

Quantum disordered ground state and relative proximity to an exactly solvable model in the frustrated magnet $\text{CeMgAl}_{11}\text{O}_{19}$

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(Received 16 June 2025; accepted 19 August 2026; published 19 October 2026)

The magnetic properties of the triangular magnet $\text{CeMgAl}_{11}\text{O}_{19}$ were investigated by magnetization and specific heat measurements down to $T = 0.03$ K on single crystals grown by the floating zone method. The formation of effective spins $S_{\text{eff}} = 1/2$ below $T < 10$ K was confirmed by both density functional theory calculations and specific heat measurements. No magnetic order was found down to $T = 0.03$ K despite the formation of magnetic correlations observed in specific heat. The measured magnetization was compared with density matrix renormalization group calculations and their agreement supports the proposal of a strongly anisotropic magnetic interaction antiferromagnetically coupling the spin components in the ab plane and ferromagnetically coupling the spin component along the c axis. However, our quantitative study of the magnetization places $\text{CeMgAl}_{11}\text{O}_{19}$ further from the ferromagnetic quantum critical point than inferred from the previous inelastic neutron scattering study. Finally, we suggest that the absence of magnetic order may be promoted by the structural disorder revealed by single-crystal x-ray diffraction [Ce deficiency, Mg/Al site mixing, and Al(5) splitting].

DOI: [10.1103/kqxq-g723](https://doi.org/10.1103/kqxq-g723)

The research on quantum magnetism is motivated by the possible formation of magnetically disordered states where quantum entanglement plays a role, such as quantum spin liquid states [1–4], valence bond solids [5,6], and random singlet (RS) states [7,8]. Over the past decade, Kitaev magnets on honeycomb lattices have attracted considerable attention due to the analytic prediction of a quantum spin liquid state at the limit of uniaxial magnetic interactions [9,10]. Interestingly, the $S = 1/2$ triangular lattice antiferromagnet (TLAF) also reaches a point where it becomes exactly solvable and a spin liquid state is predicted [11,12].

We consider the $S = 1/2$ TLAF with XXZ anisotropy and without any off-diagonal coupling [11–15]:

$$\hat{H} = \sum_{\langle i,j \rangle} J_{\perp} (S_i^x S_j^x + S_i^y S_j^y) + J_z S_i^z S_j^z. \quad (1)$$

In the case of an anisotropic interaction antiferromagnetically coupling spin components in the ab plane ($J_{\perp} > 0$) and ferromagnetically coupling the spin component along the c axis ($J_z < 0$), this Hamiltonian harbors a quantum critical point (QCP) at $J_z/J_{\perp} = -0.5$. The QCP separates a 120°

coplanar antiferromagnetic order from a ferromagnetic order [11,12]. At the QCP, the problem is exactly solvable and it harbors a spin liquid ground state describable as a superposition of various umbrella states [11–13,16].

Ce-based TLAFs are promising materials for the search of real materials in proximity to this QCP, since most of them harbor effective spins $S_{\text{eff}} = 1/2$ at low temperature [17–21] and strongly anisotropic magnetic interactions induced by the strong spin-orbit coupling [12,18,22–24]. Recently, the TLAF $\text{CeMgAl}_{11}\text{O}_{19}$ was proposed to be located close to the QCP of the XXZ Hamiltonian (1) from inelastic neutron scattering (INS) experiments [12]. $\text{CeMgAl}_{11}\text{O}_{19}$ crystallizes in the magnetoplumbite structure (space group $P6_3/mmc$) [25,26], which is characterized by a large interplane separation between the triangular magnetic layers [27,28]. The presence of horizontal and vertical mirror planes in this crystal structure cancels off-diagonal symmetric exchange, whereas they play a major role in other Ce-based TLAFs such as KCeS_2 [23] and CsCeSe_2 [24]. Importantly, an absence of antiferromagnetic order in $\text{CeMgAl}_{11}\text{O}_{19}$ down to $T = 0.05$ K was reported from specific heat measurements [12,21], neutron diffraction [12], and muon spectroscopy [21]. In addition, INS experiments pointed out a broad excitation continuum attributed to an ensemble average of spin-wave spectra over a nearly degenerate ground-state manifold [12]. Therefore, the low-temperature state of $\text{CeMgAl}_{11}\text{O}_{19}$ was interpreted as a disorder-stabilized, quantum-disordered state associated with proximity to the degenerate manifold of the ferromagnetic-antiferromagnetic XXZ Hamiltonian [12].

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In this Letter, we report a single-crystal study of $\text{CeMgAl}_{11}\text{O}_{19}$ confirming the formation of $S_{\text{eff}} = 1/2$ effective spins at low temperature and the absence of magnetic order down to $T = 0.03$ K. Considering magnetization as the appropriate probe to quantify the proximity to ferromagnetic quantum criticality, we combined magnetization measurements at temperature down to $T = 0.03$ K with density matrix renormalization group (DMRG) calculation. Our data confirm the proposal of strongly anisotropic magnetic interactions but it indicates a weaker proximity to ferromagnetic quantum criticality than previously proposed from INS [12]. These results disfavor a scenario in which the absence of magnetic order is driven primarily by close proximity to the ferromagnetic quantum critical point, and instead point to an important role of structural disorder.

The polycrystalline precursor for the crystal growth was first synthesized by sol-gel methods as described in Refs. [29,30]. Then, single crystals were grown by the floating zone method in an optical furnace in air as described in Refs. [31,32]. Powder x-ray diffraction revealed that our ingots contained grains of two different materials, the hexaaluminate $\text{CeMgAl}_{11}\text{O}_{19}$ and the spinel $\text{Mg}_x\text{Al}_{8/3-2x/3}\text{O}_4$. X-ray fluorescence spectroscopic mapping showed that these two phases are well separated and allowed confirmation of the composition of the $\text{CeMgAl}_{11}\text{O}_{19}$ phase (see the Supplemental Material [33], which includes Refs. [12,16,28,34–47]).

Magnetization measurements at temperature down to $T = 1.8$ K were performed using an magnetic properties measurement system (MPMS) magnetometer and a physical properties measurement system (PPMS) with vibrating sample magnetometer option, from Quantum Design. These measurements were extended down to $T = 0.03$ K in a Triton dilution refrigerator using the Hall probe method [28,48]. Magnetization measurements were carried out on a nearly cubic crystal with an estimated average demagnetization factor of $N \approx 0.32$ [49]. The specific heat was measured both by the relaxation method in the PPMS and the Triton dilution refrigerator and by adiabatic demagnetization refrigeration (ADR). The magnetic contribution to the specific heat was extracted using a nonmagnetic analog, $\text{LaMgAl}_{11}\text{O}_{19}$ (see the Supplemental Material [33]). The measurement by ADR involved the self-cooling of the sample from 1.8 K to 81 mK by demagnetization from 14 T to zero applied field, on a powdered sample. Constant heat flux on warming back to 1.8 K allowed calculation of the specific heat, as detailed in Refs. [39,40].

Single-crystal diffraction confirms the magnetoplumbite crystal structure with the space group $P6_3/mmc$ [12,21,25]. The diffraction results unveil three sources of structural disorder: The Al(5) ion is distributed between two off-centered positions in the oxygen bipyramid as commonly observed in the magnetoplumbite structure [12,21,31,50–52]; a partially occupied Ce site, which was refined to 10% deficiency and is similar to the 7% deficiency previously reported in crystals grown in a reducing atmosphere [12]; and mixed Mg/Al occupation at the Al(4) site. Even though it is not possible to confirm that the Mg^{2+} ions preferably sit at the Al(4) site through our single crystal x-ray diffraction results, this was previously confirmed by neutron diffraction for the $\text{LnMgAl}_{11}\text{O}_{19}$ ($\text{Ln} = \text{La–Gd}$) series [12,50,53]. Experimental

and structural details are given in the Supplemental Material [33–38].

In order to test theoretically the description of $\text{CeMgAl}_{11}\text{O}_{19}$ as an effective spin $S_{\text{eff}} = 1/2$ magnet, we calculated the crystal electric field (CEF) energy levels of the ground-state multiplets of Ce^{3+} . These calculations were performed using the recently developed method based on a combination of density functional theory plane wave code using VASP [41] and Wien-2k program [42–45] (full computational details are given in the Supplemental Material [33]). The crystal field parameters were defined from the Wannier transformed local Hamiltonian $\hat{H}_{\text{loc}}$ by expanding it into a series of spherical tensor operators $\hat{C}_q^k$:

$$\begin{aligned}\hat{H}_{\text{loc}} &= \sum_{k=2,4,6} \sum_{q=-k}^k B_q^k \hat{C}_q^k, \\ B_0^2 &= 52.16 \text{ meV}, \quad B_0^4 = 19.68 \text{ meV}, \\ B_0^6 &= -154.86 \text{ meV}, \quad B_6^6 = -122.68 \text{ meV}. \quad (2)\end{aligned}$$

Solving the crystal field Hamiltonian for D_{3h} ($-6m2$) local symmetry of Ce^{3+} , we end up with a diagonal matrix with the nonzero elements being equal to eigenvalues of three Kramers doublets

$$\begin{aligned}E_{\pm 5/2} &= -\frac{2}{7}B_0^2 + \frac{1}{21}B_0^4 = -13.96 \text{ meV}, \\ E_{\pm 3/2} &= \frac{2}{35}B_0^2 - \frac{1}{7}B_0^4 = +0.17 \text{ meV}, \\ E_{\pm 1/2} &= \frac{8}{35}B_0^2 + \frac{2}{21}B_0^4 = +13.80 \text{ meV}.\end{aligned}$$

These results imply the quasiexclusive selection of the $J_z = \pm 5/2$ magnetic state at temperatures $T \ll (E_{\pm 5/2} - E_{\pm 3/2})/k = 164$ K. Therefore, it validates previous interpretations of the magnetism of $\text{CeMgAl}_{11}\text{O}_{19}$ below $T = 10$ K as an effective spin $S_{\text{eff}} = 1/2$ magnet [12,21]. The CEF scheme was further validated from the comparison of the temperature dependence of the effective moment with experimental data as shown in the Supplemental Material [33].

The magnetic susceptibility of $\text{CeMgAl}_{11}\text{O}_{19}$ as a function of temperature down to $T = 0.03$ K is shown in Fig. 1(a). It is characterized by a strong anisotropy with the c axis as the easy magnetization axis. Below $T = 0.2$ K, the dc magnetic susceptibility shows a plateau, in agreement with the ac magnetic susceptibility recently reported [21]. A Curie-Weiss fit performed in the temperature interval $5 \text{ K} < T < 15 \text{ K}$ gives a Curie-Weiss temperature of $\theta_{\text{CW}} = 0.3 \pm 0.3$ K. The positive value hints at ferromagnetic interactions in agreement with the proposal of a magnetic interaction ferromagnetically coupling the S^z spin component [12].

Magnetization measurements at magnetic fields up to $\mu_0 H = 14$ T at a temperature of $T = 2$ K indicate a clear magnetic saturation along the easy magnetization axis $H \parallel c$ at $M_{\text{sat}} = 1.68 \mu_B/\text{Ce}$. It implies an effective g factor of $g_c = 3.36$ in a relatively good agreement with previous electron spin resonance studies in $\text{La}_{1-x}\text{Ce}_x\text{MgAl}_{11}\text{O}_{19}$ and $\text{CeMgAl}_{11}\text{O}_{19}$ giving $g_c = 3.74$ and $g_c = 3.66$, respectively [12,54]. The g factor in the ab plane was estimated at $g_{ab} \approx 0.6$ upon approaching the magnetization by a Brillouin function. It implies a large anisotropy of the effective g tensor with $g_c/g_{ab} \approx 6$. This anisotropy of the magnetization differs from

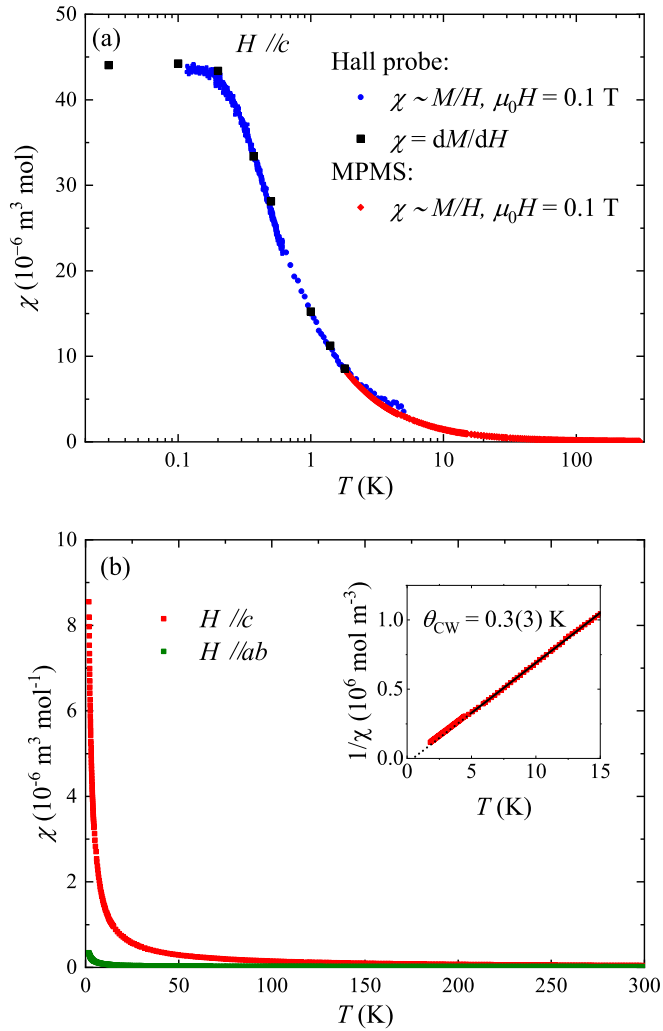


FIG. 1. (a) Magnetic susceptibility of $\text{CeMgAl}_{11}\text{O}_{19}$ along the easy magnetization axis c as a function of temperature combining data from the MPMS magnetometer and the Hall sensor. Data points obtained upon temperature scan at a magnetic field of $\mu_0 H = 0.1$ T (disks) are confirmed by direct measurement of the derivative $\partial M / \partial H$ (black squares). (b) Magnetic susceptibility of $\text{CeMgAl}_{11}\text{O}_{19}$ under magnetic field applied both along the c axis and in the ab plane. The inset shows the Curie-Weiss fit in the temperature interval $5 \text{ K} < T < 15 \text{ K}$ for magnetic field applied along the c axis.

the much weaker anisotropy reported in Ref. [12] and it agrees with data reported in Ref. [21].

The isothermal magnetization along the c axis in $\text{CeMgAl}_{11}\text{O}_{19}$ at different temperatures down to $T = 0.03$ K is represented in Fig. 2(b). At the lowest temperature of $T = 0.03$ K, the magnetization is linear in applied magnetic field until the saturation field of $\mu_0 H_{\text{sat}} \approx 0.25$ T. The absence of any magnetization plateau is consistent with the assumption of an anisotropy of the magnetic interaction $J_z < J_{\perp}$ [14,15]. The saturation field is relatively weak with a ratio of $g_c \mu_B \mu_0 H_{\text{sat}} / J_{\perp} \approx 0.9$, which is far below the theoretical value at the Heisenberg limit $g \mu_B \mu_0 H_{\text{sat}} / J = 9/2$ [14,15,55]. This result is qualitatively consistent with a proximity of $\text{CeMgAl}_{11}\text{O}_{19}$ to quantum criticality toward ferromagnetism.

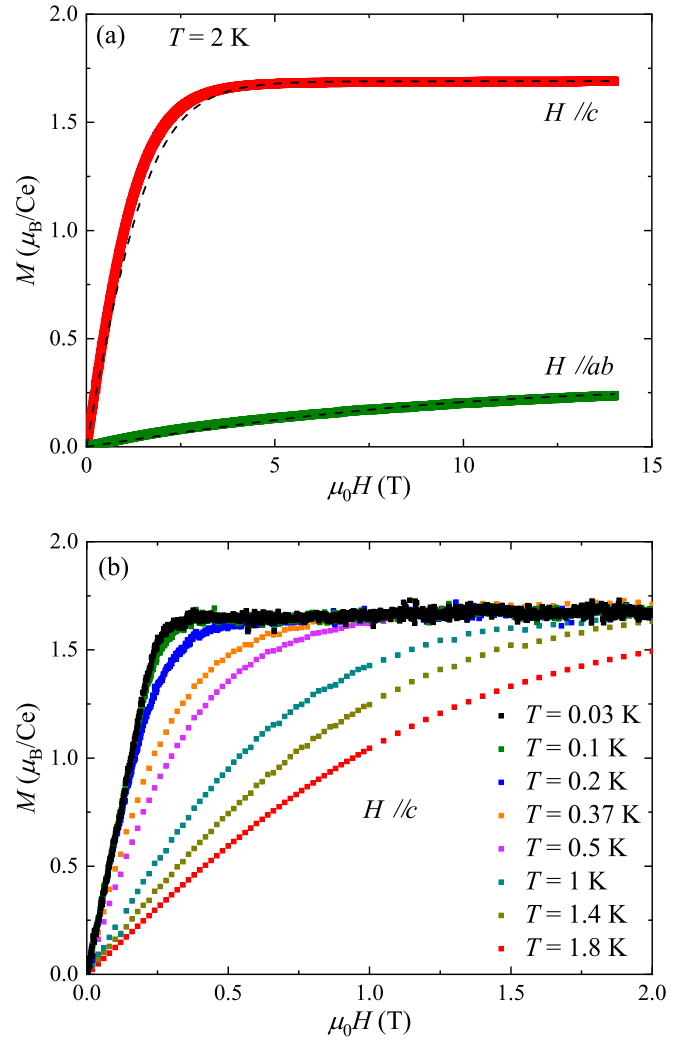


FIG. 2. (a) Magnetization at $T = 2$ K in $\text{CeMgAl}_{11}\text{O}_{19}$ along the c axis and in the ab plane as a function of magnetic field. The dashed line indicates a fit of the magnetization to Brillouin functions giving the g factor values: $g_c = 3.4$ and $g_{ab} \approx 0.6$. (b) Magnetization of $\text{CeMgAl}_{11}\text{O}_{19}$ along the c axis at different temperatures from $T = 0.03$ K to $T = 1.8$ K.

To quantify the proximity of $\text{CeMgAl}_{11}\text{O}_{19}$ to ferromagnetic quantum criticality, we analyzed magnetization using DMRG calculations based on Hamiltonian (1) for various sets of parameters. The calculations were performed using the TeNPy library (version 1.0.4) [46] with a lattice size of $L = 12 \times 9$ and cylindrical boundary conditions. Details about the calculation method can be found in the Supplemental Material [33]. To test the effect of the ratio $J_z/J_{\perp}$, we kept the strongest interaction at a fixed value $J_{\perp} = 0.056$ meV from Ref. [12] and we performed calculations for various values of J_z . The computational data at $T = 0$ were compared with the experimental data measured at $T = 0.03$ K in Fig. 3(a). To check whether the demagnetization effect could influence the fitting by DMRG, the demagnetization effect was estimated using the approximation proposed in Ref. [49]. It implies a minor correction that was taken into account in analyzing experimental results.

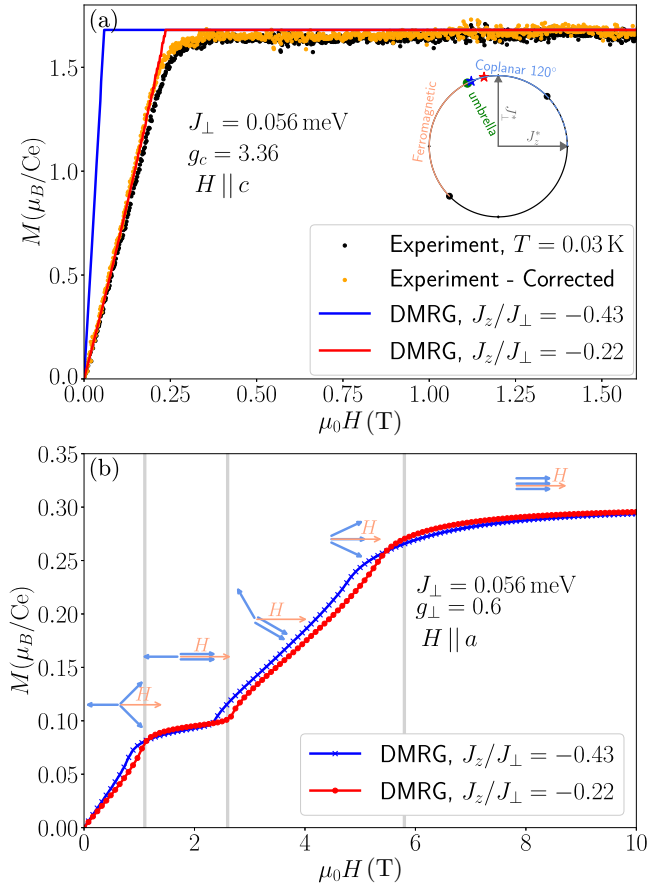


FIG. 3. (a) Comparison of the experimental magnetization at $T = 0.03$ K in $\text{CeMgAl}_{11}\text{O}_{19}$ along the c axis as a function of magnetic field with the DMRG ground-state calculations. The black and orange dots correspond to the magnetization as measured and after correction of the demagnetization effect, respectively. The red line is our best fit where $J_z/J_{\perp} = -0.22$ and the blue line shows results for $J_z/J_{\perp} = -0.43$, which was the value extracted from the best fit of INS data in Ref. [12]. The circle in the inset shows approximate ground-state phase diagram in zero magnetic field [11], with green point showing the exactly solvable critical point and colored lines show the ground-state phases. The stars represent the respective parameter solutions. Here, $J_z^* = J_z/\sqrt{J_z^2 + J_{\perp}^2}$ and $J_{\perp}^* = J_{\perp}/\sqrt{J_z^2 + J_{\perp}^2}$. (b) Theoretical ground-state magnetization along the a axis as a function of magnetic field for $J_z/J_{\perp} = -0.43$ (blue symbols) and $J_z/J_{\perp} = -0.22$ (red symbols), predicting a $1/3$ magnetization plateau. Inserted schematics show the different spin textures, which were determined from structure factor calculations (see the Supplemental Material [33]).

Our best fit value $J_z/J_{\perp} = -0.22$ confirms the presence of strongly anisotropic magnetic interactions that ferromagnetically couple the spin component S^z . However, it places $\text{CeMgAl}_{11}\text{O}_{19}$ further away from the quantum critical point at $J_z/J_{\perp} = -0.5$ than fits of INS data suggesting $J_z/J_{\perp} = -0.43$ [12]. This INS-derived parameter set was also recently shown, using a $U(1)$ -symmetric pseudo-Majorana functional renormalization group approach, to reproduce the finite-temperature out-of-plane magnetization data reported in Ref. [12] at $T = 2$ and 5 K [56]. Our analysis instead

constrains the exchange ratio from the ultralow-temperature magnetization and saturation field down to $T = 0.03$ K, where the ground-state response is more directly probed. Such quantitative differences may come from sample dependences, for instance, due to different levels of structural disorder noted in all crystals, although so far the bulk magnetic and thermodynamic probes appear consistent [12,21]. Furthermore, DMRG ground-state calculations of a perfect two-dimensional system predict the formation of a 120° coplanar order for $J_z/J_{\perp} = -0.22$ and its evolution into an umbrella state under magnetic field as shown in the Supplemental Material [33] with structure factor calculations [33]. Therefore, our experimental results do not support a ground state driven solely by close proximity to the exactly solvable point $J_z/J_{\perp} = -0.5$.

The zero-temperature magnetization in the ab plane computed by DMRG is represented in Fig. 3(b). These calculations predict a magnetization plateau with $M/M_{\text{sat}} \approx 1/3$ for $\mu_0H \approx 1.8$ T, reflecting the formation of the up-up-down order [14,15,55]. Unfortunately, these predictions could not be tested due to low magnetization and the need to measure at much higher magnetic field for $H \parallel ab$ than for $H \parallel c$, implying a much lower resolution. Experimentally, an $H \parallel ab$ magnetization measurement at $T = 0.5$ K up to 7 T shows no clear plateau or slope anomaly (see the Supplemental Material [33]), most likely because the $T = 0$ features are thermally broadened at this temperature; lower-temperature measurements in high in-plane fields would be required to test the predicted plateau directly.

The specific heat divided by temperature C_p/T in $\text{CeMgAl}_{11}\text{O}_{19}$ as a function of temperature below $T = 0.6$ K is represented in Fig. 4(b). It does not show any peak that would indicate long-range magnetic ordering. On the contrary, C_p/T in the absence of an external magnetic field harbors a broad maximum around $T \approx 0.12$ K. This maximum broadens under the application of magnetic field up to the saturation field of $\mu_0H \approx 0.25$ T excluding the formation of magnetic-field-induced long-range magnetic order. Under magnetic fields higher than the saturation field $\mu_0H_{\text{sat}} = 0.25$ T, the maximum of C_p/T shifts to higher temperature with increasing field as commonly observed in ferromagnets [28].

The entropy variation with temperature and magnetic field was obtained based on the integration of magnetization and specific heat as explained in Ref. [18] and it is represented in Fig. 4(b). The difference of entropy between magnetic saturation at $T = 0.4$ K and $\mu_0H > 3$ T and the paramagnetic regime at 5 K $< T < 15$ K and $\mu_0H = 0$ approaches $\Delta S = R \ln(2)$. This result further confirms the formation of effective spins $S_{\text{eff}} = 1/2$. The strong dependence of the entropy on magnetic field implies the possibility to cool $\text{CeMgAl}_{11}\text{O}_{19}$ samples using adiabatic demagnetization of the sample itself. We demonstrated this possibility in the same setup used in Refs. [39,40]. Demagnetization from $\mu_0H = 5$ T at $T = 2$ K yields a final temperature of $T = 0.09$ K. The subsequent warm-up curve was used to calculate $C_p(T)$, displayed as line in Fig. 4(a).

The DMRG analysis places the clean XXZ triangular-lattice model with $J_z/J_{\perp} = -0.22$ in the zero-temperature 120° coplanar region. This should not be interpreted as a direct contradiction with the absence of experimentally observed

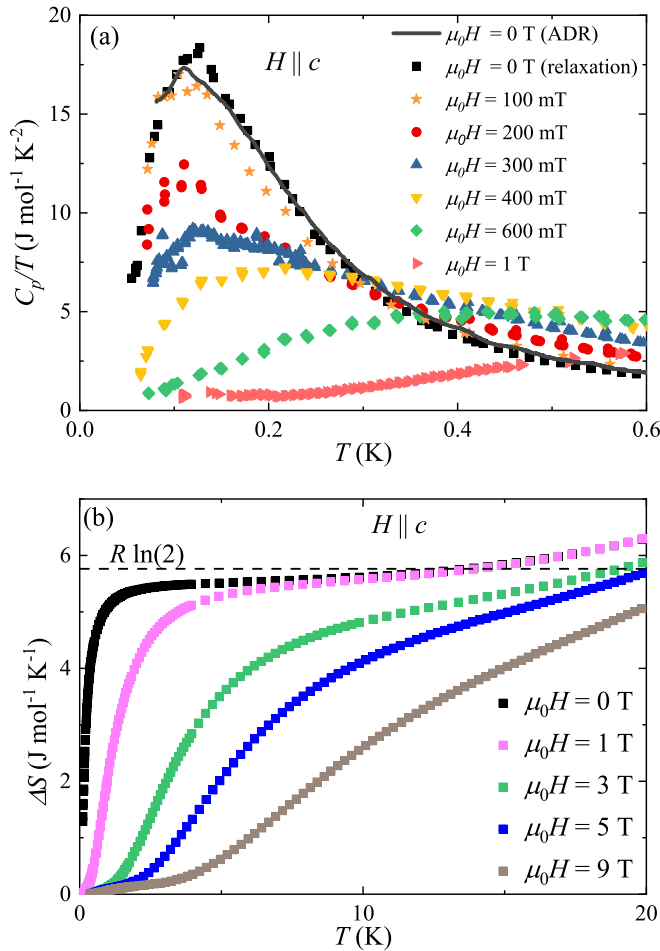


FIG. 4. (a) C_p/T in $\text{CeMgAl}_{11}\text{O}_{19}$ below $T = 0.6$ K as a function of temperature and magnetic field applied along the c axis. The zero-field specific heat was measured with both the relaxation method and adiabatic demagnetization refrigeration. (b) Variation of entropy $\Delta S = S - S(T = 0.4 \text{ K}, \mu_0 H = 9 \text{ T})$ as a function of temperature and magnetic field. The horizontal dashed line indicates the expected value of $R \ln(2)$.

magnetic long-range order down to 0.03 K, since in a strictly two-dimensional system true finite-temperature long-range ordering of the continuous in-plane spin component is not expected. Although in a real material even a weak interlayer coupling can trigger a short-range order, anisotropies or disorder can strongly influence the low-temperature response [7,8,57–60].

At present, we have no direct evidence tying the observed disorder to the magnetic response, and narrowing to a specific source (Ce deficiency, Mg/Al site mixing, and Al(5) splitting) remains beyond our DMRG calculations. The most studied disordered-induced quantum magnetic state is the RS state proposed in YbMgGaO_4 [4,7,8]; however, the occurrence of this phase in $\text{CeMgAl}_{11}\text{O}_{19}$ is unlikely since power law divergence of χ and C_p/T toward $T = 0$ was predicted for an RS

state [8,61,62] in contradiction with our data. Alternatively, we could propose the formation short-range 120° magnetic order in $\text{CeMgAl}_{11}\text{O}_{19}$ as previously proposed in YbMgGaO_4 [59] and in NaYbSe_2 [60]. This emphasis on disorder is consistent with the recent INS interpretation that weak quenched disorder can inhibit unique ground-state selection and stabilize a local distribution within a nearby degenerate manifold, although our magnetization analysis places the present sample further from the exactly solvable point [12]. In addition, the strong g -factor anisotropy substantially suppresses the spectral weight of in-plane fluctuations in INS, which may hinder direct observation of low-energy modes near the K point.

To conclude, our single-crystal study of the $S_{\text{eff}} = 1/2$ effective spin TLAF $\text{CeMgAl}_{11}\text{O}_{19}$ confirms the absence of magnetic ordering down to $T = 0.03$ K. The combination of magnetization measurement down to $T = 0.03$ K and DMRG calculation confirms the proposal of strongly anisotropic magnetic interactions antiferromagnetically coupling in-plane spin components and ferromagnetically coupling the out-of-plane spin component. However, it suggests that $\text{CeMgAl}_{11}\text{O}_{19}$ is further from the ferromagnetic quantum critical point than previously inferred, where a spin liquid was analytically predicted. Finally, we argue that the disordered magnetic ground state of $\text{CeMgAl}_{11}\text{O}_{19}$ arises from the combination of magnetic frustration, structural disorder, and proximity to quantum criticality, and the question whether this magnetic state can be classified as a quantum spin liquid remains open for future debate.

We acknowledge funding from the Charles University in Prague within the Primus research program with Grant No. PRIMUS/22/SCI/016. The work was also supported by the Ministry of Education, Youth and Sports of the Czech Republic through programs e-INFRA CZ (No. 90254) and INTER-EXCELLENCE II INTER-ACTION (IDLU-ABA24056). M.Ž. acknowledges support from the Czech Science Foundation via Project No. 26-22148S and RHC via Project No. 26-23051S. The work in Augsburg was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Grants No. 514162746 (GE 1640/11-1) and No. 492547816 (TRR 360) as well as by the Bavarian-Czech University Agency, Project No. BTHA-JC-2024-15. Crystal growth, structural analysis, and magnetic property measurements were carried out in the MGML [63], which is supported within the program of Czech Research Infrastructures (Project No. LM2023065). We acknowledge Šárka Matoušková and Jan Rohovec from the Institute of Geology of the Czech Academy of Sciences (RVO 67985831) for their help for the determination of crystal composition.

Data availability. The crystallographic data that support the findings of this article are available from the Cambridge Crystallographic Data Centre, CCDC (No. 2454784). All other experimental data are available upon reasonable request to the authors.

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