S}$, the 24-fold degenerate ground state is split into multiplets labeled by $F = 7/2 - 1$, $7/2$, and $7/2 + 1$, corresponding to a sextet ($F = 5/2$), an octet ($F = 7/2$), and a dectet ($F = 9/2$), respectively. Since $\mathbf{I} \cdot \mathbf{S} = (\mathbf{F}^2 - \mathbf{I}^2 - \mathbf{S}^2)/2$, the eigenenergy for each F multiplet is given by

$$A_{\text{HF}}\alpha(x) \frac{F(F+1) - I(I+1) - S(S+1)}{2} = \frac{A_{\text{HF}}\alpha(x)}{2} \left\{ F(F+1) - \frac{33}{2} \right\}. \quad (6)$$

For the ^{165}Ho isotope, A_{HF} is positive because, in the f^{10} configuration, it is given by $A_{\text{HF}} = 23g_{\text{N}}\mu_{\text{N}}\mu_{\text{B}} \langle r^{-3} \rangle / 15$ [45,46], where $g_{\text{N}} = 1.192$ (Table II) and $\langle r^{-3} \rangle$ is the average inverse cubic radius of the $4f$ orbital. Therefore, for $\alpha(x) > 0$, the ground state is the $F = 5/2$ sextet, whereas for $\alpha(x) < 0$, it becomes the $F = 9/2$ dectet. In $\text{HoCo}_2\text{Zn}_{20}$, where $\alpha(x = -0.0640) = 4.479$, the ground state is the $F = 5/2$ sextet. The first and second excited states are the $F = 7/2$ octet and $F = 9/2$ dectet, located at $7A_{\text{HF}}\alpha/2 = 0.556$ K and $8A_{\text{HF}}\alpha = 1.271$ K above the ground state, respectively. These eigenenergies closely match the quasioctet (~ 0.55 K) and quasidectet levels (~ 1.26 K) obtained from the numerical diagonalization of $\mathcal{H}^{\text{OT}}$. The further splitting of each F multiplet into doublets

and quartets on the millikelvin or submillikelvin scale likely originates from a small but finite occupancy of excited CEF states by f electrons.

V. DISCUSSION

A. Eigenenergy diagram for $W > 0$

To investigate the hyperfine level scheme in general cubic Ho compounds, we calculated $\alpha(x)$ and constructed the eigenenergy diagram over a wide range of x . Figure 8(b) shows the x dependence of the eigenenergies of $A_{\text{HF}}\mathbf{I} \cdot \mathbf{J}$ normalized by A_{HF} , i.e., $\varepsilon/A_{\text{HF}}$, for $W > 0$ within the CEF ground-state subspace, together with the variation of $\alpha(x)$. Details of the construction of this figure are provided in Appendix D. The Γ_5 state is the ground level for $-38/83 < x \leq 1$, except at $x = 2/3$. Within this range, $\alpha(x)$ changes sign at $x = x_0$ (~ 0.64), where $\alpha(x_0) = 0$, and the $F = 9/2$ dectet becomes the ground state for $x > x_0$. This result demonstrates that the hyperfine level scheme is governed not only by A_{HF} , which sets the overall energy scale of the hyperfine splitting, but also by the CEF parameter x .

B. Multichannel Kondo effect in materials containing nuclear spins

Finally, we comment on the multichannel Kondo (MCK) effect in realistic materials. The MCK effect, in which a local moment at a magnetic site is overscreened by electrons from multiple conduction bands [47], has attracted considerable attention because of the associated residual entropy [48–50], which reflects the presence of exotic quasiparticles such as Majorana and Fibonacci anyons [51]. In bulk systems, experimental evidence for the MCK effect has recently been reported in the diluted Pr compound $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$ ($x \ll 1$) [52], based on ultrasonic [53], thermal expansion [54], and magnetization [55] measurements in the temperature range of 10^1 – 10^2 mK. In this compound, the MCK effect is interpreted as a quadrupole Kondo effect, in which the electric quadrupole moment of the localized f electrons is overscreened by conduction electrons [56]. The CEF ground state of the Pr ions in $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$ is a nonmagnetic Γ_3 doublet [52,53,55], which carries an active electric quadrupole moment but no magnetic dipole moment. Furthermore, the first excited CEF state lies about 30 K above the Γ_3 ground state [52], which justifies restricting the discussion to the Γ_3 subspace when considering hyperfine splitting of the CEF ground state. Consequently, although natural Pr has a nuclear spin of $I = 5/2$ with 100% natural abundance [26], the Γ_3 ground state does not split under hyperfine coupling. Thus, the influence of hyperfine coupling can be safely neglected in the quadrupole Kondo effect of $\text{Y}_{1-x}\text{Pr}_x\text{Ir}_2\text{Zn}_{20}$.

In contrast, the magnetic MCK effect, in which a localized magnetic dipole is overscreened, has been theoretically proposed in several systems [57,58], and more recently in cubic Ho-based compounds [59,60]. According to Ref. [59], the magnetic MCK effect can be realized in cubic Ho-based compounds when the CEF ground state is the Γ_5 triplet. Since $\text{HoCo}_2\text{Zn}_{20}$ has a Γ_5 CEF ground state, diluted Ho systems $\text{R}'_{1-x}\text{Ho}_x\text{Co}_2\text{Zn}_{20}$ ($x \ll 1$), in which Ho is substituted by a nonmagnetic element R' to suppress magnetic order, are also

expected to retain the Γ_5 CEF ground state. However, due to the presence of hyperfine coupling at magnetic sites—which has not been considered in theoretical studies of the MCK effect—the Γ_5 CEF states are no longer exact eigenstates except under special conditions [61]. Whether the magnetic MCK effect can occur in the presence of hyperfine coupling remains an open question, both experimentally and theoretically. In the case of cubic Ho compounds, the diluted system $R'_{1-x}\text{Ho}_x\text{Co}_2\text{Zn}_{20}$ may serve as a promising candidate for testing the realization of the MCK effect in a system with an $F = 5/2$ sextet ground state. In Appendix E, we propose a detailed experimental procedure for capturing the residual entropy associated with the MCK effect in diluted cubic systems containing nuclear spins. At the same time, theoretical investigations are required to explore the possibility of the MCK effect when the hyperfine-split ground state is either an $F = 5/2$ sextet or an $F = 9/2$ decet.

VI. CONCLUSION

In this study, we revealed that $\text{HoCo}_2\text{Zn}_{20}$ exhibits antiferromagnetic order and determined its CEF parameters, magnetic exchange constant, and hyperfine coupling constant using macroscopic thermodynamic probes. These refined parameters clarified the hyperfine level scheme in the paramagnetic state, where the ground state is identified as an $F = 5/2$ quasisextet formed by the coupling between the f electron and the ^{165}Ho nuclear spin. It is noteworthy that the energy width of the CEF ground-state splitting due to hyperfine coupling exceeds 1 K at 0 T. This finding indicates that, if an Ho-based compound remains in the paramagnetic state down to subkelvin temperatures at 0 T, understanding the electron-nuclear level scheme may be essential for interpreting phenomena at very low temperatures, such as magnetic MCK effects [59,60] and frustrated magnetism [62,63]. Furthermore, the eigenenergy diagram of the hyperfine coupling within the subspace of the CEF ground state shows that the CEF parameters can be critically important in determining the low-energy electron-nuclear level scheme. We believe that our study on $\text{HoCo}_2\text{Zn}_{20}$ represents a first step toward determining hyperfine electron-nuclear entangled states via macroscopic measurements in a variety of materials with nuclear spins.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically

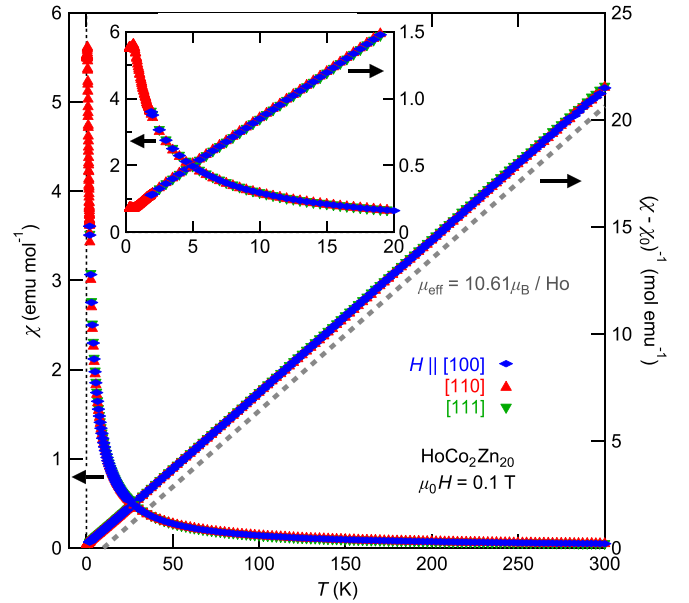


FIG. 9. Temperature dependence of χ (left axis) and $(\chi - \chi_0)^{-1}$ (right axis) in $\text{HoCo}_2\text{Zn}_{20}$ for $H \parallel [100]$, $[110]$, and $[111]$. The data for $H \parallel [110]$ are the same as those in Fig. 1. The gray dashed line indicates the expected slope of the inverse susceptibility for free Ho^{3+} ions. The inset shows an enlarged view of χ and $(\chi - \chi_0)^{-1}$ below 20 K.

feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: SUPPLEMENTAL FIGURES OF MAGNETIZATION

Figure 9 shows the temperature dependence of χ and $(\chi - \chi_0)^{-1}$ along the $[100]$, $[110]$, and $[111]$ directions, confirming the absence of magnetic anisotropy in weak magnetic fields.

Figure 10 compares the experimental results with calculations including the contribution of Ho nuclear spins to $\chi^{-1}(T)$, $\chi(T)$, and $M(H)$. The calculated curves, based on the refined parameters from Secs. IV B and IV C, reproduce the experimental data well.

APPENDIX B: SPECIFIC HEAT OF $\text{LuCo}_2\text{Zn}_{20}$

1. Experimental results

Figure 11 shows the temperature dependence of the observed specific heat, $C_{\text{obs}}(T)$, in $\text{LuCo}_2\text{Zn}_{20}$, a nonmagnetic reference compound for $\text{HoCo}_2\text{Zn}_{20}$. The magnetic field was applied along $[110]$, as in the specific heat measurements of $\text{HoCo}_2\text{Zn}_{20}$ shown in Fig. 2. Comparing C_{obs} at 0, 1, 4, 6, and 9 T at the same temperature, we find that C_{obs} increases with increasing field, and this increase becomes more pronounced at lower temperatures. These behaviors indicate the presence of nuclear specific heat arising from Lu, Co, and Zn isotopes listed in Table II. However, comparison of the vertical scales in Figs. 2(a) and 11 clearly shows that, in $\text{HoCo}_2\text{Zn}_{20}$, the nuclear contributions of Co and Zn, $C_{\text{Co}}^{\text{nuc}}$ and $C_{\text{Zn}}^{\text{nuc}}$, are much

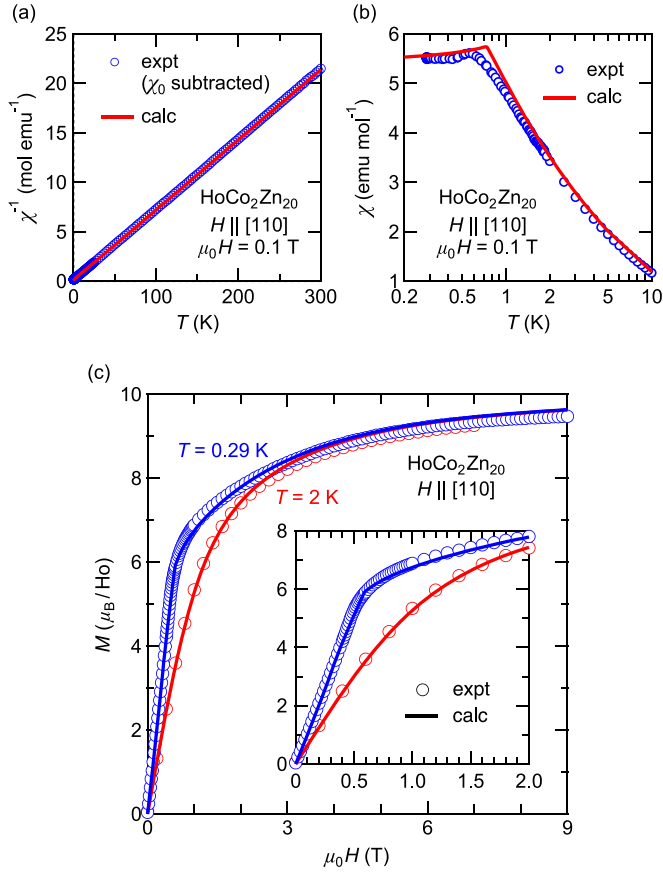


FIG. 10. Comparison of (a) $\chi^{-1}(T)$, (b) $\chi(T)$, and (c) $M(H)$ for $H \parallel [110]$ between experimental data (open circles) and calculations (solid lines) including all terms in Eq. (5). The refined parameters (W , x , J_{ex} , and A_{HF}) obtained in Secs. IV B and IV C were used in the calculations. The experimental data in panel (a) are plotted as $(\chi - \chi_0)^{-1}$. The inset in panel (c) shows an enlarged view of $M(H)$ below 2 T.

smaller than the specific heat contribution from Ho sites, C_{Ho} , at least within the temperature range of the present study.

2. Estimation of $C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$

Although $C_{\text{Lu}}^{\text{nuc}}$ in Eq. (1) is negligible in our study, $C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$ of $\text{HoCo}_2\text{Zn}_{20}$ can be estimated using the specific heat data of $\text{LuCo}_2\text{Zn}_{20}$. The observed specific heat of $\text{LuCo}_2\text{Zn}_{20}$, C_{obs} , can be decomposed into four contributions:

$$C_{\text{obs}} = C_{\text{nonmag}} + C_{\text{Lu}}^{\text{nuc}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}, \quad (\text{B1})$$

where $C_{\text{Lu}}^{\text{nuc}}$ is the nuclear specific heat of Lu. To subtract $C_{\text{Lu}}^{\text{nuc}}$ from C_{obs} , we analytically calculated $C_{\text{Lu}}^{\text{nuc}}$.

Since Lu occupies a site with cubic symmetry, the nuclear quadrupole interaction at Lu sites can be neglected [32]. Thus, the degeneracy of the Lu nuclear spins is lifted only by the nuclear Zeeman effect. For an isotope of Lu with mass number i ($= 175, 176$), the eigenenergy is $E^i = \mu_0 \mu_N g_N^i H I_z^i$ ($I_z^i = -I^i, -I^i + 1, \dots, I^i$), where g_N^i and I^i are the nuclear g factor and nuclear spin, respectively. The corresponding free energy

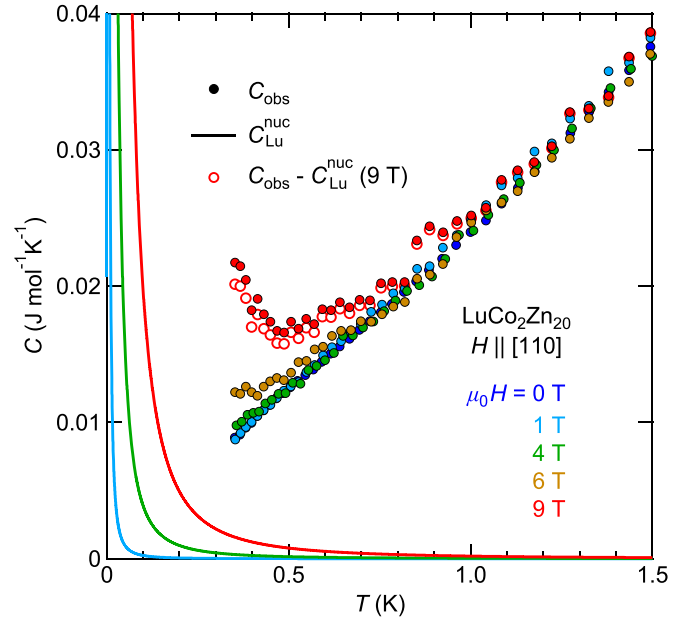


FIG. 11. Temperature dependence of the specific heat in $\text{LuCo}_2\text{Zn}_{20}$, C_{obs} (closed circles), and the nuclear specific heat contribution of Lu calculated from Eq. (B4), $C_{\text{Lu}}^{\text{nuc}}$ (solid lines). Open circles represent $C_{\text{obs}} - C_{\text{Lu}}^{\text{nuc}}$ at $\mu_0 H = 9$ T, corresponding to $C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$ at 9 T (see Appendix B 2 for details).

is

$$F^i = -k_B T \ln \sum_{I_z^i = -I^i}^{I^i} \exp(-\beta \mu_0 \mu_N g_N^i H I_z^i), \quad (\text{B2})$$

where k_B is the Boltzmann constant and $\beta = (k_B T)^{-1}$. The specific heat for each Lu isotope is then given by

$$c^i = -T \frac{\partial^2 F^i}{\partial T^2} = -\varepsilon_I^i \frac{\partial B_I(\beta \varepsilon_I^i)}{\partial T}, \quad (\text{B3})$$

where $B_I(x)$ is the Brillouin function and $\varepsilon_I^i = \mu_0 \mu_N g_N^i H I^i$. Finally, using the natural abundance n_a^i of Lu isotopes and the Avogadro constant N_A , the molar nuclear specific heat of Lu is obtained as

$$C_{\text{Lu}}^{\text{nuc}} = N_A (n_a^{175} c^{175} + n_a^{176} c^{176}). \quad (\text{B4})$$

Based on n_a^i , I^i , and g_N^i listed in Table II, we calculated $C_{\text{Lu}}^{\text{nuc}}$ at $\mu_0 H = 1, 4$, and 9 T using Eq. (B4). The results are shown as solid lines in Fig. 11. We then subtracted $C_{\text{Lu}}^{\text{nuc}}$ from C_{obs} to obtain $C_{\text{nonmag}} + C_{\text{Co}}^{\text{nuc}} + C_{\text{Zn}}^{\text{nuc}}$. Since the calculated $C_{\text{Lu}}^{\text{nuc}}$ at 1 and 4 T is nearly zero above 0.3 K, only the data for $C_{\text{obs}} - C_{\text{Lu}}^{\text{nuc}}$ at 9 T are shown in Fig. 11.

APPENDIX C: ANALYSIS METHOD

In this Appendix, we define $\mathcal{H}'_\alpha$ as $\mathcal{H}_\alpha$ with the term $J_{\text{ex}} \langle \mathbf{J}_\alpha \rangle \cdot \langle \mathbf{J}_\beta \rangle / 2$ in Eq. (5) removed.

1. Method in Sec. IV B

In Sec. IV B, the cubic CEF parameters W and x , along with the magnetic exchange constant J_{ex} , were refined by comparing the observed magnetization curves $M(H)$ at 2 and

10 K for $H \parallel [100]$, $[110]$, and $[111]$ (Fig. 6) with calculations based on a model without Ho nuclear spins. The six $M(H)$ curves, sampled every 0.5 T from 0.5 to 7 T, provided a total of 84 data points for the optimization. The magnetization per Ho site, neglecting nuclear spins, was calculated as

$$M_{\text{cal}} = -\frac{\mu_B g_J}{d} \mathbf{e}_H \cdot (\langle \mathbf{J}_A \rangle + \langle \mathbf{J}_B \rangle), \quad (\text{C1})$$

where $\mathbf{e}_H = \mathbf{H}/H$ is the unit vector along $\mathbf{H}$ and d is the number of sublattices ($d = 1$ for $J_{\text{ex}} \geq 0$ and $d = 2$ for $J_{\text{ex}} < 0$). For $J_{\text{ex}} \geq 0$, the $\langle \mathbf{J}_B \rangle$ term was omitted. We refined W , x , and J_{ex} simultaneously by minimizing the sum of squared deviations $(M_{\text{cal}} - M)^2$ over 84 data points. To locate the global optimum within $|W| \leq 0.2$ K, $|x| \leq 1$, and $|J_{\text{ex}}| \leq 0.2$ K, we employed the JADE algorithm [64], an improved variant of differential evolution [65]. The method for evaluating $\langle \mathbf{J}_A \rangle$ and $\langle \mathbf{J}_B \rangle$ under trial values of W , x , and J_{ex} generated by the JADE algorithm is described below.

To solve the self-consistent total Hamiltonian of Eq. (4) for both $J_{\text{ex}} \geq 0$ and $J_{\text{ex}} < 0$, we searched for the x , y , and z components of $\langle \mathbf{J}_A \rangle$ that minimize the free energy per Ho site:

$$F = \begin{cases} -\frac{1}{\beta} \ln Z_A + \frac{J_{\text{ex}}}{2} \langle \mathbf{J}_A \rangle^2 & (J_{\text{ex}} \geq 0), \\ -\frac{1}{2\beta} \ln(Z_A Z_B) + \frac{J_{\text{ex}}}{2} \langle \mathbf{J}_A \rangle \cdot \langle \mathbf{J}_B \rangle & (J_{\text{ex}} < 0), \end{cases} \quad (\text{C2})$$

where $Z_i = \sum_n \exp(-\beta \varepsilon_n^i)$ ($i = A, B$; ε_n^i : eigenenergy of $\mathcal{H}_i'$) is the partition function of $\mathcal{H}_i'$. In the case of $J_{\text{ex}} < 0$, $\langle \mathbf{J}_B \rangle = \text{Tr}(\mathbf{J}_B e^{-\beta \mathcal{H}_B'})/Z_B$ depends on $\langle \mathbf{J}_A \rangle$ because $\mathcal{H}_B'$ contains $\langle \mathbf{J}_A \rangle$. The optimal $\langle \mathbf{J}_A \rangle$ was obtained using the JADE algorithm under the constraint $|\langle \mathbf{J}_A \rangle| \leq J$ ($= 8$). For $J_{\text{ex}} < 0$, the procedure was as follows: (1) substitute a trial $\langle \mathbf{J}_A \rangle$ generated by JADE into $\mathcal{H}_B'$, and solve for the eigenenergies ε_n^B and eigenfunctions $|n_B\rangle$ of $\mathcal{H}_B'$; (2) calculate $\langle \mathbf{J}_B \rangle$ via

$$\langle \mathbf{J}_B \rangle = \frac{1}{Z_B} \sum_n \langle n_B | \mathbf{J}_B | n_B \rangle \exp(-\beta \varepsilon_n^B); \quad (\text{C3})$$

(3) substitute $\langle \mathbf{J}_B \rangle$ into $\mathcal{H}_A'$, and solve for the eigenenergies ε_n^A of $\mathcal{H}_A'$; (4) evaluate the free energy F using Eq. (C2); and (5) repeat steps (1)–(4) until convergence within the JADE algorithm. For $J_{\text{ex}} \geq 0$, step (1) was applied with the suffix “B” replaced by “A”, and steps (2) and (3) were omitted. Because $J = 8$ and nuclear spins were neglected, the dimension of each $\mathcal{H}_\alpha'$ matrix was 17.

With the refined parameters $W = 0.0443$ K, $x = -0.0640$, and $J_{\text{ex}} = -0.0511$ K, we then calculated the specific heat contribution from Ho sites, $C_{\text{Ho}}(T)$, shown in Fig. 7(b). Since the entropy of Ho sites is given by $S(T) = \int_0^T C_{\text{Ho}}(T)/T dT$, the calculated $C_{\text{Ho}}(T)$ was obtained from

$$C_{\text{Ho}}^{\text{cal}}(T) = T \frac{S(T + \Delta T) - S(T - \Delta T)}{2\Delta T}, \quad (\text{C4})$$

with a temperature increment $\Delta T = 0.5$ mK. Here, $S(T \pm \Delta T)$ was evaluated using the calculated F from Eq. (C2) and the thermal average of the energy per Ho site,

$$E = \frac{1}{2} \sum_{i=A,B} \sum_n \frac{\varepsilon_n^i e^{-\beta \varepsilon_n^i}}{Z_i}, \quad (\text{C5})$$

via the relation $S(T) = (E(T) - F(T))/T$.

2. Method in Sec. IV C

In Sec. IV C, the hyperfine coupling constant A_{HF} was refined by comparing the specific heat $C_{\text{Ho}}(T)$ below 3 K at 1, 4, and 9 T. As shown by the open circles in Fig. 7(a), five equally spaced interpolated points of $C_{\text{Ho}}(T)$ on a logarithmic temperature scale between 0.38 and 3 K were used for this refinement. We optimized A_{HF} with the JADE algorithm by minimizing the sum of squared deviations $(C_{\text{Ho}}^{\text{cal}} - C_{\text{Ho}})^2$ over 15 data points, where $C_{\text{Ho}}^{\text{cal}}$ was calculated using Eq. (C4). The search range was set not to $|A_{\text{HF}}| \leq 2000$ MHz (0.096 K) but to $0 \leq A_{\text{HF}} \leq 2000$ MHz, since $A_{\text{HF}} = 23 g_N \mu_N \mu_B \langle r^{-3} \rangle / 15$ [45,46] is positive, as mentioned in Sec. IV D. In these calculations, the refined values of W , x , and J_{ex} obtained in Appendix C 1 were fixed. Unlike Appendix C 1, the nuclear Zeeman term $\mu_0 g_N \mu_N \mathbf{I}_\alpha \cdot \mathbf{H}$ and the hyperfine interaction term $A_{\text{HF}} \mathbf{I}_\alpha \cdot \mathbf{J}_\alpha$ were included in $\mathcal{H}_\alpha'$. The solution of the total Hamiltonian [Eq. (4)] followed the procedure described in the second paragraph of Appendix C 1. Since the system considered consists of the $I = 7/2$ nuclear spin coupled with f electrons of $J = 8$, the dimension of each $\mathcal{H}_\alpha'$ matrix was $8 \times 17 = 136$.

With the refined value $A_{\text{HF}} = 0.0355$ K, we calculated the specific heat and magnetization, shown in Figs. 7(c) and 10, respectively. The magnetization per Ho site was given by

$$M_{\text{cal}} = -\frac{\mu_B}{2} \mathbf{e}_H \cdot \sum_{i=A,B} (g_J \langle \mathbf{J}_i \rangle + g_N \langle \mathbf{I}_i \rangle), \quad (\text{C6})$$

where $\langle \mathbf{I}_i \rangle$ is the thermal average of $\mathbf{I}_i$. Finally, in the H – T phase diagram of Fig. 5, we plotted the transition temperature at which $\langle \mathbf{J}_A \rangle = \langle \mathbf{J}_B \rangle$ changes to $\langle \mathbf{J}_A \rangle \neq \langle \mathbf{J}_B \rangle$.

APPENDIX D: SPLITTING OF THE CEF GROUND LEVEL DUE TO HYPERFINE COUPLING

In this Appendix, we explain how to construct the energy diagram shown in Fig. 8(b). When the cubic CEF parameter W is positive, the CEF ground state for $J = 8$ can be classified into four cases depending on the other cubic CEF parameter x : (1) a Γ_1 singlet, (2) a Γ_5 triplet, (3) a direct sum of a Γ_1 singlet and a Γ_5 triplet, and (4) a direct sum of a Γ_3 doublet and a Γ_5 triplet. These cases are realized for (1) $-1 \leq x < -38/83$, (2) $-38/83 < x \leq 1$ with $x \neq 2/3$, (3) $x = -38/83$, and (4) $x = 2/3$, respectively. The f -electron wave functions for the Γ_1 , Γ_3 , and Γ_5 CEF states can be expressed in terms of the J_z components [38]:

$$|\Gamma_1\rangle = \frac{\sqrt{390}}{48}(|8\rangle + |-8\rangle) + \frac{\sqrt{42}}{24}(|4\rangle + |-4\rangle) + \frac{\sqrt{33}}{8}|0\rangle, \quad (\text{D1})$$

$$|\Gamma_3; \alpha\rangle = a_1(x)(|8\rangle + |-8\rangle) + a_2(x)(|4\rangle + |-4\rangle) + a_3(x)|0\rangle, \quad (\text{D2})$$

$$|\Gamma_3; \beta\rangle = b_1(x)(|6\rangle + |-6\rangle) + b_2(x)(|2\rangle + |-2\rangle), \quad (\text{D3})$$

$$|\Gamma_5; \pm\rangle = c_1(x)|\pm 7\rangle + c_2(x)|\pm 3\rangle + c_3(x)|\mp 1\rangle + c_4(x)|\mp 5\rangle, \quad (\text{D4})$$

$$|\Gamma_5; 0\rangle = d_1(x)(|6\rangle - |-6\rangle) + d_2(x)(|2\rangle - |-2\rangle), \quad (\text{D5})$$

where the coefficients a_i , b_i , c_i , and d_i depend on x . Below, we consider a composite system consisting of the nuclear spin space $I = 7/2$ of the ^{165}Ho isotope and the CEF ground subspace corresponding to each of the above cases, and describe how the ground multiplet is split by the hyperfine coupling $A_{\text{HF}} \mathbf{I} \cdot \mathbf{J}$.

(1) Γ_1 subspace: Since the magnetic dipole moment is inactive in the Γ_1 state, the f electrons do not couple to the nuclear spin via hyperfine interaction. Therefore, the eightfold nuclear-spin multiplet remains unsplit, and the eigenfunctions are simply given by the product states $|\Gamma_1\rangle|I_z\rangle$ with $I_z = -7/2, -5/2, \dots, 7/2$.

(2) Γ_5 subspace: We numerically calculated the matrix elements of J_z , the raising operator $J_+ (= J_x + iJ_y)$, and the lowering operator $J_- (= J_x - iJ_y)$ in the Γ_5 state space. These operators share the same coefficient $\alpha(x)$:

$$J_z = \alpha(x) \begin{pmatrix} \langle \Gamma_5; + | & \langle \Gamma_5; 0 | & \langle \Gamma_5; - | \end{pmatrix} \begin{pmatrix} |\Gamma_5; +\rangle & |\Gamma_5; 0\rangle & |\Gamma_5; -\rangle \\ 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad (\text{D6})$$

$$J_+ = J_-^\dagger = \alpha(x) \begin{pmatrix} \langle \Gamma_5; + | & \langle \Gamma_5; 0 | & \langle \Gamma_5; - | \end{pmatrix} \begin{pmatrix} |\Gamma_5; +\rangle & |\Gamma_5; 0\rangle & |\Gamma_5; -\rangle \\ 0 & \sqrt{2} & 0 \\ 0 & 0 & \sqrt{2} \\ 0 & 0 & 0 \end{pmatrix}. \quad (\text{D7})$$

The 3×3 matrices in Eqs. (D6) and (D7) correspond to the z component S_z and the raising operator S_+ , respectively, in the $S = 1$ spin space. Thus, $\mathbf{J}$ can be expressed as $\mathbf{J} = \alpha(x) \mathbf{S}$. The numerically obtained values of $\alpha(x)$ are plotted in Fig. 8(b) as the light-purple line.

By coupling the effective spin $S = 1$ with the nuclear spin $I = 7/2$ via the hyperfine interaction $A_{\text{HF}}\alpha(x)\mathbf{I} \cdot \mathbf{S}$, the 24-fold multiplet splits into a sextet, an octet, and a dactet. As stated in Sec. IV D, each multiplet is characterized by the new total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{S}$. We calculated the eigenenergies for each multiplet using Eq. (6), and plotted the $F = 5/2$ sextet, $F = 7/2$ octet, and $F = 9/2$ dactet in Fig. 8(b) as blue, green, and orange lines, respectively. We note that the wave functions of each multiplet can be expressed using the Clebsch-Gordan coef-

ficients $\langle I = 7/2, I_z, S = 1, S_z | F, F_z \rangle$. For example, the $F_z = \pm 1/2, \pm 3/2, \pm 5/2$ wave functions of the $F = 5/2$ sextet can be written as

$$|F_z = \pm 1/2\rangle = \sqrt{\frac{5}{14}} |\pm 3/2, \mp\rangle - \sqrt{\frac{3}{7}} |\pm 1/2, 0\rangle + \sqrt{\frac{3}{14}} |\mp 1/2, \pm\rangle, \quad (\text{D8})$$

$$|F_z = \pm 3/2\rangle = \frac{1}{2} \sqrt{\frac{15}{7}} |\pm 5/2, \mp\rangle - \sqrt{\frac{5}{14}} |\pm 3/2, 0\rangle + \frac{1}{2} \sqrt{\frac{3}{7}} |\pm 1/2, \pm\rangle, \quad (\text{D9})$$

$$|F_z = \pm 5/2\rangle = \frac{\sqrt{3}}{2} |\pm 7/2, \mp\rangle - \sqrt{\frac{3}{14}} |\pm 5/2, 0\rangle + \frac{1}{2\sqrt{7}} |\pm 3/2, \pm\rangle, \quad (\text{D10})$$

where the basis states on the right-hand side of Eqs. (D8)–(D10), $|I_z, S_z\rangle$, denote the product states of the nuclear spin with I_z and the f -electron effective spin with S_z ($S_z = \pm, 0$ correspond to $|\Gamma_5; \pm\rangle$ and $|\Gamma_5; 0\rangle$, respectively).

(3) $\Gamma_1 \oplus \Gamma_5$ subspace: The coefficients of the Γ_5 wave functions, $c_i(x)$ and $d_i(x)$, for $x = -38/83$ are given by

$$(c_1, c_2, c_3, c_4) = \frac{C}{64} (95\sqrt{65}, 59\sqrt{105}, -113\sqrt{11}, -13\sqrt{91}), \quad (\text{D11})$$

$$(d_1, d_2) = C \left(\sqrt{39}, \frac{\sqrt{385}}{2} \right), \quad (\text{D12})$$

with $C = \sqrt{2/541}$. At this parameter, the matrices of J_z and $J_\pm$ in the $\Gamma_1 \oplus \Gamma_5$ subspace are

$$J_z = \begin{pmatrix} \langle \Gamma_1 | & \langle \Gamma_5; + | & \langle \Gamma_5; 0 | & \langle \Gamma_5; - | \end{pmatrix} \begin{pmatrix} |\Gamma_1\rangle & |\Gamma_5; +\rangle & |\Gamma_5; 0\rangle & |\Gamma_5; -\rangle \\ 0 & 0 & 0 & 0 \\ 0 & \alpha' & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\alpha' \end{pmatrix} \quad (\text{D13})$$

and

$$J_+ = J_-^\dagger = \begin{pmatrix} \langle \Gamma_1 | & \langle \Gamma_5; + | & \langle \Gamma_5; 0 | & \langle \Gamma_5; - | \end{pmatrix} \begin{pmatrix} |\Gamma_1\rangle & |\Gamma_5; +\rangle & |\Gamma_5; 0\rangle & |\Gamma_5; -\rangle \\ 0 & 0 & 0 & 0 \\ 0 & 0 & \alpha'\sqrt{2} & 0 \\ 0 & 0 & 0 & \alpha'\sqrt{2} \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad (\text{D14})$$

with $\alpha' = 77899/17312 \approx 4.50$. Because the off-diagonal block matrices in Eqs. (D13) and (D14) vanish, the Γ_1 and Γ_5 subspaces can be treated independently. Therefore, the composite system reduces to two independent

cases: the coupling of the $I = 7/2$ nuclear spin with the Γ_1 state [case (1)] and with the Γ_5 state [case (2)]. As a result, the 32-fold manifold splits into $F = 5/2, 7/2$, and $9/2$ multiplets, together with a noninteracting octet.

The eigenenergies of these F multiplets, obtained from Eq. (6) by substituting $\alpha = \alpha'$, and the eigenenergy of the noninteracting octet are plotted as closed circles in Fig. 8(b).

(4) $\Gamma_3 \oplus \Gamma_5$ subspace: The coefficients of the Γ_3 and Γ_5 wave functions for $x = 2/3$ are

$$(a_1, a_2, a_3) = \frac{1}{16} \left(\frac{\sqrt{5}}{2}, \sqrt{91}, -\frac{\sqrt{286}}{2} \right), \quad (\text{D15})$$

$$(b_1, b_2) = (d_1, d_2) = \left(\frac{1}{\sqrt{2}}, 0 \right), \quad (\text{D16})$$

$$(c_1, c_2, c_3, c_4) = \frac{1}{32\sqrt{2}} (3\sqrt{15}, \sqrt{455}, -\sqrt{429}, 7\sqrt{21}). \quad (\text{D17})$$

In contrast to case (3), the off-diagonal block matrices of J_z and $J_+ (= J_-^\dagger)$ contain nonzero elements:

$$\langle \Gamma_3; \alpha | J_z | \Gamma_5; 0 \rangle = 6, \quad (\text{D18})$$

$$\langle \Gamma_3; \alpha | J_+ | \Gamma_5; - \rangle = \langle \Gamma_5; + | J_+ | \Gamma_3; \alpha \rangle = 3\sqrt{2}, \quad (\text{D19})$$

$$\langle \Gamma_3; \beta | J_+ | \Gamma_5; + \rangle = \langle \Gamma_5; - | J_+ | \Gamma_3; \beta \rangle = 3\sqrt{6}. \quad (\text{D20})$$

Therefore, we must consider the composite state formed by coupling the $I = 7/2$ nuclear spin with the direct-sum space $\Gamma_3 \oplus \Gamma_5$. Numerical diagonalization of the $A_{\text{HF}} \mathbf{I} \cdot \mathbf{J}$ matrix of size 40×40 shows that the 40-fold manifold splits into seven quartets and six doublets. The eigenenergies, shown as triangles in Fig. 8(b), are $\varepsilon/A_{\text{HF}} = -24.7$ (4), -19.4 (2), -17.5 (4), -10.6 (2), -8.2 (4), -4.9 (2), 2.5 (2), 2.9 (4), 5.5 (4), 16.9 (2), 18.1 (4), 20.4 (2), and 21.6 (4), where the numbers in parentheses indicate degeneracy.

APPENDIX E: EXPERIMENTAL PROPOSAL FOR CAPTURING THE RESIDUAL ENTROPY

In this Appendix, we outline an experimental strategy for capturing the residual entropy, which is one of the most compelling experimental signatures of the MCK effect, in diluted cubic systems containing nuclear spins at magnetic sites. At 0 T, the MCK effect may occur because the electron-nuclear spin degrees of freedom remain active owing to the degeneracy of the hyperfine-coupled states. In contrast, under finite magnetic fields, the MCK effect is not expected, since the Zeeman splitting removes the degeneracy of these states. In this regime, the Hamiltonian for each magnetic site can be described by a single-site model consisting of the cubic CEF Hamiltonian, the Zeeman terms for the electronic and nuclear

moments, and the hyperfine interaction:

$$\mathcal{H}_{\text{single}} = \mathcal{H}_{\text{CEF}}^{\text{cubic}} + \mu_0 (g_J \mu_B \mathbf{J} + g_N \mu_N \mathbf{I}) \cdot \mathbf{H} + A_{\text{HF}} \mathbf{I} \cdot \mathbf{J}. \quad (\text{E1})$$

The experimental procedure for estimating the residual entropy at 0 T is as follows. (1) First, the cubic CEF parameters and the hyperfine coupling constant A_{HF} in Eq. (E1) are determined from experiments performed under finite magnetic fields. As shown in Secs. IV B and IV C, these parameters can be obtained from magnetization and specific heat measurements in magnetic fields. (2) Next, isothermal magnetization measurements are performed as a function of magnetic field from $B = 0$ to B' , where $B = \mu_0 H$, at two nearby temperatures, $T = T' + \Delta T$ and $T = T' - \Delta T$. Here, T' is a fixed temperature and ΔT is a small temperature interval. B' is defined as a magnetic field such that the Hamiltonian at magnetic sites can be well described by Eq. (E1). Accordingly, B' is chosen so that the experimentally measured thermodynamic quantities quantitatively agree with those calculated from Eq. (E1) using the refined parameters. The entropy at $T = T'$ and $B = 0$ can then be estimated using Maxwell's relation $(\partial S / \partial B)_T = (\partial M / \partial T)_B$:

$$S(T', B = 0) = S(T', B') - \int_0^{B'} \left(\frac{\partial M}{\partial T} \right)_B dB, \quad (\text{E2})$$

$$\approx S(T', B') - \frac{1}{2\Delta T} \int_0^{B'} \Delta M dB, \quad (\text{E3})$$

where

$$\Delta M = M(T' + \Delta T, B) - M(T' - \Delta T, B), \quad (\text{E4})$$

and the entropy $S(T', B')$ is calculated from Eq. (E1) using the refined parameters.

As an alternative to isothermal magnetization measurements, the quantity $(\partial M / \partial T)_B$ in Eq. (E2) can also be obtained from measurements of the magnetic Grüneisen ratio Γ_H under adiabatic conditions [66] and the specific heat from $B = 0$ to B' at $T = T'$, using the thermodynamic relation $(\partial M / \partial T)_B = -C_m \Gamma_H$ [66]. Here, C_m denotes the specific heat contribution from magnetic sites. In the case of diluted Ho systems, C_m corresponds to C_{Ho} in this paper.

(3) Finally, the temperature dependence of the specific heat at $B = 0$ is measured down to the lowest accessible temperatures, and the entropy at $B = 0$ is evaluated via $S(T) = S(T') - \int_T^{T'} C_m(T)/T dT$. If $S(T)$ asymptotically approaches a finite positive value as $T \rightarrow 0$, the presence of residual entropy can be established. We note that T' is, in principle, an arbitrary finite temperature; however, in practice, it should be chosen below a few kelvin, where $C_m(T)$ can be accurately estimated.

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