

Absence of long-range order and magnetic anisotropy in the triangular magnet $\text{NdMgAl}_{11}\text{O}_{19}$

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(Received 13 February 2026; revised 28 July 2026; accepted 17 August 2026; published 2 September 2026)

We investigated the rare-earth triangular-lattice antiferromagnet $\text{NdMgAl}_{11}\text{O}_{19}$ using single-crystal magnetization ($1.8 \text{ K} \leq T \leq 300 \text{ K}$, $\mu_0 H \leq 7 \text{ T}$) and specific-heat measurements down to 45 mK. The dc susceptibility confirms a well-isolated Kramers doublet ground state with pronounced Ising-type anisotropy, with $g_c \approx 3.7$, $g_{ab} \approx 1.45$. Curie-Weiss fits yield weak, anisotropic antiferromagnetic exchange, with $\theta_c = -0.54 \text{ K}$ and $\theta_{ab} = -0.87 \text{ K}$. Heat capacity measurements show no long-range magnetic order down to 40 mK. Instead, C_m/T exhibits a broad maximum near 0.081 K whose magnitude and field evolution are consistent with short-range correlations in an anisotropic triangular lattice. Applied magnetic fields open a Zeeman gap where the specific-heat anomaly follows $\Delta = g\mu_B\mu_0 H$, and $M(H, T)$ is well described by a Brillouin function for a $J_{\text{eff}} = \frac{1}{2}$ moment. The field tuning of the low-temperature entropy manifold allows self-cooling from 1.8 K to 53 mK by adiabatic demagnetization from a $\mu_0 H = 9 \text{ T}$ field. These results identify $\text{NdMgAl}_{11}\text{O}_{19}$ as a nearly ideal weak-exchange triangular magnet with a field-tunable correlated ground state.

DOI: [10.1103/x1rf-xmrd](https://doi.org/10.1103/x1rf-xmrd)

I. INTRODUCTION

Geometrically frustrated magnets on triangular lattices often exhibit unconventional magnetic states, including quantum spin liquids (QSLs), owing to the inherent competition among spins that prevents classical long-range order [1–3]. In such systems, pronounced quantum fluctuations can lead to anomalous thermodynamic responses, persistent spin dynamics, or partially ordered phases. The suppression of magnetic order through frustration has proven technologically important with several frustrated magnets proving useful for adiabatic demagnetization refrigerant (ADR) [4,5]. Rare-earth triangular-lattice magnets are especially interesting because strong spin-orbit coupling (SOC) and crystal electric field (CEF) effects frequently yield large anisotropy and unusual correlation phenomena [6,7]. Among rare-earth triangular-lattice systems, Yb- and Tm-based materials have received extensive focus [5,8–12]. YbMgGaO₄ stirred

significant debate over potential gapless QSL behavior and the role of site disorder [8,13–15]. TmMgGaO₄ revealed essential insights into triangular-lattice quantum Ising physics and complex topological transitions [12,16–18]. Kramers ions (Nd^{3+} , Yb^{3+} , Ce^{3+}), which possess half-integer total angular momentum J , are guaranteed to host at least a doubly degenerate ground state due to time-reversal symmetry. In contrast, non-Kramers ions (Tm^{3+} , Pr^{3+}) with integer J may have nondegenerate or quasidegenerate ground states, depending sensitively on the crystal electric field environment. This distinction leads to qualitatively different low-temperature behaviors, especially in the presence of disorder or quantum fluctuations. This anisotropy often manifests in Ising-type or XY-type characters, thereby stabilizing novel partially disordered states, spin-ice-like phases, field-induced magnetic super-solid phases, or nontrivial excitations with strong quantum fluctuations [19,20]. In recent years, the rare-earth hexaaluminate family $\text{Ln}(\text{Mg/Zn})\text{Al}_{11}\text{O}_{19}$ (Ln = lanthanide) has emerged as a model platform for frustrated triangular-lattice magnetism [21–29]. Across the series, the interplay of geometric frustration, SOC, and crystal-field anisotropy stabilizes a range of unconventional low-temperature states: $\text{PrMgAl}_{11}\text{O}_{19}$ hosts a disorder-stabilized quantum Ising-like state with induced transverse field [21,22], $\text{CeMgAl}_{11}\text{O}_{19}$ lies proximate to an XXZ-type quantum critical point and exhibits a broad excitation continuum [24,25], while the magnetic response of $\text{SmMgAl}_{11}\text{O}_{19}$ is strongly quenched [27].

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However, the properties of $\text{NdMgAl}_{11}\text{O}_{19}$ remain less explored, despite Nd^{3+} being a Kramers ion that often displays notable low-temperature anisotropy and collective phenomena [30–33]. This work focuses on the novel rare-earth triangular lattice compound $\text{NdMgAl}_{11}\text{O}_{19}$. The Nd^{3+} ion has a total angular momentum of $J = 9/2$, which, in the presence of crystal-field splitting, often results in a low-lying Kramers doublet, significantly influencing magnetic anisotropy at low temperatures [29,33]. Our motivation is to explore how weak antiferromagnetic interactions, g -factor anisotropy, and geometric frustration collectively shape the unusual magnetic and thermodynamic responses observed in $\text{NdMgAl}_{11}\text{O}_{19}$, as well as highlighting its magnetocaloric refrigeration potential. We compare and contrast our single crystal study to the polycrystal study of the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29].

II. METHODS

The synthesis and subsequent crystal growth of $\text{NdMgAl}_{11}\text{O}_{19}$ were successfully carried out using a solid-state reaction and the optical floating zone (OFZ) method. Initial precursor binary oxides (Nd_2O_3 , MgO , and Al_2O_3 of 99.99% purity) were calcined at 800°C for 24 hours to remove moisture or carbonate contamination. Following calcination, the oxides were weighed in stoichiometric ratio, thoroughly mixed, and ground to ensure homogeneity. The mixture was pressed into cylindrical rods with dimensions of 6 mm in diameter and 100 mm in length. Densification was achieved using a quasihydrostatic pressure of two tons for 15 minutes. The rods were sintered in air at 1200°C for 72 hours to promote solid-state reaction and improve density. Growth was performed under an air atmosphere with a slight overpressure of 1 atm and a flow rate of 3 L/min. The sintered rods were used as both feed and seed material for crystal growth. The feed and seed rods were counterrotated at 30 rpm to improve temperature distribution and material mixing in the molten zone. The growth rate was maintained at 2 mm/h. Further details about the synthesis method are reported elsewhere [22,25]. The growth resulted in a light purple ingot containing multiple grains, with visible grain boundaries. Individual grains were separated mechanically using a wire saw and gentle cleavage, taking advantage of the strong natural cleavage parallel to the ab plane. The single-crystalline nature of the resulting pieces was confirmed using backscattered Laue x-ray diffraction. Single-crystal x-ray diffraction (SCXRD) measurements were carried out at 95 K using a Rigaku SuperNova diffractometer equipped with an Atlas S2 CCD detector and a mirror-collimated $\text{Mo K}\alpha$ radiation source ($\lambda = 0.71073 \text{ \AA}$) from a microfocus sealed tube. Data integration, scaling, and absorption correction were performed with CrysAlis Pro [34], incorporating an empirical absorption correction based on spherical harmonics [35] and an analytical numerical correction using Gaussian integration over a multifaceted crystal model via the SCALE3 ABSPACK scaling algorithm. The crystal structure was solved by the charge-flipping method using Superflip [36] and refined through full-matrix least-squares on F^2 using Jana2020 [37]. The dc magnetic susceptibility was measured using a Quantum Design SQUID Magnetic Property Measurement System (MPMS). Specific heat was

measured from room temperature down to 40 mK, using a combination of the Quantum Design Physical Property Measurement System (PPMS), the PPMS ^3He option, and an Oxford Instruments Triton dilution refrigerator. Heat capacity was determined using the relaxation method across all instruments. The small specimen (0.21 mg) measured in the dilution refrigerator was cross-calibrated against PPMS ^3He data collected on all overlapping fields over the temperature window where the two setups coincide, with a small, field-independent mass correction applied to account for the weighing uncertainty of such a small crystal. Multiple pieces of single crystals from two growths were used for the bulk measurements, with consistent results confirming the sample quality and reproducibility of response. This report shows results obtained from pieces of 8.87 and 4.39 mg. $\text{LaMgAl}_{11}\text{O}_{19}$ single crystals were also grown in air using the floating zone method, and their specific heat measurement was used to isolate the magnetic contribution (C_m) of the Nd counterpart. Crystals of $\text{LaMgAl}_{11}\text{O}_{19}$ contain a tiny amount of magnetic impurities influencing the specific heat at low temperatures; therefore, in the low-temperature limit ($T < 1 \text{ K}$), the phononic background was obtained from a T^3 extrapolation of the measured specific heat of $\text{LaMgAl}_{11}\text{O}_{19}$. The measurement of cooling potential by adiabatic demagnetization refrigeration (ADR) involved the self-cooling of the sample from 1.8 K by demagnetization from 9 T to zero applied field over a period of 30 minutes quasiadiabatically, on a pressed disk of powdered sample (1 g) intimately mixed with silver powder (1 g) for enhanced thermal conductivity, within a Quantum Design PPMS (For more information, see [4,5,38]). Electron paramagnetic resonance (EPR) spectra were acquired using a Radiopan ES/X spectrometer equipped with an Oxford Instruments helium-flow cryostat, operating over 1.5 to 300 K. Single-crystal fragments were mounted on a rotating quartz holder to probe signal anisotropy.

III. RESULTS

A. Structural characterization and point-charge CEF calculations

Our single-crystal x-ray diffraction measurements confirm that $\text{NdMgAl}_{11}\text{O}_{19}$ adopts the expected magnetoplumbite structure, consistent with previous reports on related $\text{LaMgAl}_{11}\text{O}_{19}$ compounds (La = lanthanides), seen in Fig. 1 [22,24,25,39–41]. In this structure, Nd^{3+} ions occupy 12-fold coordinated oxygen cages arranged in triangular layers, with each NdO_{12} polyhedron sharing a single in-plane oxygen vertex with its neighbors. These magnetic layers are separated by approximately 11 Å along the c axis by a nonmagnetic slab of edge- and face-sharing Al and Mg polyhedra, forming a spinel-like block. The single-crystal structure, refined at 95 K, is in excellent agreement with those of the Pr and Ce analogs and is provided in the Supplemental Material [42] (see also Ref. [43]). As with other members of this family, four sources of inherent local structural disorder are found possible: (i) a mixed Mg/Al occupancy at the Al(4)/Mg(1) site, located far from the Nd layers and having negligible magnetic impact; (ii) a positionally disordered Al(5) ion residing in a double-well

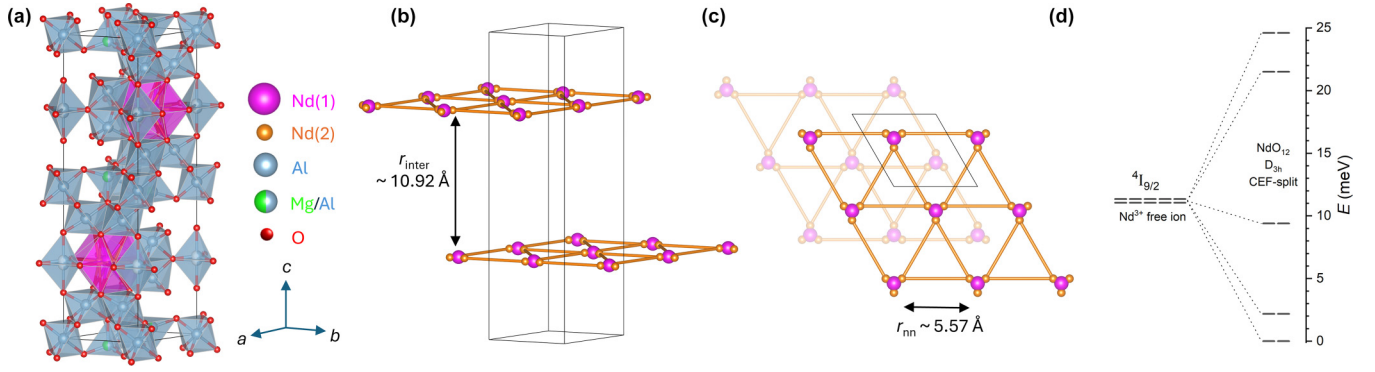


FIG. 1. The magnetoplumbite structure of $\text{NdMgAl}_{11}\text{O}_{19}$ (a), with magnetic Nd^{3+} triangular lattice interplanar (b) and intraplanar (c) separation distances. (d) A representation of the point-charge-approximation calculation of crystal electric field splitting of the $\text{Nd}^{3+} 4I_{9/2}$ free ion electronic levels, resulting in a Kramers doublet ground state, separated from the first excited doublet level by 2.174 meV (25.2 K).

potential within the bipyramidal oxygen cage, off-centered along c by $\delta \approx 0.31 \text{ \AA}$; (iii) a positional disorder of the Nd^{3+} ion within its 12-fold cage, described below; and (iv) a refined incomplete occupancy of the Nd site(s). Refinement quality was significantly improved by incorporating a second Nd site displaced by $0.83 \text{ \AA}$ from the high-symmetry $2d$ Wyckoff position, leading to refined occupancies of $0.852(4):0.0188(9)$ for $\text{Nd}_{12d} : \text{Nd}_{26h}$, respectively. This off-center site, situated on a $6h$ Wyckoff position, is supported by residual electron density maxima in Fourier difference maps and aligns with similar positional disorder reported in $\text{PrMgAl}_{11}\text{O}_{19}$ [22,44]. The final composition $\text{Nd}_{0.908}\text{MgAl}_{11}\text{O}_{19}$ suggests a $9.2(4)\%$ total Nd^{3+} deficiency, also commonly seen in several other members of this family [22,24,25,44]. While such positional disorder can significantly affect the magnetic properties of non-Kramers ions due to sensitivity to shift of gaps between low-lying singlet CEF levels, its impact in the present case is expected to be much smaller. Nd^{3+} is a Kramers ion and thus retains a degenerate doublet ground state regardless of local point symmetry. Moreover, our zero-field specific heat data show no low-lying Schottky anomalies (specific heat section), further indicating a well-isolated ground-state Kramers doublet at temperatures below 10 K. To further investigate the crystal-field scheme, we performed point-charge CEF calculations using the PyCrystalField software package [45], considering only the majority-occupied $2d\text{Nd}^{3+}$ Wyckoff site. Our point-charge calculations take into account the first coordination sphere of the Nd^{3+} ion, the NdO_{12} polyhedron, so it should be taken as an initial estimation only. However, they lead to the splitting of the tenfold $\text{Nd}^{3+} 4I_{9/2}$ levels into five pairs of Kramers degenerate doublet states. Full details of the calculated splittings are given in the Supplemental Material [42,46,47], and graphically shown in Fig. 1(d), but importantly for interpretation of the low temperature magnetic behaviors these preliminary calculations show that the Kramers doublet ground state is separated from the first excited doublet state by an energy of 2.17 meV (25.2 K). Such splittings are often seen for Nd^{3+} oxides, leading to the common description of the ground-state properties resulting from an effective $J_{\text{eff}} = 1/2$ state. The anisotropic coordination within the NdO_{12} polyhedron results in significant single ion anisotropy ($g_{x,y} = 1.35$ vs $g_z = 4.45$, $g_z/g_x = 3.31$, close to EPR results detailed in Sec. III C) perpendicular to the

triangular lattice planes. While these calculations provide an initial estimate only of the CEF scheme in $\text{NdMgAl}_{11}\text{O}_{19}$, we can compare with spectroscopic measurements of the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29]. Inelastic neutron scattering observes dispersionless excitations $\sim 3.5, 9, 23.8$, and 70 meV . The first three excitations appear close in energy to several predicted by our preliminary point-charge model, at $E_{\text{CEF}} = 2.17, 9.39, 21.50, 24.59 \text{ meV}$ [42], however the model chosen for $\text{NdZnAl}_{11}\text{O}_{19}$ assigns the observed excitations to a mixture of the Nd_{12d} and Nd_{26h} sites, using only two of the excitations for determination of their Nd_{12d} CEF scheme. Nevertheless, the conclusion of a separated, anisotropic ($g_c/g_{ab} \sim 3.3$), ground-state doublet dominating the low-temperature physics remains consistent between analyses of these isostructural analogs.

B. Magnetization

Figure 2 shows the magnetic susceptibility measured in a field of $\mu_0 H = 0.1 \text{ T}$ applied parallel to the c axis, $\chi_c(T)$, and in the ab plane, $\chi_{ab}(T)$. Down to 1.8 K no signature of long-range magnetic order appears. At sufficiently high temperatures the Curie-Weiss law is obeyed; however, deviations from linear behavior emerge below 100 K , likely due to changing occupation of the low-lying crystal-electric-field levels. Because the Nd^{3+} ion ($J = 9/2$) is split into five Kramers doublets by the CEF, the temperature dependence of thermodynamic and magnetic properties of $\text{NdMgAl}_{11}\text{O}_{19}$ are strongly influenced by this level scheme. Fitting the linear region of $\chi^{-1}(T)$ between 1.8 and 7.5 K , where we can reasonably expect significant thermal population of only the lowest lying electronic doublet ($< 7.5 \text{ K}$), yields small negative Curie-Weiss temperatures of $\theta_{\text{CW}}^c = -0.54(5) \text{ K}$ for $H \parallel c$ and $\theta_{\text{CW}}^{ab} = -0.86(2) \text{ K}$ for $H \parallel ab$, indicating weak antiferromagnetic interactions. The ratio $\chi_c/\chi_{ab} \approx 10$ at 2 K reflects a strong single-ion uniaxial anisotropy, consistent with the CEF calculations, and with a predominantly c -axis moment, similar to that seen in $\text{CeMgAl}_{11}\text{O}_{19}$ [25]. The effective moment extracted from the Curie-Weiss fit is $\mu_{\text{eff}} \approx 3.42 \mu_B$, and $\mu_{\text{eff}} \approx 1.33 \mu_B$ corresponding to an effective g factor of $g_{\text{eff}} \approx 3.95$ and $g_{\text{eff}} \approx 1.54$, respectively, for $H \parallel c$ and $H \parallel ab$ close to that estimated by our point-charge calculations and EPR measurements. Figure 3 shows the isothermal

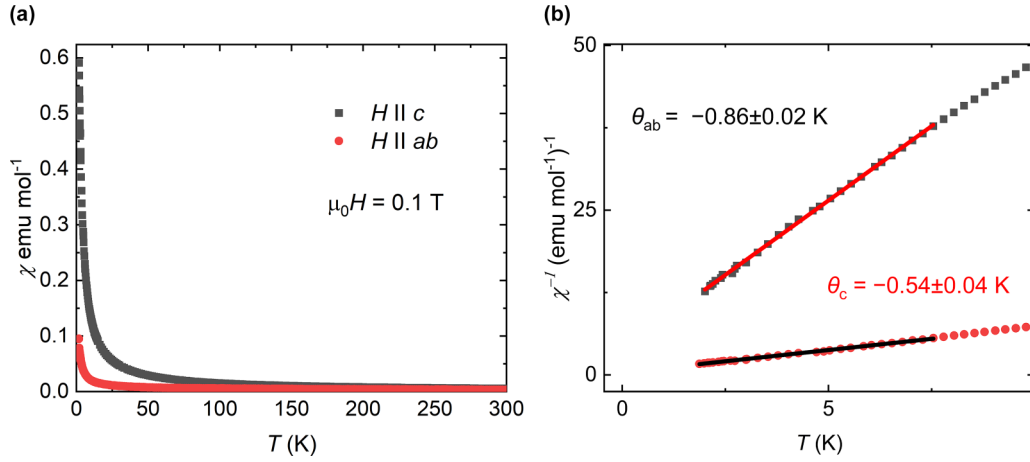


FIG. 2. Magnetic susceptibility measured with an external magnetic field $\mu_0 H = 0.1$ T, applied parallel to the c axis, and perpendicular to the c axis (a). Corresponding inverse susceptibility with CW fits at low temperatures. The solid red lines represent the Curie-Weiss fitting of the inverse susceptibility, described in the text.

magnetization $M(H)$ for both field orientations. The data up to 10 K are well described by a Brillouin function for a $J_{\text{eff}} = \frac{1}{2}$ doublet, supplemented by a small temperature-independent background term,

$$M(H, T) = M_s B_J(x) + a\mu_0 H, \quad (1)$$

where $M_s = gJ\mu_B$ and

$$B_J(x) = \frac{2J+1}{2J} \coth\left(\frac{2J+1}{2J}x\right) - \frac{1}{2J} \coth\left(\frac{x}{2J}\right), \quad (2)$$

with

$$x = \frac{gJ\mu_B\mu_0 H}{k_B T}. \quad (3)$$

A global fit to the $T = 2, 5$, and 10 K datasets (within the regime where only the ground-state doublet is significantly thermally populated) yields $g = 3.75(1)$ for $H \parallel c$, with negligible a , confirming that the magnetization is dominated by the Kramers doublet ground state. For $H \parallel ab$, the magnetization reaches $\sim 0.75 \mu_B/\text{Nd}$ at $\mu_0 H = 7$ T without full saturation; the corresponding global fit gives $g = 1.54(1)$ and an expected

saturation moment of $\approx 0.77 \mu_B/\text{Nd}$. The excellent agreement between the experimental data and the Brillouin model in both field orientations demonstrates that, for $10 \text{ K} \gtrsim T \gtrsim 2 \text{ K}$, the magnetization is governed by weakly interacting $J_{\text{eff}} = \frac{1}{2}$ moments, with exchange interactions becoming relevant only at lower temperatures. The breakdown of the single-doublet Brillouin fit at $T \gtrsim 15$ K is consistent with thermal population of the first excited Kramers doublet, predicted at ~ 25 K (2.17 meV) by our point-charge CEF calculation and observed near 3.5 meV by inelastic neutron scattering in the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29], as well as with the gradual deviation from Curie-Weiss behavior observed in $\chi^{-1}(T)$ above ~ 10 K.

C. Electron paramagnetic resonance

To probe the single-ion anisotropy directly and complement the thermodynamic characterization of the ground-state Kramers doublet, we performed continuous-wave X-band electron paramagnetic resonance (EPR) measurements at $\nu = 9.4$ GHz on a single crystal of $\text{NdMgAl}_{11}\text{O}_{19}$ at 1.5 K. The

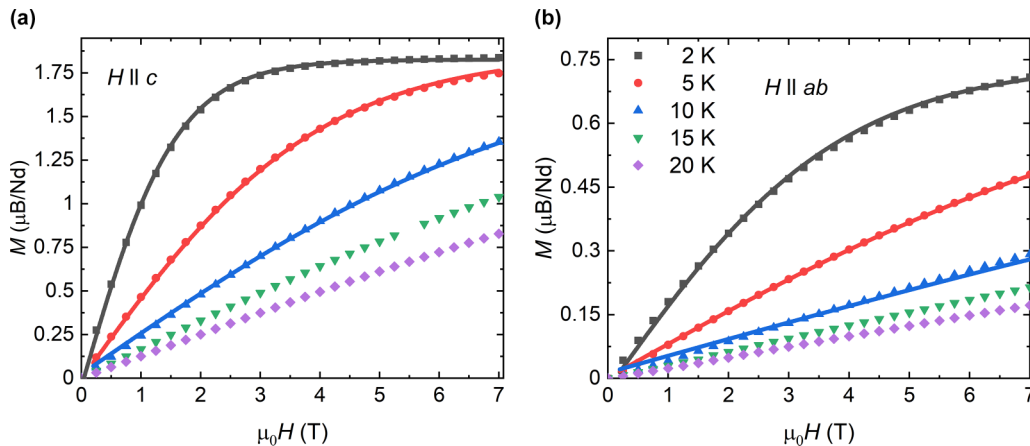


FIG. 3. Isothermal magnetization as a function of applied magnetic field for (a) $\mu_0 H \parallel c$ and (b) $\mu_0 H \parallel ab$. Data are shown for $T = 2, 5, 10, 15$, and 20 K. Solid lines represent global Brillouin fits for a $J_{\text{eff}} = \frac{1}{2}$ doublet to the $T = 2, 5$, and 10 K datasets.

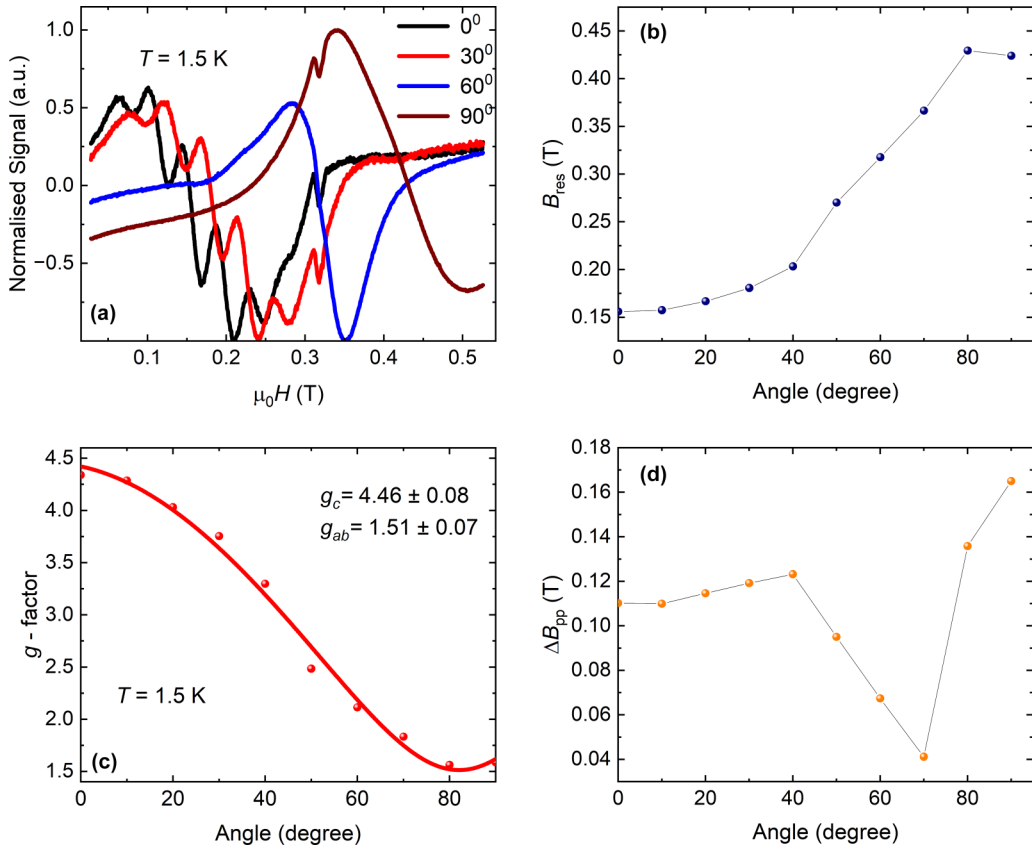


FIG. 4. Angle-dependent X-band EPR characterization of $\text{NdMgAl}_{11}\text{O}_{19}$ at 1.5 K ($\nu = 9.4$ GHz). (a) Normalized first-derivative spectra dI/dB vs $\mu_0 H$ for representative rotation angles $\theta = 0^\circ$, 30° , 60° , and 90° . The multihump fine structure resolved at $\theta = 0^\circ$ ($H \parallel c$) progressively weakens upon rotation toward the hard plane ($H \parallel ab$). The weak feature near 314 mT is an extrinsic carbon-cavity resonance and is excluded from all analysis. (b) Resonance field B_{res} versus θ , obtained from the extrema-midpoint construction. (c) Effective g factor $g(\theta)$; the solid red curve is a fit to the uniaxial anisotropy model, yielding $g_c = 4.46(8)$ and $g_{ab} = 1.51(7)$. (d) Peak-to-peak linewidth $\Delta B_{\text{pp}}(\theta)$, showing maximum broadening along the easy axis attributed to unresolved hyperfine structure and CEF mixing, with narrowing at intermediate angles.

crystal was mounted on a rotating quartz holder such that the rotation angle θ varied the orientation of the applied field continuously between $\theta = 0^\circ$ ($H \parallel c$) and $\theta = 90^\circ$ ($H \parallel ab$). Spectra were recorded in first-derivative mode dI/dB using lock-in detection, with a linear background removed by least-squares detrending [Fig. 4(a)]. A weak extrinsic resonance visible at $\mu_0 H \approx 314$ mT originates from a carbon-related impurity in the microwave cavity and was excluded from the analysis. For each angle, the resonance field B_{res} and peak-to-peak linewidth ΔB_{pp} were extracted using the standard extrema-midpoint construction [48,49],

$$B_{\text{res}} = \frac{B_{\text{max}} + B_{\text{min}}}{2}, \quad \Delta B_{\text{pp}} = B_{\text{min}} - B_{\text{max}}, \quad (4)$$

where B_{max} and B_{min} are the field positions of the positive and negative derivative extrema. The effective g factor follows from the resonance condition $g = h\nu/(\mu_B B_{\text{res}})$. The angular variation of B_{res} [Fig. 4(b)] and the derived $g(\theta)$ [Fig. 4(c)] evolve smoothly between the two principal orientations and are well captured by the standard uniaxial model

$$g(\theta) = \sqrt{g_c^2 \cos^2(\theta - \theta_0) + g_{ab}^2 \sin^2(\theta - \theta_0)}, \quad (5)$$

yielding $g_c = 4.46(8)$, $g_{ab} = 1.51(7)$, and a small misalignment offset $\theta_0 = -7.92(16)^\circ$ between the crystal c axis and the goniometer rotation axis. The strongly anisotropic, easy-axis g tensor with $g_c/g_{ab} \approx 3.0$ is characteristic of Nd^{3+} in trigonal coordination and is in quantitative agreement both with our point-charge CEF calculation ($g_{zz} = 4.45$, $g_{xx} = g_{yy} = 1.35$) and with the values reported for the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29]. The Zeeman splitting $\Delta(H)$ extracted independently from the field-dependent specific heat (below) returns $g_c = 3.86(5)$, in good agreement with the EPR value and confirming a fully consistent picture of the single-ion anisotropy across all probes. The peak-to-peak linewidth $\Delta B_{\text{pp}}(\theta)$ [Fig. 4(d)] is largest near the easy axis, narrows substantially upon rotation, and reaches a minimum near $\theta \approx 60^\circ$ – 70° before broadening again at $\theta = 90^\circ$. This nonmonotonic behavior reflects the angular dependence of the spin-lattice and hyperfine contributions discussed below, both of which are most effective when the field is aligned with the crystal-field axis and progressively average out upon tilting. A prominent multihump fine structure is clearly resolved in the $\theta = 0^\circ$ spectrum, weakens systematically upon rotation, and is no longer discernible above approximately 60° , where the spectrum coalesces into a single broad line. We attribute this fine structure to two concurring mechanisms. First, spin-

orbit-induced admixture of the two lowest Kramers doublets of the $4f^3$ ($J = 9/2$) multiplet under a field along the trigonal symmetry axis, which generates closely spaced resonances with slightly different effective g_c values [49,50]. Second, hyperfine coupling to the magnetic Nd isotopes ^{143}Nd and ^{145}Nd (both $I = 7/2$, natural abundances 12.2% and 8.3%, respectively), each of which produces a $2I + 1 = 8$ -line multiplet governed by the hyperfine term

$$\mathcal{H}_{\text{hf}} = A_{\parallel} S_z I_z + A_{\perp} (S_x I_x + S_y I_y), \quad (6)$$

with $A_{\parallel} \gg A_{\perp}$ in the strongly axial limit relevant here. The adjacent-peak separations in the $\theta = 0^\circ$ spectrum are $\Delta B \approx 40\text{--}45$ mT, yielding via $\Delta B \approx A_{\parallel}/(g_c \mu_B)$ an axial hyperfine coupling $A_{\parallel} \approx 0.09$ cm $^{-1}$ (≈ 2.7 GHz), consistent with values reported for Nd^{3+} in oxide hosts [50]. The multiplet remains incompletely resolved owing to substantial inhomogeneous broadening: The $\sim 9.2\%$ Nd-site deficiency together with the positional disorder between the majority Nd(1) $2d$ and minority Nd(2) $6h$ Wyckoff positions introduces a distribution of local crystal-field environments at the Nd sites, smearing out the individual hyperfine lines. As the field rotates away from the c axis, both the CEF-mixing-induced splitting and the projection of the axial hyperfine tensor onto the field direction decrease, naturally explaining the progressive suppression of the fine structure and the convergence to a single broad line in the hard plane. In summary, the angle-dependent EPR data quantitatively confirm the strongly anisotropic easy-axis g tensor of $\text{NdMgAl}_{11}\text{O}_{19}$, $g_c = 4.46(8)$ and $g_{ab} = 1.51(7)$, in agreement with the point-charge CEF analysis and with the g_c value extracted from the field-dependent specific heat. The fine structure along the easy axis arises from a superposition of CEF mixing within the Kramers manifold and unresolved hyperfine coupling to the ^{143}Nd and ^{145}Nd nuclei, with $A_{\parallel} \approx 0.09$ cm $^{-1}$ (≈ 2.7 GHz) inferred from the observed peak separations, broadened by the intrinsic structural disorder of this magnetoplumbite family. A quantitative separation of these contributions would require multifrequency high-field EPR or measurements on isotopically enriched samples and is left for future work.

D. Specific heat

The specific heat $C(T)$ of $\text{NdMgAl}_{11}\text{O}_{19}$ was measured down to 40 mK. No sharp λ -type anomaly is observed, excluding a conventional long-range ordering transition down to the base temperature [Fig. 5(a)]. In zero field, $C(T)$ exhibits a broad hump with a maximum at $T^* = 81(2)$ mK, corresponding to the onset of short-range magnetic correlations. To isolate the magnetic contribution, we subtract the phonon background using the nonmagnetic analog $\text{LaMgAl}_{11}\text{O}_{19}$, defining $C_m = C_{\text{tot}} - C_p$ (no electronic term is expected for this insulating oxide). The resulting $C_m(T)$ shows a broad, nondivergent maximum ($C_m^{\text{max}} \sim 1.6$ J mol $^{-1}$ K $^{-1}$) and then decreases approximately as T^{-1} up to ~ 0.3 K, with no additional anomalies between 0.3 and 5 K. Integration of C_m/T reveals that the entropy release is strongly incomplete in zero field: By 15 K we recover $S_m \approx 2.59$ J mol $^{-1}$ K $^{-1} \approx 0.45 R \ln 2$, leaving a missing entropy of $R \ln 2 - S_m \approx 3.17$ J mol $^{-1}$ K $^{-1} \approx 0.55 R \ln 2$ within our experimental window down to 40 mK [Fig. 5(d)]. This large entropy deficit,

together with the absence of any transition feature, points to persistent low-energy correlations and fluctuating degrees of freedom that retain a substantial fraction of the ground-state entropy below the base temperature, consistent with the absence of ordering or spin-freezing anomalies down to 50 mK in χ_{AC} investigations of the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29]. To probe the field evolution of the low-energy spectrum, we measured $C(T)$ in magnetic fields up to 3 T with $H \parallel c$ and extracted $C_m(T)$ [Fig. 5(b)]. With increasing field, the broad low- T anomaly shifts to higher temperature and becomes more Schottky-like, consistent with Zeeman splitting of the ground-state Kramers doublet and progressive suppression of correlation effects as the response becomes increasingly single-ion dominated [13,14,22,51]. In this regime we fit $C_m(T)$ using a two-level Schottky model,

$$C_m(T) = \eta R \left(\frac{\Delta}{T} \right)^2 \frac{e^{-\Delta/T}}{(1 + e^{-\Delta/T})^2}, \quad (7)$$

where $\Delta \equiv \Delta_E/k_B$ is the Zeeman splitting in kelvin and η quantifies the fraction of Nd^{3+} ions contributing to the anomaly. This model describes the data well for fields 0.75 – 3 T [solid curves in Fig. 5(b)], while for $\mu_0 H \lesssim 1$ T the fitted η drops sharply and the extracted gap becomes ill-defined, signaling that weak fields do not fully quench the dominant correlations responsible for the zero-field anomaly, which remains discernible below 0.1 K in the $\mu_0 H = 0\text{--}0.5$ T data. The same crossover is visible in the dielectric response of this compound, which carries clear crystal-field signatures: Field sweeps at 0.3 and 2 K show a well-defined field-induced crossover at $\mu_0 H \approx 0.8$ T separating a low-field correlated regime from a high-field single-ion one [52]. From the reliable Schottky fits we obtain $\Delta(H)$ and find a strictly linear field dependence [inset of Fig. 5(c)]. Fitting $\Delta(H) = \Delta_0 + (g_{\text{eff}} \mu_B/k_B) \mu_0 H$ yields $g_{\text{eff}} = 3.92(1)$ with Δ_0 consistent with zero, in good agreement with the single-ion anisotropy obtained independently from Curie-Weiss analysis, Brillouin-function fits, point-charge CEF estimates, and EPR. The fitted $\eta \approx 0.81\text{--}0.87$ between 1 and 3 T indicates a modest reduction of the effective Nd^{3+} spectral weight, consistent with the small Nd-site deficiency established by SCXRD. Turning to the entropy [Fig. 5(d)], at 3 T the uncorrected S_m reaches ≈ 4.98 J mol $^{-1}$ K $^{-1}$ by 15 K, falling short of $R \ln 2$ by ~ 0.78 J mol $^{-1}$ K $^{-1}$. This deficit matches quantitatively the spectral-weight loss expected from $\eta < 1$; after correcting for it (gray shaded region), the 3 T entropy saturates at $R \ln 2$ by 15 K, confirming a $J_{\text{eff}} = \frac{1}{2}$ Kramers doublet ground state. The residual entropy shortfall that persists in zero field beyond this Nd-deficiency contribution (magenta shaded region) therefore reflects genuine entropy retained below 40 mK, consistent with persistent correlated fluctuations rather than symmetry breaking. Above ~ 15 K, C_m rises again and S_m begins to exceed $R \ln 2$ in both field orientations, signaling the onset of thermal population of the first excited Kramers doublet at $E_{\text{CEF}} = 2.17$ meV (25.2 K) predicted by our point-charge calculations. The resulting Schottky-like upturn in C_m and the accompanying excess entropy are calorimetric signatures of the CEF level scheme, and coincide precisely with the temperatures at which the single-doublet Brillouin description of $M(H)$ begins to break down, providing a fully

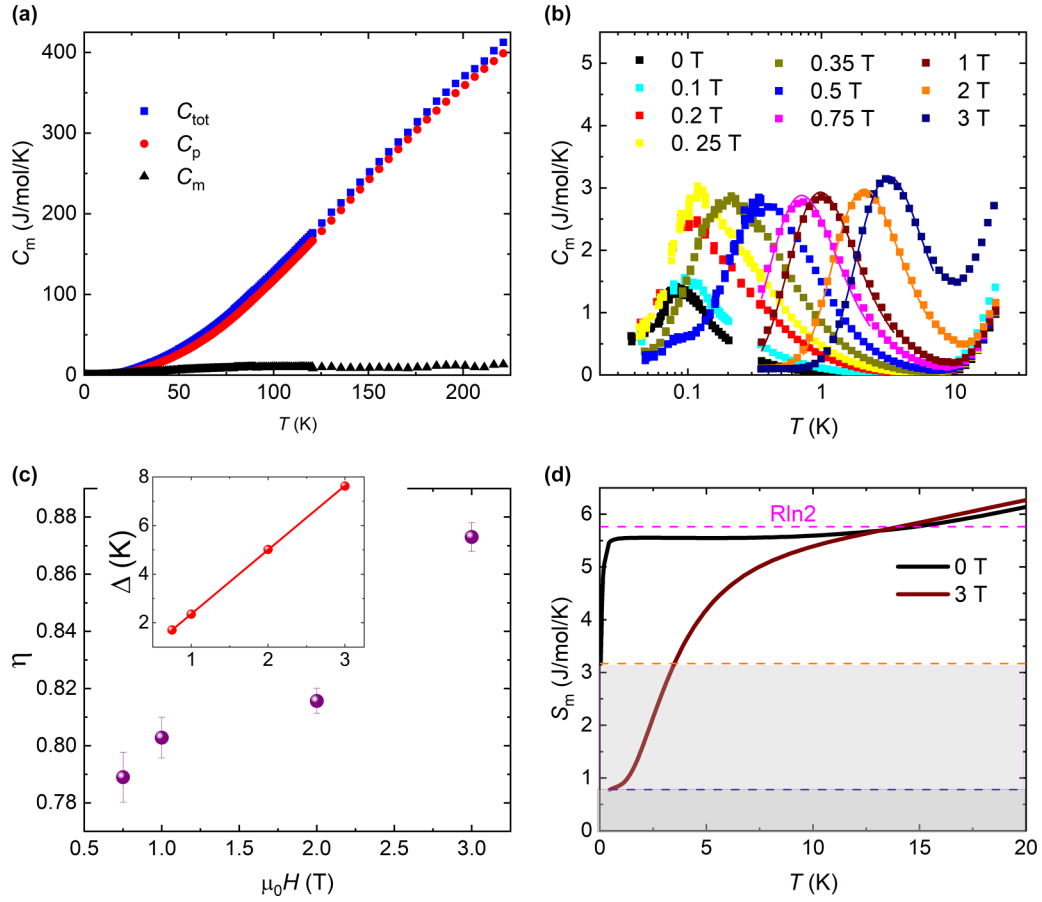


FIG. 5. (a) Total specific heat (C_{tot}), nonmagnetic (phonon) contribution (C_p), and the resulting magnetic specific heat (C_m) obtained by subtraction, shown as functions of temperature in zero magnetic field. (b) Magnetic specific heat $C_m(T)$ measured in various magnetic fields. Solid lines show fits to the two-level Schottky model, while dashed lines indicate extrapolated curves inferred from the Schottky analysis. (c) Field dependence of η , the fraction of ions contributing to the Schottky anomaly; inset: extracted Schottky gap Δ as a function of magnetic field. (d) Magnetic entropy $S_m(T)$. The semitransparent gray shaded area below the dashed magenta line highlights the entropy deficit attributed to Nd deficiency, while the transparent gray shaded area between the dashed orange and dashed magenta lines marks the residual entropy remaining below 40 mK.

self-consistent picture of the CEF excitation structure across both magnetic and thermodynamic probes.

E. Adiabatic demagnetization refrigerant potential

To directly assess the suitability of $\text{NdMgAl}_{11}\text{O}_{19}$ for ADR, we performed a self-cooling (demagnetization) experiment in a Quantum Design PPMS using a pressed disk of powdered sample. After stabilizing the sample at $T_i = 1.8$ K in an applied field of $\mu_0 H = 9$ T, the residual helium gas used for thermalization was absorbed in a custom-built *in situ* cryopump. Under these quasiadiabatic conditions, the field was ramped continuously down to zero over 30 minutes, corresponding to an average sweep rate $|d(\mu_0 H)/dt| \approx 0.3 \text{ T min}^{-1}$. As shown in Fig. 6, the sample temperature decreased significantly during the field ramp, reaching a minimum temperature of $T_f = 52$ mK at $\mu_0 H = 0$ T. This corresponds to a self-cooling factor of $T_i/T_f \approx 35$, demonstrating that substantial magnetic entropy remains available for refrigeration well below 1 K. The pronounced cooling is consistent with the low-temperature heat-capacity anomaly and the incomplete release of $R \ln 2$ entropy in zero field,

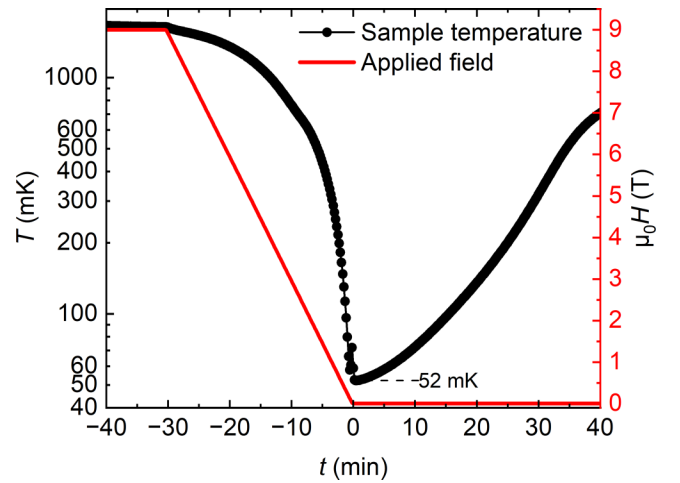


FIG. 6. Self-cooling curve for a pressed disk of $\text{NdMgAl}_{11}\text{O}_{19}$ powder during adiabatic demagnetization from $\mu_0 H = 9$ T at a start temperature of 1.8 K.

indicating a dense manifold of low-energy magnetic degrees of freedom that can be efficiently polarized at high field and exploited for ADR upon demagnetization. The nonideal adiabaticity of the setup then results in a slow-warming curve as heat is leaked back to the sample from the measurement puck's thermal bath.

IV. DISCUSSION

The combination of SCXRD, magnetization, and specific-heat measurements on $\text{NdMgAl}_{11}\text{O}_{19}$ together with point-charge CEF calculations, reveals a triangular-lattice rare-earth magnet in which geometric frustration, weak exchange interactions, and strong single-ion anisotropy cooperatively suppress conventional magnetic order down to at least 40 mK. Magnetization data exhibit pronounced anisotropy, and low-temperature Curie-Weiss fits (valid when only the ground-state Kramers doublet is populated) yield $\theta_{\text{CW}}^{ab} \approx -0.87$ K and $\theta_{\text{CW}}^c \approx -0.54$ K, indicating weak antiferromagnetic coupling that is stronger in the ab plane. Brillouin-function fits reproduce the field-dependent magnetization with high fidelity for both orientations, confirming that the system behaves as a collection of well-defined, only weakly interacting Kramers pseudospins [13,51,53]. Despite its paramagnetic behavior above 2 K, the specific heat displays a broad anomaly centered at 81 mK, signaling the development of short-range correlations among Nd^{3+} moments. Crucially, no sharp λ -type feature or other indication of long-range magnetic order appears down to 40 mK, implying that any ordering transition lies below our experimental base temperature or is entirely suppressed by frustration and anisotropy. While our ZFC-FC susceptibility measurements are restricted to $T \geq 1.8$ K, far above any plausible freezing scale in this compound, several observations argue against spin freezing: The broad specific-heat anomaly evolves smoothly and Schottky-consistently with applied field, in contrast to the weak field dependence typical of glassy freezing, and ac susceptibility (down to 50 mK) and μSR (down to 0.5 K) measurements on the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ reveal no spin freezing [29]. Using the direction-dependent Weiss temperatures and noting that no ordering occurs down to $T_N < 40$ mK, we obtain frustration indices $f_{ab} = |\theta_{\text{CW}}^{ab}|/T_N > 0.87/0.04 \approx 22$ and $f_c = |\theta_{\text{CW}}^c|/T_N > 0.54/0.04 \approx 13$, both above the value of 10 usually needed to assign a system as highly frustrated. These values should be regarded as approximate: The Curie-Weiss temperatures carry the usual mean-field uncertainties, while the anomaly in zero-field specific heat appears near 81 mK and a field of 0.7–1 T is required to suppress the correlated regime, suggesting the true interaction scale lies between these bounds. Since our measurements extend only down to 40 mK, ordering at lower temperatures cannot be excluded, and the calculated indices should therefore be considered lower bounds. In zero applied field, only a portion of the $J_{\text{eff}} = \frac{1}{2}$ entropy is released below 1 K, indicating that a substantial fraction of the ground-state entropy remains locked into low-energy degrees of freedom. This is consistent with persistent spin-dynamics observed in the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ using μSR down to 0.5 K [29]. Between 1 and 6 K the entropy shows no further evolution, consistent with a frustration-stabilized manifold without any intervening

ordering or freezing transition. Only at higher temperatures does the entropy begin to increase again due to thermal population of the first excited crystal-field doublet. When a 3 T magnetic field is applied along the c axis, a Zeeman gap of $\Delta \approx 0.67$ meV opens, producing a Schottky anomaly near 4 K. This field lifts the ground-state degeneracy and restores nearly the full $R \ln 2$ entropy by 15 K, with additional entropy released compared to that measured in zero field. The field-induced recovery of entropy confirms that the “missing” entropy at zero field resides in a macroscopically degenerate, frustration-driven manifold rather than in inaccessible CEF levels, in close analogy to observations in $\text{Ba}_3\text{Yb}(\text{BO}_3)_3$ [54]. The 9.2(4)% Nd-site vacancies ($p = 0.908$) will further effect the magnetic properties. Since the occupation level is well above the two-dimensional site percolation threshold ($p_c = 0.5$), the triangular lattice remains fully connected. In the hypothetical undiluted limit ($p = 1$), $|\theta_{\text{CW}}|$ would increase by approximately 10%, indicating only a minor renormalization effect due to the dilution that we observe from diffraction. Classical Monte Carlo simulations of the site-diluted triangular-lattice Ising antiferromagnet have long shown that nonmagnetic vacancies liberate nearly free $S = \frac{1}{2}$ “orphan” spins: Early studies by Grest and Gabl demonstrated defect-induced spin-glass freezing in this model [55], and Creswick identified a corresponding depletion of zero-net-field sites in the local-field distribution [56]. Building on these, Moessner and Berlinsky quantified that each Henley-type defect triad (three isolated vacancies) produces one orphan moment, leading to the rule of thumb of roughly one orphan per three vacancies [57]. A simple geometric estimate then gives

$$n_{\text{orphan}} \approx \frac{1}{3}(1 - p) = 0.031 \quad \text{per Nd site,}$$

and the corresponding maximum entropy is

$$S_{\text{orphan}} = n_{\text{orphan}} R \ln 2 \approx 0.18 \text{ J mol}^{-1} \text{ K}^{-1},$$

which remains far below the missing $R \ln 2$ entropy we observe up to 15 K in zero field. Moreover, in a quantum Kramers-ion system, zero-point fluctuations and transverse exchange will further suppress the orphan contribution, underscoring that degenerate correlated states resulting from the geometric frustration, not dilution-induced orphan spins, dominantly retains the residual entropy. The pronounced self-cooling observed under quasiadiabatic demagnetization (Fig. 6) places $\text{NdMgAl}_{11}\text{O}_{19}$ within a broader class of low-temperature magnetocaloric materials in which frustration and/or disorder suppress long-range order and retain a sizable entropy reservoir to deeply subkelvin temperatures [5,58]. A key limitation of the present compound, however, is its comparatively low rare-earth moment density: Even if the full Kramers-doublet entropy $R \ln 2$ per Nd is available, the maximum entropy density is

$$s_{\text{max}} = \frac{ZR \ln 2}{N_A V_{\text{cell}}} \simeq 32.5 \text{ mJ cm}^{-3} \text{ K}^{-1},$$

using $Z = 2$ and the refined hexagonal unit cell. This is comparable to historic hydrated-salt refrigerants [4], but remains modest compared with dense Gd^{3+} -based refrigerants, where the spin-only $R \ln(8)$ entropy per ion combined with a high concentration of magnetic ions yields substantially larger volumetric entropy changes and cooling power; recent

frustrated Gd-rich oxides in particular demonstrate state-of-the-art low-temperature performance [59,60], alongside established benchmarks such as rare-earth garnets [61] and other chemically robust Gd-based lattices [62,63]. Nevertheless, the strong magnetic anisotropy of $\text{NdMgAl}_{11}\text{O}_{19}$, coupled with the availability of reasonably sized single crystals, makes this system a promising candidate for exploring rotational magnetocaloric effects. A definitive determination of the true ground state will require local probes. Inelastic neutron scattering can map out the full spin-excitation spectrum and provide a direct determination of the exchange tensor; μSR or NMR can resolve slow dynamics and possible critical slowing down. Only a combination of such local-probe measurements and theory constrained by the experimentally determined exchange parameters can unambiguously distinguish between quantum-correlated or quantum-disordered scenarios, such as quantum spin liquid, valence bond solid, or random singlet ground states. In the discussion, we considered the possible effects of Nd deficiency on the magnetic and thermodynamic response; however, for clarity and simplicity, heat capacity and magnetic property data are plotted and analyzed per mol formula unit, i.e., neglecting the Nd deficiency.

V. CONCLUSION

Our combined SCXRD, magnetization, specific-heat measurements on single-crystalline $\text{NdMgAl}_{11}\text{O}_{19}$ supported by point-charge calculation reveal a triangular-lattice system composed of well-isolated Kramers doublets with strong axial anisotropy. The extracted $\theta_{\text{CW}} \approx -0.54$ K indicates the presence of antiferromagnetic interactions, yet no evidence of long-range magnetic order or spin-glass freezing is observed down to our base temperature of 40 mK. This robust suppression of ordering, despite magnetic exchange and measurable structural disorder, points to strong geometric frustration amplified by the system's quasi-two-dimensionality and $\sim 9.2\%$ Nd-site dilution. The thermal anomaly centered at ~ 81 mK, combined with the large residual entropy and its full recovery under applied field, further confirms the presence of a frustrated manifold that allows self-cooling down to 52 mK on demagnetization from a field of $\mu_0 H = 9$ T. The proximity to a quantum-disordered state with persistent spin dynamics cannot be excluded, as has been suggested in the isostructural analog $\text{NdZnAl}_{11}\text{O}_{19}$ [29]. In this context, $\text{NdMgAl}_{11}\text{O}_{19}$ emerges as a promising candidate for a triangular-lattice quantum spin liquid, readily grown as a single crystal. Further experimental investigations, such as low-temperature inelastic

neutron scattering and muon spin relaxation will be essential to clarify the nature of its ground state.

ACKNOWLEDGMENTS

We acknowledge funding from Charles University in Prague within the Primus research program with Grant No. PRIMUS/22/SCI/016, and the Grant Agency of Univerzita Karlova (Grant No. 438425). The work was also supported by the Ministry of Education, Youth and Sports of the Czech Republic through program INTER-EXCELLENCE II INTER-ACTION (LUABA24056) and the Czech Science Foundation (Project No. 26-23051S). Work at the University of Augsburg was supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Grant No. 514162746 (GE 1640/11-1) and the Bavarian-Czech Academic Agency BTHA, Grant No. JC-2024-15 (BaC-QuERel). Crystal growth, structural analysis, and magnetic properties measurements were carried out in the MGML [64], supported within the Czech Research Infrastructures program (Project No. LM2023065). The first author (S.K.) would like to acknowledge also the TERA FIT project – CZ.02.01.01/00/22_008/0004594, cofounded by the European Union (EU) and the Czech Ministry of Education, Youth and Sports (MEYS).

S. Kumar prepared the samples, verified crystal quality, reproduced the magnetization and specific-heat datasets, performed point-charge crystal-field calculations, carried out data analysis, and drafted the manuscript. G. Bastien initiated the project, performed the initial magnetization and specific-heat measurements, and supervised parts of the analysis. J. Prokleška performed dilution refrigerator measurements of specific heat. M. Kempirski and W. Kempirski conducted the EPR measurements. K. Załeski reproduced magnetization measurements on additional single crystals. A. Kancko and C. Correa performed the single-crystal x-ray diffraction measurements and structural analysis. T. Treu and P. Gegenwart carried out the ADR performance tests. M. Śliwińska-Bartkowiak supervised and coordinated all experimental work performed in Poznań. R. H. Colman supervised the crystal growth, data analysis, and manuscript preparation.

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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- [1] A. P. Ramirez and S. V. Syzranov, Short-range order and hidden energy scale in geometrically frustrated magnets, *Mater. Adv.* **6**, 1213 (2025).
 - [2] L. Balents, Spin liquids in frustrated magnets, *Nature (London)* **464**, 199 (2010).
 - [3] L. Savary and L. Balents, Quantum spin liquids: A review, *Rep. Prog. Phys.* **80**, 016502 (2017).
 - [4] Y. Tokiwa, S. Bachus, K. Kavita, A. Jesche, A. A. Tsirlin, and P. Gegenwart, Frustrated magnet for adiabatic demagnetization cooling to milli-Kelvin temperatures, *Commun. Mater.* **2**, 42 (2021).
 - [5] T. Treu, M. Klinger, N. Oefele, P. Telang, A. Jesche, and P. Gegenwart, Utilizing frustration in Gd- and Yb-based oxides for milli-Kelvin adiabatic demagnetization refrigeration, *J. Phys.: Condens. Matter* **37**, 013001 (2025).
 - [6] Y.-Y. Qin, Y. Shen, G. Chen, and J. Zhao, Magnetic frustration and quantum fluctuation in rare-earth triangular-lattice magnets, *Physics* **50**, 454 (2021).

- [7] M. Xie, W. Zhuo, Y. Cai, Z. Zhang, and Q. Zhang, Rare-earth chalcogenides: An inspiring playground for exploring frustrated magnetism, *Chin. Phys. Lett.* **41**, 117505 (2024).
- [8] Y. Li, H. Liao, Z. Zhang, S. Li, F. Jin, L. Ling, L. Zhang, Y. Zou, L. Pi, *et al.*, Gapless quantum spin liquid ground state in the two-dimensional spin-1/2 triangular antiferromagnet YbMgGaO_4 , *Sci. Rep.* **5**, 16419 (2015).
- [9] U. K. Voma, S. Bhattacharya, E. Kermarrec, J. Alam, Y. M. Jana, B. Sana, P. Khuntia, S. K. Panda, and B. Koteswararao, Electronic structure and magnetic properties of the effective spin- $J_{\text{eff}} = \frac{1}{2}$ two-dimensional triangular lattice $\text{K}_3\text{Yb}(\text{VO}_4)_2$, *Phys. Rev. B* **104**, 144411 (2021).
- [10] K. Somesh, S. S. Islam, S. Mohanty, G. Simutis, Z. Guguchia, C. Wang, J. Sichelschmidt, M. Baenitz, and R. Nath, Absence of magnetic order and emergence of unconventional fluctuations in the $J_{\text{eff}} = \frac{1}{2}$ triangular-lattice antiferromagnet YbBO_3 , *Phys. Rev. B* **107**, 064421 (2023).
- [11] J. Khatua, S. Bhattacharya, A. M. Strydom, A. Zorko, J. S. Lord, A. Ozarowski, E. Kermarrec, and P. Khuntia, Magnetic properties and spin dynamics in the spin-orbit driven $J_{\text{eff}} = \frac{1}{2}$ triangular lattice antiferromagnet $\text{Ba}_6\text{Yb}_2\text{Ti}_4\text{O}_{17}$, *Phys. Rev. B* **109**, 024427 (2024).
- [12] Y. Shen, C. Liu, Y. Qin, S. Shen, Y.-D. Li, R. Bewley, A. Schneidewind, G. Chen, and J. Zhao, Intertwined dipolar and multipolar order in the triangular-lattice magnet TmMgGaO_4 , *Nat. Commun.* **10**, 4530 (2019).
- [13] X. Rao, G. Hussain, Q. Huang, W. J. Chu, N. Li, X. Zhao, Z. Dun, E. S. Choi, T. Asaba, *et al.*, Survival of itinerant excitations and quantum spin state transitions in YbMgGaO_4 with chemical disorder, *Nat. Commun.* **12**, 4949 (2021).
- [14] I. Kimchi, A. Nahum, and T. Senthil, Valence bonds in random quantum magnets: Theory and application to YbMgGaO_4 , *Phys. Rev. X* **8**, 031028 (2018).
- [15] Y. Li, D. Adroja, R. I. Bewley, D. Voneshen, A. A. Tsirlin, P. Gegenwart, and Q. Zhang, Crystalline electric-field randomness in the triangular lattice spin-liquid YbMgGaO_4 , *Phys. Rev. Lett.* **118**, 107202 (2017).
- [16] H. Li, Y. D. Liao, B.-B. Chen, X.-T. Zeng, X.-L. Sheng, Y. Qi, Z. Y. Meng, and W. Li, Kosterlitz–Thouless melting of magnetic order in the triangular quantum Ising material TmMgGaO_4 , *Nat. Commun.* **11**, 1111 (2020).
- [17] C.-J. Huang, X. Wang, Z. Wang, and G. Chen, Emergent Halperin–Saslow mode, gauge glass and quenched disorders in quantum Ising magnet TmMgGaO_4 , *Int. J. Mod. Phys. B* **38**, 2450040 (2024).
- [18] C. Liu, C.-J. Huang, and G. Chen, Intrinsic quantum Ising model on a triangular lattice magnet TmMgGaO_4 , *Phys. Rev. Res.* **2**, 043013 (2020).
- [19] S. T. Bramwell and M. J. Harris, The history of spin ice, *J. Phys.: Condens. Matter* **32**, 374010 (2020).
- [20] L. Seabra and N. Shannon, Competition between supersolid phases and magnetization plateaus in the frustrated easy-axis antiferromagnet on a triangular lattice, *Phys. Rev. B* **83**, 134412 (2011).
- [21] Z. Ma, S. Zheng, Y. Chen, R. Xu, Z.-Y. Dong, J. Wang, H. Du, J. P. Embs, S. Li, *et al.*, Possible gapless quantum spin liquid behavior in the triangular-lattice Ising antiferromagnet $\text{PrMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **109**, 165143 (2024).
- [22] S. Kumar, M. Klicpera, A. Eliáš, M. Kratochvílová, A. Kancko, C. Correa, K. Załęski, M. Śliwińska-Bartkowiak, R. H. Colman, and G. Bastien, Induced quantum magnetism on a triangular lattice of non-Kramers ions in $\text{PrMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **111**, 174444 (2025).
- [23] H. Bu, M. Ashtar, T. Shiroka, H. C. Walker, Z. Fu, J. Zhao, J. S. Gardner, G. Chen, Z. Tian, and H. Guo, Gapless triangular-lattice spin-liquid candidate $\text{PrZnAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **106**, 134428 (2022).
- [24] B. Gao, T. Chen, C. Liu, M. L. Klemm, S. Zhang, Z. Ma, X. Xu, C. Won, G. T. McCandless, *et al.*, Spin excitation continuum from degenerate states in the mixed ferro-antiferromagnetic exchange system $\text{CeMgAl}_{11}\text{O}_{19}$, *Sci. Adv.* **12**, ead7778 (2026).
- [25] G. Bastien, A. Eliáš, V. Anderle, A. Kancko, C. A. Corrêa, S. Kumar, P. Proschek, J. Prokleška, L. Nádherný, *et al.*, Quantum disordered ground state and relative proximity to an exactly solvable model in the frustrated magnet $\text{CeMgAl}_{11}\text{O}_{19}$, *Phys. Rev. Res.* (2026).
- [26] S. Kumar, G. Bastien, R. H. Colman, M. Savinov, P. Proschek, M. Vališka, M. Kempínski, W. Kempínski, M. Śliwińska-Bartkowiak, and S. Kamba, Crystal-field-driven magnetoelectric coupling in the non-Kramers hexaaluminate $\text{PrMgAl}_{11}\text{O}_{19}$, *J. Alloys Compd.* **1061**, 187535 (2026).
- [27] S. Kumar, B. Salajová, A. Kancko, C. A. Corrêa, S. Halder, and R. H. Colman, Strongly quenched Kramers doublet magnetism in $\text{SmMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **113**, 224443 (2026).
- [28] S. Kumar, G. Bastien, M. Savinov, P. Proschek, A. Eliáš, K. Załęski, M. Śliwińska-Bartkowiak, R. H. Colman, and S. Kamba, Crystal field driven magnetoelectricity in the triangular quantum magnet $\text{CeMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **112**, 224431 (2025).
- [29] Y. Cao, H. Bu, T. Shiroka, H. C. Walker, Z. Fu, Z. Tian, J. Zhao, and H. Guo, Magnetic ground state and persistent spin fluctuations in the triangular-lattice antiferromagnet $\text{NdZnAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **112**, 144409 (2025).
- [30] M. Ashtar, M. A. Marwat, Y. X. Gao, Z. T. Zhang, L. Pi, S. L. Yuan, and Z. M. Tian, $\text{REZnAl}_{11}\text{O}_{19}$ ($\text{RE} = \text{Pr}, \text{Nd}, \text{Sm-Tb}$): A new family of ideal 2D triangular lattice frustrated magnets, *J. Mater. Chem. C* **7**, 10073 (2019).
- [31] L. Soderholm, C.-K. Loong, G. L. Goodman, and B. D. Dabrowski, Crystal-field splittings and magnetic properties of Pr^{3+} and Nd^{3+} in $\text{RBa}_2\text{Cu}_3\text{O}_7$, *Phys. Rev. B* **43**, 7923 (1991).
- [32] J. R. Chamorro, A. R. Jackson, A. K. Watkins, R. Seshadri, and S. D. Wilson, Magnetic order in the $S_{\text{eff}} = \frac{1}{2}$ triangular-lattice compound NdCd_3P_3 , *Phys. Rev. Mater.* **7**, 094402 (2023).
- [33] T. Arh, B. Sana, M. Pregelj, P. Khuntia, Z. Jagličić, M. D. Le, P. K. Biswas, P. Manuel, L. Mangin-Thro, *et al.*, The Ising triangular-lattice antiferromagnet neodymium heptatantalate as a quantum spin liquid candidate, *Nat. Mater.* **21**, 416 (2022).
- [34] Rigaku Oxford Diffraction, CrysAlisPro software system, version 1.171.43.143a, rigaku Oxford Diffraction Ltd., Oxford, UK (2024).
- [35] R. C. Clark and J. S. Reid, The analytical calculation of absorption in multifaceted crystals, *Acta Crystallogr. A* **51**, 887 (1995).
- [36] L. Palatinus and G. Chapuis, SUPERFLIP – a computer program for the solution of crystal structures by charge flipping in arbitrary dimensions, *J. Appl. Crystallogr.* **40**, 786 (2007).
- [37] V. Petříček, L. Palatinus, J. Plášil, and M. Dušek, Jana2020 – a new version of the crystallographic computing system Jana, *Z. Kristallogr. Cryst. Mater.* **238**, 271 (2023).

- [38] M. Klinger, T. Treu, F. Kreisberger, C. Heil, A. Klinger, A. Jesche, and P. Gegenwart, Sub-1 K adiabatic demagnetization refrigeration with rare-earth borates $\text{Ba}_3\text{XB}_9\text{O}_{18}$ and $\text{Ba}_3\text{XB}_3\text{O}_9$, $X = (\text{Yb}, \text{Gd})$, *Appl. Sci.* **16**, 290 (2025).
- [39] N. Li, A. Rutherford, Y. Y. Wang, H. Liang, Q. J. Li, Z. J. Zhang, H. Wang, W. Xie, H. D. Zhou, and X. F. Sun, Ising-type quantum spin liquid state in $\text{PrMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **110**, 134401 (2024).
- [40] D. Saber and A. M. Lejus, Elaboration and characterization of lanthanide aluminate single crystals with the formula $\text{LnMgAl}_{11}\text{O}_{19}$, *Mater. Res. Bull.* **16**, 1325 (1981).
- [41] M. Gasperin, M. C. Saine, A. Kahn, F. Laville, and A. M. Lejus, Influence of M^{2+} ions substitution on the structure of lanthanum hexaaluminates with magnetoplumbite structure, *J. Solid State Chem.* **54**, 61 (1984).
- [42] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/x1rf-xmrd> for structural analysis and point charge calculations.
- [43] K. Momma and F. Izumi, VESTA 3 for three-dimensional visualization of crystal, volumetric and morphology data, *J. Appl. Crystallogr.* **44**, 1272 (2011).
- [44] Y. Cao, H. Bu, Z. Fu, J. Zhao, J. S. Gardner, Z. Ouyang, Z. Tian, Z. Li, and H. Guo, Synthesis, disorder and Ising anisotropy in a new spin liquid candidate $\text{PrMgAl}_{11}\text{O}_{19}$, *Mater. Futures* **3**, 035201 (2024).
- [45] A. Scheie, PyCrystalField: Software for calculation, analysis and fitting of crystal electric field Hamiltonians, *J. Appl. Crystallogr.* **54**, 356 (2021).
- [46] J.-F. Wyart, A. Meftah, A. Bachelier, J. Sinzelle, W.-U. L. Tchang-Brillet, N. Champion, N. Spector, and J. Sugar, Energy levels of $4f^3$ in the Nd^{3+} free ion from emission spectra, *J. Phys. B: At. Mol. Opt. Phys.* **39**, L77 (2006).
- [47] D. Wagner, *Introduction to the Theory of Magnetism*, International Series in Natural Philosophy (Pergamon Press, Oxford, 1972).
- [48] J. A. Weil, J. R. Bolton, and J. E. Wertz, *Electron Paramagnetic Resonance: Elementary Theory and Practical Applications*, 2nd ed. (Wiley, Hoboken, 2006).
- [49] T. Bodziony, S. M. Kaczmarek, and J. Hanuza, EPR and optical studies of $\text{LiNbO}_3\text{:Yb}$ and $\text{LiNbO}_3\text{:Yb, Pr}$ single crystals, *J. Alloys Compd.* **451**, 240 (2008).
- [50] L. K. Aminov, I. N. Kurkin, and B. Z. Malkin, Superhyperfine structure in the EPR spectra and optical spectra of impurity f ions in dielectric crystals: A review, *Phys. Solid State* **55**, 1343 (2013).
- [51] J. Khatua, Q. P. Ding, M. S. Ramachandra Rao, K. Y. Choi, A. Zorko, Y. Furukawa, and P. Khuntia, Magnetic properties of a spin-orbit entangled $J_{\text{eff}} = \frac{1}{2}$ honeycomb lattice, *Phys. Rev. B* **108**, 054442 (2023).
- [52] S. Kumar, G. Bastien, M. Savinov, M. Śliwińska-Bartkowiak, R. H. Colman, and S. Kamba, Dielectric signatures of crystal-field and low-temperature correlated dynamics in $\text{NdMgAl}_{11}\text{O}_{19}$, *Phys. Rev. B* **114**, L111103 (2026).
- [53] J. Khatua, M. Pregelj, A. Elghandour, Z. Jagličić, R. Klingeler, A. Zorko, and P. Khuntia, Magnetic properties of the triangular-lattice antiferromagnets $\text{Ba}_3\text{RB}_9\text{O}_{18}$ ($R = \text{Yb}, \text{Er}$), *Phys. Rev. B* **106**, 104408 (2022).
- [54] R. Bag, M. Ennis, C. Liu, S. E. Dissanayake, Z. Shi, J. Liu, L. Balents, and S. Haravifard, Realization of quantum dipoles in triangular lattice crystal $\text{Ba}_3\text{Yb}(\text{BO}_3)_3$, *Phys. Rev. B* **104**, L220403 (2021).
- [55] G. S. Grest and E. G. Gabl, Monte Carlo study of spin-glass ordering on dilute frustrated lattices, *Phys. Rev. Lett.* **43**, 1182 (1979).
- [56] R. J. Creswick, H. A. Farach, C. P. Poole, Jr., and J. M. Knight, Monte Carlo study of the local-field distribution in the dilute antiferromagnetic Ising model on the triangular lattice, *Phys. Rev. B* **32**, 5776 (1985).
- [57] R. Moessner and A. J. Berlinsky, Magnetic susceptibility of diluted pyrochlore and $\text{SrCr}_{9-9x}\text{Ga}_{3+9x}\text{O}_{19}$ antiferromagnets, *Phys. Rev. Lett.* **83**, 3293 (1999).
- [58] V. K. Dwivedi, Recent development of magnetocaloric effect in pyrochlore oxides, in *Smart Materials for Energy Storage and Biomedical Applications*, edited by A. Mishra and V. Pathak (Springer Nature Switzerland, Cham, 2025) pp. 15–41.
- [59] Y. Zhang, Y. Na, X. Liu, J. Xiang, F. Chen, H.-F. Li, P. Sun, S. Zhou, X. Zhang, and L. Li, Refrigeration down to 0.16 K using a frustrated magnet $\text{Gd}_2\text{B}_2\text{MoO}_9$, *Nat. Commun.* **17**, 1554 (2026).
- [60] Z. W. Yang, G. Gong, F. Yuan, S. Tang, G. Zhang, X. Ye, Z. Pan, Y.-J. Zeng, and Y. Long, Disorder-tuned magnetism and magnetocaloric response in a layered $\text{SrGd}_2\text{Al}_2\text{O}_7$ single crystal, *Mater. Today Phys.* **60**, 101970 (2026).
- [61] M. Kleinhans, K. Eibensteiner, J. C. Leiner, C. Resch, L. Worch, M. A. Wilde, J. Spallek, A. Regnat, and C. Pfeleiderer, Magnetocaloric properties of $R_3\text{Ga}_5\text{O}_{12}$ ($R = \text{Tb}, \text{Gd}, \text{Nd}, \text{Dy}$), *Phys. Rev. Appl.* **19**, 014038 (2023).
- [62] C. Delacotte, T. A. Pomelova, T. Stephant, T. Guizouarn, S. Cordier, N. G. Naumov, and P. Lemoine, NaGdS_2 : A promising sulfide for cryogenic magnetic cooling, *Chem. Mater.* **34**, 1829 (2022).
- [63] B. Shanta, X. Zhao, N. Li, and X. F. Sun, Magnetocaloric effect in frustrated magnetic oxides, *J. Sci.: Adv. Mater. Devices* **10**, 100932 (2025).
- [64] <http://mgml.eu/>.