

Field-induced magnetic phase transition driven by a ground-state level crossing in CsErSe₂

Hope Whitelock¹, Allen O. Scheie², Marissa McMaster¹, Ian A. Leahy¹, Li Xiang³, Mykhaylo Ozerov³, Dmitry Smirnov³, Eun Sang Choi³, C. dela Cruz⁴, M. O. Ajeesh^{2,5}, Eliana S. Krakovsky^{6,7}, Daniel A. Rehn⁷, Jie Xing⁴, Athena S. Sefat⁸, and Minhyea Lee^{1,*}

¹Department of Physics, *University of Colorado*, Boulder, Colorado 80309, USA

²Los Alamos National Laboratory, Los Alamos, New Mexico, USA

³National High Magnetic Field Laboratory, Tallahassee, Florida, USA

⁴Neutron Scattering Division, *Oak Ridge National Laboratory*, Oak Ridge, Tennessee 37831, USA

⁵Department of Physics, *Indian Institute of Technology Palakkad*, Kanjikode, Kerala 678623, India

⁶Department of Materials Science & Engineering, *Stanford University*, Stanford, California 94305, USA

⁷Computational Physics Division, *Los Alamos National Laboratory*, Los Alamos, New Mexico 87545, USA

⁸Materials Science and Technology Division, *Oak Ridge National Laboratory*, Oak Ridge, Tennessee 37831, USA



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We report a comprehensive study of the low-temperature magnetic properties of the insulating rare-earth triangular magnet CsErSe₂. We uncover a field-induced level crossing at the crystal electric field ground state, which gives rise to a first-order phase transition as well as to distinctive magnetic properties. This crossing is identified by the accurate determination of the single-ion Hamiltonian, the reliability of which is substantially enhanced by field-dependent optical spectroscopy that directly tracks the Zeeman splittings of Kramers doublets. We also observe spontaneous antiferromagnetic ordering in CsErSe₂ below $T_N \approx 110$ mK and resolve the corresponding magnetic structure using elastic neutron scattering. We discuss how the rich magnetic behavior of CsErSe₂ arises from the interplay of nontrivial field-dependent single-ion physics and spontaneous ordering, and highlight the implications of these results for understanding magnetic phenomena across a wide range of insulating rare-earth magnets.

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I. INTRODUCTION

A primary goal at the interface of theoretical and experimental efforts in quantum magnetism is to understand exotic spin states, most notably quantum spin liquids, which host fractionalized excitations, long-ranged entanglement, and topological order [1,2]. Magnetic insulators with strong spin-orbit coupling (SOC) are now widely recognized as a promising platform for accessing such unconventional ground states by extending the nature of magnetic frustration.

Compounds based on 4f rare-earth (RE) ions have provided fertile ground for quantum magnetism research, thanks to their unique combination of geometric frustration and strong SOC [3–5]. In these materials, however, a detailed understanding of the microscopic physics is a prerequisite for developing minimal spin models for effective theories of many-body phenomena [3,4].

The key to a microscopic description of the magnetic interactions in RE magnets is a reliable determination of the single-ion crystal electric-field (CEF) Hamiltonian ($\mathcal{H}_{\text{CEF}}$),

comprising a linear combination of Stevens' operators ($\hat{O}_n^m$) allowed by the site symmetry of RE ion and their coefficients (B_n^m). CEF effects split the degeneracy of the ground multiplet of the total angular momentum J [6,7]. This produces a set of Kramers doublets for RE ions with half-odd-integer J . At sufficiently low temperatures, the resulting dynamics are confined to the lowest doublet, thereby justifying an effective pseudospin-1/2 description and enabling the application of the extensive theoretical framework developed for frustrated magnetism.

The delafossite (ARESe₂, $A = \text{K, Na, Cs}$ and $\text{RE} = \text{rare-earth ions}$) family, which hosts a two-dimensional triangular net of RE ions layered along the c axis [8], has attracted much attention. Of particular interest are the compounds with Kramers ions such as Yb^{3+} ($J = 7/2$) [9–13] and Er^{3+} ($J = 15/2$) [14–16], which undergo Zeeman splitting in magnetic fields [12]. When multiple CEF levels are closely spaced, their field-induced splittings can host level crossings, resulting in distinctive field-dependent spectra. When such a crossing occurs in the ground state, it gives rise to a field-induced phase transition, profoundly impacting the magnetic properties. Figure 1(a) schematically shows the Zeeman splittings of the two lowest-lying doublets upon applying a magnetic field. The discontinuous change in the slope of the ground-state energy at the critical field $B = B_C$ indicates a first-order phase transition at zero temperature. Furthermore, the crossing of excited states, marked as B_x , results in the nonmonotonic Zeeman gap that closes and reopens at B_C , which has a profound impact

*Contact author: minhyea.lee@colorado.edu

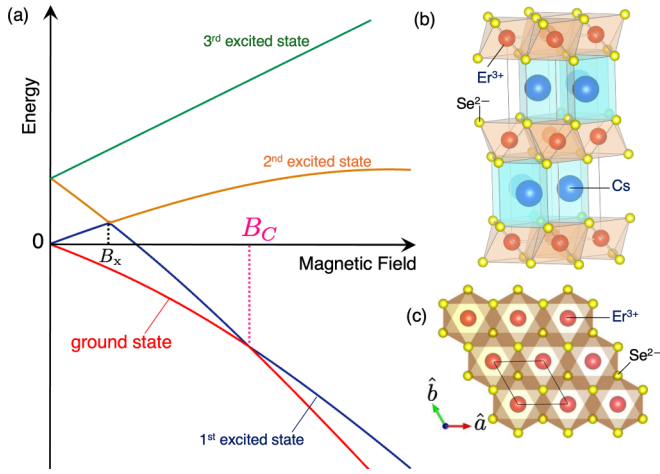


FIG. 1. (a) Schematic sketch for field-induced level splitting of the two lowest-energy doublets in CsErSe₂ is shown. The ground state undergoes a first-order quantum phase transition at B_C , where the derivative of the energy with respect to B discontinuously changes. Another crossing between the first and second excited states shown at B_x . (b) Crystal structure of CsErSe₂ adopting $P6_3/mmc$ space group [8]. (c) Er³⁺ (red) triangular layers on the ab plane formed by edge-sharing ErSe₆ octahedra, exhibiting D_{3d} site symmetry.

to give rise to complex and rich field dependence of magnetic properties.

In this work, we identify a field-induced phase transition arising from the level crossing in the ground state of the delafossite CeErSe₂ at $B_C \approx 5.4$ T, when magnetic field (B) is applied along the crystalline c axis ($\mathbf{B} \parallel c$). To our best knowledge, such field-induced level crossings involving the ground state and the corresponding field-induced transition have not been explicitly predicted from a well-characterized CEF Hamiltonian.

This first-order transition is manifested in a steplike change in magnetization (M_c) and a corresponding conspicuous peak at B_C in magnetic susceptibility as a function of field. We performed ac susceptibility measurements and verified a peak at B_C . In $B < B_C$, the nonmonotonic Zeeman gap exhibits its maximum at B_x —the crossing of the first and second excited states [Fig. 1(a)]—which leads to a plateaulike field dependence of magnetization before its steplike increase at B_C .

We predicted this transition and associated magnetic properties based on our model Hamiltonian $\mathcal{H}_{\text{tot}}$ for the system, which consists of the CEF ($\mathcal{H}_{\text{CEF}}$), Zeeman ($\mathcal{H}_Z$), and XXZ-like exchange interactions ($\mathcal{H}_{\text{xxz}}$) within the Weiss mean-field (MF) approximation:

$$\mathcal{H}_{\text{tot}} = \mathcal{H}_{\text{CEF}} + \mathcal{H}_Z + \mathcal{H}_{\text{xxz}}. \quad (1)$$

Because of a weak contribution from exchange interaction in the f -electron systems like CsErSe₂, the robust determination of $\mathcal{H}_{\text{CEF}}$ is most crucial to accurately predicting magnetic behavior. We achieve this by directly probing the field dependence of the CEF level splitting using magneto-optical spectroscopy. We find that leveraging the magnetic field offers a unique opportunity to determine the parameters of $\mathcal{H}_{\text{CEF}}$, leading us to accurate predictions.

Finally, we report long-range magnetic ordering in CsErSe₂ below $T_N = 110$ mK. Our neutron diffraction reveals stripe antiferromagnetic ordering. Experimentally observed T_N is consistent with in-plane exchange energy obtained from our fitting, which reassures us that the validity of the MF approximation extends to low temperatures.

With an accurately determined microscopic Hamiltonian, our work reveals the rich CEF physics of a single ion under high magnetic field. This opens a new avenue for exploring frustrated magnetism in rare-earth systems, enables identification of the origin of unusual magnetic properties, and establishes a firm foundation for disentangling collective inter-ion interactions from single-ion physics.

This paper is organized as follows. Section II introduces the CsErSe₂ sample and experimental methods. In Sec. III A, we first present magneto-optical spectroscopic data obtained from far-infrared (FIR) and Raman experiments and the determination of the set of parameters in the Hamiltonian in Eq. (1) based on those data. Section III B reports ac magnetic susceptibility measurements as a function of temperature and field, compared with the predictions of our Hamiltonian. Section III C presents the long-range magnetic order revealed by elastic neutron scattering experiments. Section IV discusses the overall implications of our work and Sec. V summarizes the study.

II. METHODS

CsErSe₂ crystallizes in a $P6_3/mmc$ space group [8,14] as illustrated in Fig. 1(b). The triangular-lattice planes [Fig. 1(c)] consist of edge-sharing ErSe₆ octahedra, allowing D_{3d} site symmetry for Er³⁺, and stacks along the c axis. Within the ab plane, the Se atoms mediate antiferromagnetic superexchange interactions between nearest-neighbor Er³⁺ ion pairs.

Single crystals of CsErSe₂ used in these experiments were grown from powder samples using CsCl flux as described in Ref. [8]. Typical sizes of single crystals used for optical spectroscopy are $(1\text{--}2) \times (0.3\text{--}0.8) \times (0.02\text{--}0.04)$ mm³.

The magneto-optical spectroscopy was performed with both Raman and far-infrared measurements at the National High Magnetic Field Laboratory. Raman experiments were done in a Quantum Design Physical Property Measurement System with a magnetic field up to 14 T. Scattered light was collected in backscattering geometry from the ab plane of a CsErSe₂ crystal with the magnetic field applied along the c axis (Faraday geometry) using an unpolarized 532 nm laser focused to a 1–2 μm diameter spot with the incident power of 0.5–1 mW. Collected signal spectra were analyzed using a monochromator (HRS750, Teledyne Princeton Instruments, 1200 g/mm grating) and recorded by a liquid-nitrogen-cooled CCD camera (PyLoN:400BR, Teledyne Princeton Instruments) with a spectral resolution of approximately 1.3 cm^{−1}.

FIR spectroscopy was performed in a 17.5 T superconducting magnet with a Fourier-transform infrared (FTIR) spectrometer by Bruker Vertex 80v. Broadband IR light provided by the FTIR spectrometer is guided via evacuated brass light pipes to the sample space, situated at the center of the magnet. Transmitted IR light is delivered to a composite Si bolometer (IR Labs, Inc.) mounted in the same cryogenic

environment, cooled by low-pressure helium gas to a temperature of approximately 5 K. We used both Faraday ($\mathbf{B} \parallel c$) and Voigt ($\mathbf{B} \parallel b$) geometries with the wave vector of the incoming IR light to measure the CsErSe₂ transmission spectra. To highlight the field-dependent absorption modes, we normalized the spectrum at each field to the maximum intensity value and then subtracted the zero-field spectrum. The resulting normalized transmission is sensitive only to intensity changes induced by the magnetic field while eliminating contributions from field-independent vibrational absorption and instrumental artifacts.

Ac magnetic susceptibility was measured in a nesting coil set, consisting of a primary coil that generates a very small ac magnetic field and a pair of sensing coils, coaxially placed within the primary coil. The two sensing coils are counterwound and connected in series to ensure equal mutual inductance with opposite sign. This results in zero net induced voltage in a null state. Six single crystal pieces of CsErSe₂ samples were stacked together along the c axis and placed in one of the sensing coils. The change of magnetic flux contributed by the sample induces a directly proportional nonzero net voltage. We find that the background signal of coils is negligible compared to the signal generated by the presence of the samples. The coil set is mounted on a dilution refrigerator such that the dc magnetic field generated by the superconducting magnet is aligned to the c axis.

Neutron diffraction spectra were measured at the HB2A diffractometer at the Oak Ridge National Laboratory's High Flux Isotope Reactor [17]. We measured 3 g of loose powder of CsErSe₂ (the same samples as used in Ref. [14]) in a copper can mounted in a dilution refrigerator at temperatures $T = 0.5$ K and $T = 0.05$ K using neutrons with $\lambda = 2.41$ Å. The magnetic scattering was isolated by subtracting the high-temperature data from the low-temperature data. We performed Reitveld refinements to the neutron data using the Fullprof software package [18].

Phonon modes in CsErSe₂ are calculated from density functional theory (DFT), which is performed using the Vienna *ab Initio* Simulation Package (VASP) version 5.4.4 [19–22] using the projector augmented wave (PAW) [23,24] method and the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [25]. A Γ -centered Monkhorst-Pack [26] k -mesh is used for all calculations. Phonons are calculated using the Phonopy software [27,28].

III. RESULTS

A. Magneto-optical spectroscopy and the determination of $\mathcal{H}_{\text{CEF}}$

The FIR and Raman spectra observed under a magnetic field are shown in Figs. 2(a)–2(c): Panels (a) and (b) display colormaps of the field dependence of the normalized FIR absorption measured at $T = 5$ K, with applied field (B) up to 17.5 T along the crystalline b axis ($\mathbf{B} \parallel b$) and c axis, respectively. Panel (c) displays the Raman shift at 5 K as well, as a function of field up to 14 T along the c axis ($\mathbf{B} \parallel c$), the intensity of which is normalized with the maximum value. The red dashed lines in each figure indicate the calculated peak positions of the transitions from the ground state. Strong

single-ion magnetoanisotropy of CsErSe₂ for the different field orientations is self-evident from the distinctly anisotropic field dependence of the CEF levels.

We note that the FIR spectra of CsErSe₂ suffer from complete absorption in $130 \text{ cm}^{-1} \leq E \leq 210 \text{ cm}^{-1}$, and as such, above 210 cm^{-1} , few discernible field-dependent features were observed. The Raman shift data display three field-dependent spectra in $E > 120 \text{ cm}^{-1}$ (Fig. 7), however, with far weaker intensity. It turns out that three highest-energy doublets exhibit an exceptionally small transition probability that cannot be discernible. We will discuss further in Sec. IV. Hence, our analysis is focused on only field-dependent spectra originating from CEF levels below $E < 120 \text{ cm}^{-1}$.

Despite the fundamentally different nature of FIR and Raman in $\mathbf{B} \parallel c$, the close correspondence between panels (b) and (c) is remarkable. Apart from the field-independent peak at $E \approx 49 \text{ cm}^{-1}$ in panel (c), the two techniques clearly probe the same transitions between CEF energy levels. The field-independent line at 49 cm^{-1} is identified as Raman-active E_{2g}^1 phonon from the phonon bands calculation as shown in Table II.

FIR and Raman shift measurements reveal the transitions from the lowest states to the Zeeman split excited states of the Er^{3+} ion. In order to describe the intricate field dependencies shown above, we consider the three contributions in our Hamiltonian as expressed in Eq. (1). $\mathcal{H}_{\text{CEF}}$ describes the single-ion energy scales in terms of the linear combination of the six Stevens operators allowed by Er^{3+} 's site symmetry, $\mathcal{H}_{\text{CEF}} = B_2^0 \hat{O}_2^0 + B_4^0 \hat{O}_4^0 + B_4^3 \hat{O}_4^3 + B_6^0 \hat{O}_6^0 + B_6^3 \hat{O}_6^3 + B_6^6 \hat{O}_6^6$. This plays the most crucial role in determining the energy landscape of CsErSe₂: At zero field, the energy eigenstates of $\mathcal{H}_{\text{CEF}}$ split the $J = 15/2$ multiplet into eight Kramers doublets. Upon applying field, each doublet splits into two states according to the Zeeman interaction, which corresponds to the second term in Eq. (1): $\mathcal{H}_Z = -\mu_B g_J \mathbf{B} \cdot \sum_i \hat{\mathbf{J}}_i$, where $\hat{\mathbf{J}}$ refers to the total angular momentum operators and Landé g factor, $g_J = 6/5$, is used for Er^{3+} . Beyond the single-ion picture, the triangular-lattice symmetries of Er^{3+} sites permit a nearest-neighbor exchange interaction with XXZ symmetry [29]. Since the pseudodipolar anisotropic exchange terms vanish within a Weiss mean-field treatment [30], we confine our analysis to the minimal XXZ $\mathcal{H}_{\text{xxz}}$, featuring nearest-neighbor $J = 15/2$ moments coupled by $\mathcal{J}_\perp$ and $\mathcal{J}_z$ for transverse and longitudinal J components: $\mathcal{H}_{\text{xxz}} = \sum_{\langle i,j \rangle} [\mathcal{J}_\perp (\hat{J}_{i,x} \hat{J}_{j,x} + \hat{J}_{i,y} \hat{J}_{j,y}) + \mathcal{J}_z \hat{J}_{i,z} \hat{J}_{j,z}]$, where the indices $\langle i, j \rangle$ refer to nearest-neighbor lattice sites and x, y, z label the components of $J = 15/2$ moment on site i [31]. In addition, there is a nonzero c -axis exchange that produces the long-range order shown in Fig. 4, which is not included here.

To specify $\mathcal{H}_{\text{tot}}$ in Eq. (1), a total of eight parameters—six coefficients of Stevens' operators, B_n^m 's in $\mathcal{H}_{\text{CEF}}$, and two exchange energies in $\mathcal{H}_{\text{xxz}}$ —are required. At a given high-symmetry field orientation, either $\mathbf{B} \parallel c$ or $\mathbf{B} \parallel b$, there are seven free parameters that must be determined. We fit the field-dependent spectra in the Raman shift data [Fig. 2(c)] directly to Eq. (1), leveraging the field dependence and obtain seven parameters simultaneously for $\mathbf{B} \parallel c$. The fittings were performed using the *scipy* optimize package [32] and PyCrystalField [33].

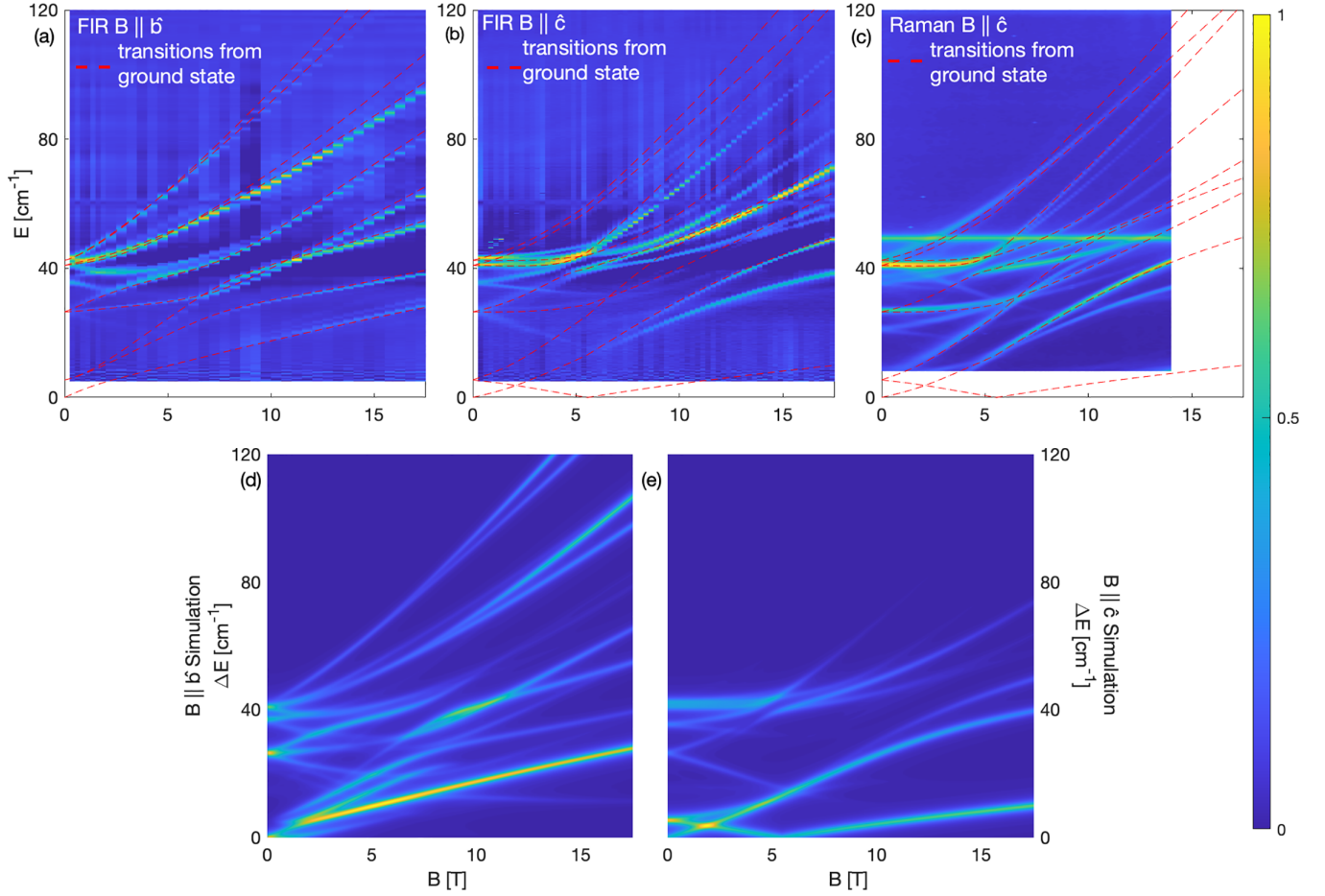


FIG. 2. Normalized FIR absorption spectral intensity at $T = 5$ K as functions of incoming light energy E and the applied magnetic field B for the two high-symmetry field orientations, $\mathbf{B} \parallel b$ [panel (a)] and $\mathbf{B} \parallel c$ [panel (b)], respectively. (c) Normalized peak intensity of Raman shift, denoted as E in $\mathbf{B} \parallel c$ at $T = 5$ K. Red dashed lines in panels (a)–(c) show the field-dependent transition from the CEF ground state as a function of B calculated from our analysis for $E < 120$ cm^{-1} (see text). Simulated transition intensity spectrum for $E < 120$ cm^{-1} for (d) $\mathbf{B} \parallel b$ and (e) $\mathbf{B} \parallel c$.

The fitting parameters obtained are listed in Table I. They agree well with values obtained using an entirely independent experimental method—zero-field inelastic neutron spectroscopy and low-field magnetization measurements [14]. This agreement highlights the accuracy of our CEF parameter determination, which is essential for predicting magnetic properties governed by single-ion physics in rare-earth compounds.

Figures 2(d) and 2(e) display the calculated normalized transition amplitudes for $\mathbf{B} \parallel b$ and $\mathbf{B} \parallel c$, respectively. They are calculated by Fermi's golden rules using the results of

solving the eigenequation for Eq. (1) with the parameters in Table I. The majority of the field-dependent spectral lines observed experimentally in Figs. 2(a)–2(c) are captured in Figs. 2(d) and 2(e).

In particular, the red lines in panels (a)–(c) represent the peak positions of transitions from only the ground state to higher excited states, calculated from our model Hamiltonian. In $\mathbf{B} \parallel c$ geometry, the calculations reveal multiple level crossings, including one involving the ground state. However, the direct experimental observation of the crossing on the ground state is precluded by the low-energy cutoff of our spectroscopic techniques.

The energy gap between the two lowest doublets at zero field is approximately 5.3 cm^{-1} (≈ 0.67 meV), corresponding to a temperature scale about 8 K. Thus, at the nominal temperature of the measurement $T = 5$ K, there can be thermal populations above the ground state, which allows finite transition amplitudes to higher-energy states [Fig. 1(a)]. We will discuss this further in Sec. IV. We also note that, as seen in Table I, the exchange energies in CsErSe₂ are on the order of 10^{-3} meV or less, far smaller than both the CEF interaction and the Zeeman splitting, typical for f -electron systems. Consequently, the observed magnetic properties, including

TABLE I. The values of the coefficients of Stevens' operators of CsErSe₂ obtained from fits to Raman shift in Fig. 2(c). Units of all shown quantities are meV.

meV		meV	
B_2^0	$-3.721(1) \times 10^{-2}$	B_4^0	$-3.880(1) \times 10^{-4}$
B_4^3	$-1.407(1) \times 10^{-2}$	B_6^0	$+3.187(1) \times 10^{-6}$
B_6^3	$-3.593(1) \times 10^{-6}$	B_6^6	$+3.491(1) \times 10^{-5}$
$\mathcal{J}_z$	$-2.64(1) \times 10^{-3}$	$\mathcal{J}_\perp$	$-0.53(1) \times 10^{-3}$

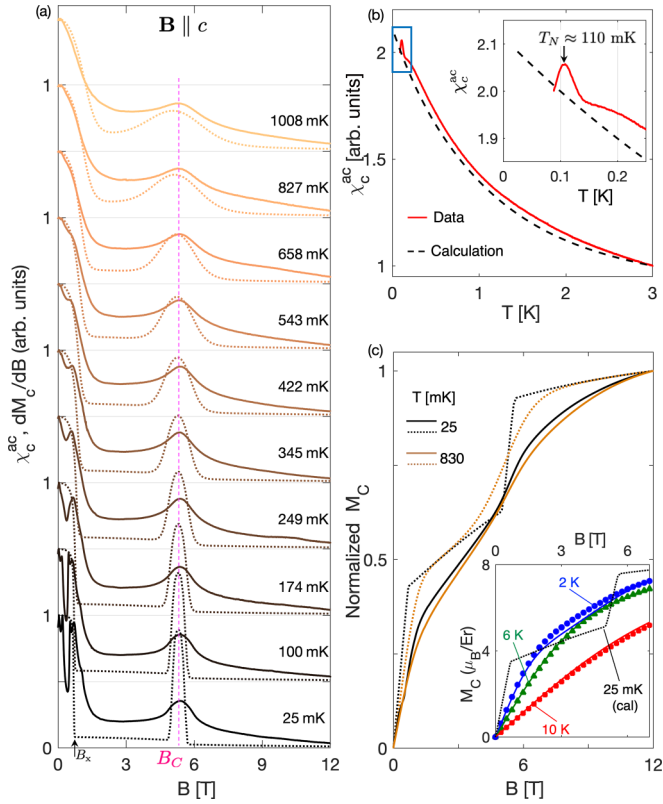


FIG. 3. (a) Measured susceptibility, χ_c^{ac} , is shown as a function of field at various temperatures in solid lines. Dotted lines display calculated χ_c^{ac} . Offset added for clarity. The vertical broken line indicates $B_C \approx 5.4$, where the first excited state crosses the ground state, while a black arrow marks B_x , where the first and second excited states cross. (b) $\chi_c^{\text{ac}}(T)$ measured at $B = 0.05$ T is shown as a solid red line, with the corresponding calculation shown as a black dashed line. A peak of χ_c^{ac} at $T = 110$ mK indicates a long-range order, but is not captured in the calculation. (c) Calculated $M_c(B)$ (dotted) and normalized magnetization $M_c(B)$ obtained from numerical integration of $\chi_c^{\text{ac}}(B)$ (solid) at 25 and 830 mK. All curves normalized at $B = 12$ T for comparison. Inset: Experimentally measured magnetization M_c (symbols) and calculated magnetization (solid lines) at 2 K (blue), 6 K (green), and 10 K (red) in solid lines. Calculated $M_c(B)$ at $T = 25$ mK is shown as a dotted line for comparison.

conspicuous magnetic anisotropy, are governed by the single-ion ground-state properties.

B. Field dependence of ac magnetic susceptibility

The most striking feature revealed by our model Hamiltonian is the ground-state inversion at $B_C \approx 5$ T [Figs. 2(c) and 2(e)], indicating a field-induced first-order phase transition. To examine the corresponding macroscopic magnetic properties, we present the ac magnetic susceptibility (χ_c^{ac}) measurements as a function of temperature and field in $\mathbf{B} \parallel c$.

In Figs. 3(a) and 3(b), we show $\chi_c^{\text{ac}}(B)$ at various temperatures as indicated and $\chi_c^{\text{ac}}(T)$ measured at the zero-field limit (at 0.05 T), respectively. $\chi_c^{\text{ac}}(T)$ exhibits a sharp peak at $T_N \approx 110$ mK (solid red line). This is evidence of long-range magnetic ordering arising from the interactions between magnetic ions and hence beyond the single-ion picture. The calculation

of $\chi_c^{\text{ac}}(T)$ [inset of Fig. 3(b)], therefore cannot capture this peak. In fact, such long-range order has been previously reported in a sister compound with the same structure, KErSe₂ at $T_N \approx 200$ mK, with a similar stripe antiferromagnetic order [16,34].

To probe the nature of the ordering further, we performed elastic neutron scattering at 50 mK. We will discuss this in Sec. III C.

$\chi_c^{\text{ac}}(B)$'s feature a prominent local maximum at $B_C \approx 5.4$ T, as well as a peak in the limit of $B \rightarrow 0$. We identify the former as a first-order field-induced transition due to a level crossing and subsequent ground-state inversion. This transition is an intrinsic consequence of the single-ion physics governed by $\mathcal{H}_{\text{CEF}}$ and $\mathcal{H}_Z$ in Eq. (1). Accordingly, the corresponding peak position remains at B_C , independent of temperature, as indicated by the vertical dashed line in Fig. 3(a). Its amplitude, on the other hand, decreases with increasing temperature as thermal population spreads over multiple CEF levels, yet the feature remains clearly visible up to 1.008 K.

We compare this with the calculated dM_c/dB , shown as dotted lines using the parameters in Table I, and find good agreement, except for a clear broadening. We note that the calculated values of dM_c/dB and magnetization were obtained using only the parameters listed in Table I; no additional fitting parameters were introduced.

The agreement is even more evident in Fig. 3(c), where the integration of the measured $\chi_c^{\text{ac}}(B)$ at $T = 25$ and 830 mK is plotted in comparison to calculated M_c . Upon further increasing temperature, the rapid increase in M_c near B_C disappears entirely by $T = 2$ K, which is approximately the lowest temperature accessible with a conventional magnetometer [inset of Fig. 3(c)]. We think this is likely due to the stacking faults common to layered materials [35,36] and the consequent distortions of octahedral environments. This can lead to a finite distribution of values of B_C , due to the spread of CEF parameters. This broadening can also be attributed to partial degradation common to layered materials containing alkali metal layers [Fig. 1(b)] [37].

Another unusual property in $\chi_c^{\text{ac}}(B)$ of CsErSe₂ is found near zero field, specifically $0 < B < B_x$ in both data and calculation. The conspicuous peak in $\chi_c^{\text{ac}}(B)$ in the limit of $B \rightarrow 0$ is a natural consequence of the Weiss MF approximation [38]; however, two peculiar B -dependent features overlap on this: One is the abrupt drop at B_x followed by remaining at a finite $\chi_c^{\text{ac}}(B)$ in $B_x < B < B_C$. The other is an abrupt dip centered at around 0.2 T, which completely disappears for $T > 830$ mK.

The first feature is best explained by examining the lowest two levels under field, where the Zeeman gap exhibits non-monotonic field dependence, having a maximum at B_x , the field corresponding to the level crossing between the first and second excited states [Fig. 1(a)]. Starting from zero field, the Zeeman gap initially increases with B , making thermal excitation across the gap more difficult at a fixed temperature and causing dM_c/dB to decrease rapidly. For $B > B_x$, however, the gap decreases again, so excitations become progressively easier, and the system continues to polarize until the gap closes at B_C . As a result, M_c increases smoothly with field, leading to a finite $\chi_c^{\text{ac}}(B)$ in the range of $B_x < B < B_C$.

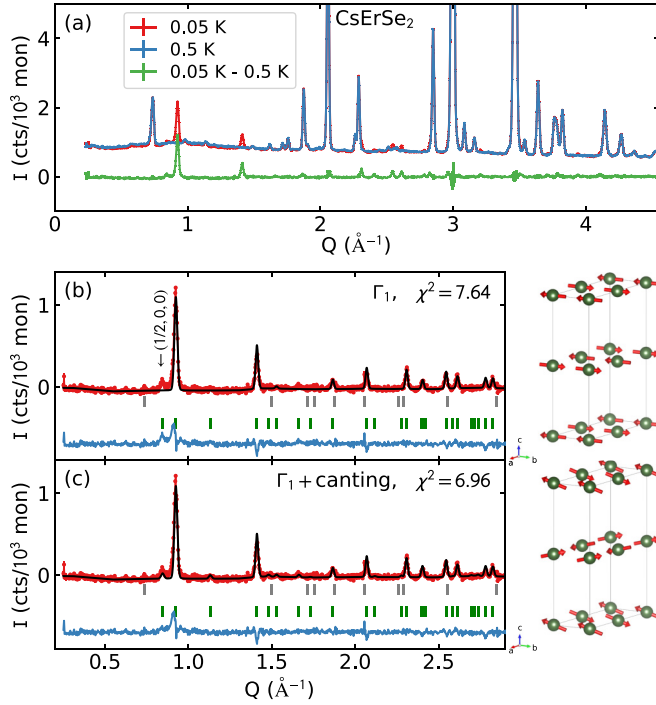


FIG. 4. (a) Neutron diffraction on CsErSe_2 , showing magnetic reflection with propagation vector $\mathbf{k} = (\frac{1}{2}, 0, 0)$ appearing at $T = 50$ mK, which is about $1/2$ of $T_N \approx 110$ mK. (b), (c) Refinements of the CsErSe_2 magnetic structure with $(\frac{1}{2}, 0, 1)\Gamma_1$ order and with an added $(\frac{1}{2}, 0, 0)$ canting along the c axis. This additional canting accounts for the $(\frac{1}{2}, 0, 0)$ Bragg peak, and noticeably lowers the reduced χ^2 . The refined moment is 71% of the theoretical maximum value from the CEF ground-state doublet $3.27(15)\mu_B$, indicating weak fluctuations.

The second feature, while we find it experimentally robust, is *not* captured by our model Hamiltonian, and at present its origin remains unclear. We speculate that this feature is associated with the onset of the spontaneous long-range order. It appears only at low temperatures in $T < 500$ mK, the same temperature range in which the predominant stripe antiferromagnetic order with wave vector $(1/2, 0, 1)$ also vanishes (see Sec. III C). This abrupt suppression of the magnetic susceptibility within the ordered phase may therefore potentially indicate a spin-flop-like transition at low fields [38] or magnetic domains at play.

C. Magnetic structure analysis with neutron scattering

The anomaly at $T = 110$ mK in $\chi_c^{\text{ac}}(T)$ [Fig. 3(b)] indicates the formation of long-range order at zero field and prompts investigation of the nature of long-range ordering in CsErSe_2 . Long-range order has been reported in similar materials, such as KErSe_2 , in both powder and single crystal samples [15,16]. This long-range order is mostly governed by interactions among pseudospins ($\mathcal{H}_{\text{XXZ}}$), of which the energy scale is two orders of magnitude smaller than CEF energy scales (Table I). To gain insight into the many-body magnetic structure, we performed elastic neutron scattering at $T = 50$ mK. We refined the magnetic structure of CsErSe_2 by performing irreducible representational analysis for the

$P6_3/mmc$ space group and the $(1/2, 0, 1)$ propagation vector. The four irreducible representations and further information are available in the Appendixes. There are many possible combinations of irreducible representations that will account for this magnetic structure, but we here show the simplest option that preserves uniform moment size: alternating canting along the c axis, shown in Fig. 4(b). These intensities match a $\mathbf{k} = (1/2, 0, 1)$ ordering wave vector (antiferromagnetic modulation along the c axis), with a very weak $\mathbf{k} = (1/2, 0, 0)$ also present, no modulation along the c axis.

We find that the ground-state magnetic order is predominantly $(1/2, 0, 1)$ stripe antiferromagnetism, which disappears by $T = 500$ mK. There is a weak $(1/2, 0, 0)$ intensity visible only at low temperature, indicating a weak canting $17.4(8)^\circ$ away from the triangular plane, corresponding to an amplitude $0.70(4)\mu_B$ along the z direction.

The sister compound KErSe_2 exhibits similar ordering, but the canting was not reported [16].

IV. DISCUSSION

We have shown that a quantitative description of the field evolution of the low-lying CEF levels, including the level crossing in the ground state, is essential for understanding the unusual properties of CsErSe_2 under a magnetic field. To our best knowledge, such level crossings involving the ground state and the corresponding field-induced transition have not been explicitly predicted from well-characterized CEF Hamiltonian, although it could be inferred from the phase diagram constructed from thermodynamic measurements [39].

This is partly attributed to the challenges in determining Stevens' coefficients in $\mathcal{H}_{\text{CEF}}$. Fits obtained from zero-field spectroscopy alone are often highly underconstrained and suffer from substantial parameter degeneracy, especially for ions with lower site symmetry [40–42]. Here, we overcome this limitation by directly exploiting the characteristic field-dependent splitting of the Kramers doublets, thereby determining the model Hamiltonian in a robust manner.

Figure 5 displays all splittings of the eight doublets of CsErSe_2 calculated with the parameters in Table I under two high-symmetry field orientations, $\mathbf{B} \parallel b$ [panel (a)] and $\mathbf{B} \parallel c$ [panel (b)]. The lowest five doublets are clustered below 40 cm^{-1} ($\approx 5 \text{ meV}$) at zero field, while the other three are clustered at around $190\text{--}200 \text{ cm}^{-1}$ ($\approx 25 \text{ meV}$).

The g -factor values are found $g_\perp = 6.55$ in $\mathbf{B} \parallel b$ and $g_z = 6.98$ in $\mathbf{B} \parallel c$ calculated from $g_{z(\perp)} = 2g_J \langle 0_\pm | \hat{J}_{z(y)} | 0_\pm(\mp) \rangle$, where $|0_\pm\rangle$ are two doubly degenerate ground states in the $B \rightarrow 0$ limit. Despite the similar g -factor values, the strong magnetic anisotropy of CsErSe_2 is manifested by the unique field evolution of the Zeeman splitting of each doublet.

In particular, the key distinction between Figs. 5(a) and 5(b) lies in the ground-state evolution under applied field. For $\mathbf{B} \parallel b$, no level crossings occur among the three lowest-energy states, whereas for $\mathbf{B} \parallel c$, level crossings appear in both the ground and first excited states. These crossings are directly responsible for the unusual magnetic behavior discussed in the previous sections. Consequently, the calculated field dependences of the magnetization, M_b and M_c for $\mathbf{B} \parallel b$ and $\mathbf{B} \parallel c$, respectively, differ dramatically, as shown in panels (c) and (d).

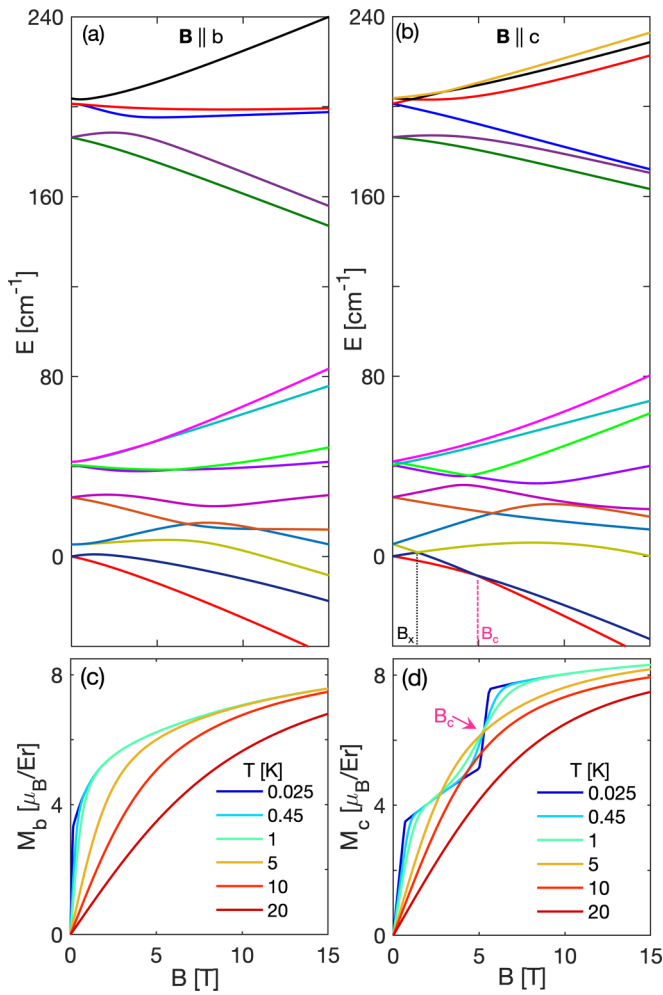


FIG. 5. Field-dependent splittings of CEF levels for the two high-symmetry axes, $\mathbf{B} \parallel b$ [panel (a)] and $\mathbf{B} \parallel c$ [panel (b)]. The energy levels are calculated from Eq. (1) using the parameters shown in Table I. Panels (c) and (d) display the calculated magnetization, M_b and M_c , as a function of applied field along the two corresponding orientations at various temperatures as shown.

Another feature that can be revealed only by a reliable determination of the CEF parameters is the quadratic low-field field splitting of the ground state for $\mathbf{B} \parallel b$ as $B \rightarrow 0$, in contrast to the strictly linear splitting for $\mathbf{B} \parallel c$. This quadratic term significantly modifies the low-temperature magnetic susceptibility, especially for temperatures below the zero-field CEF gap between the two lowest doublets. As a result, the susceptibility deviates from the effective pseudospin-1/2 description [12], which could be misinterpreted as other than a single-ion origin without an accurate understanding of the CEF spectra at finite field.

By comparing Figs. 2(a)–2(c) with Figs. 5(a) and 5(b), two key features of the observed CEF spectra become apparent. First, several spectral lines in Figs. 2(a)–2(c) cannot be explained solely by transitions originating from the ground state. Second, the three doublets with energies above 180 cm^{-1} are not present in the measured spectra.

This is due to transitions from thermally populated low-lying excited states. The zero-field CEF gap between the two

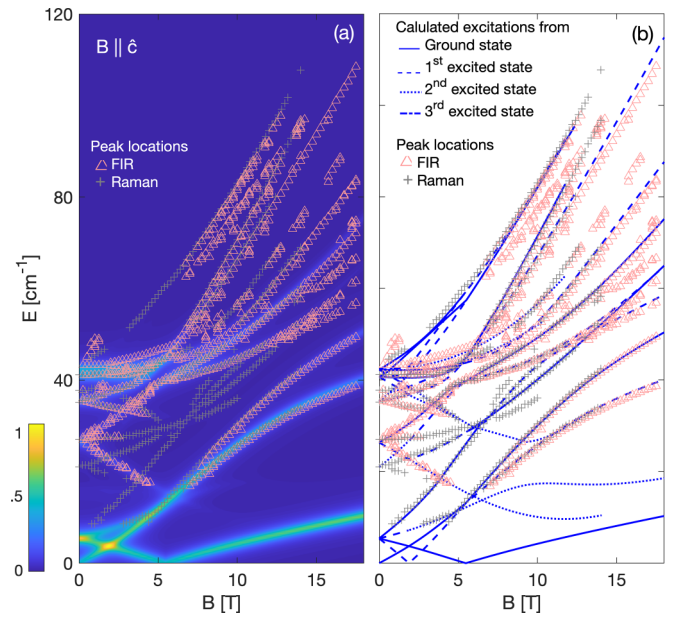


FIG. 6. (a) Simulated field-dependent spectrum at $T = 10$ K in $\mathbf{B} \parallel c$ is shown as a normalized color map. Triangles (FIR) and crosses (Raman) correspond to the peak positions from the data, Figs. 2(b) and 2(c), respectively. (b) Calculated transitions with probability $\rho > 0.001$ are shown. Solid, long-dashed, dotted, and long-short-dashed lines represent the transitions from ground, first, second, and third excited states, respectively. In $E > 160$ cm^{-1} , only two transitions with $\rho \geq 0.001$ emerge, which are not identifiable in the data [see Fig. 7(a)].

lowest doublets is found to be 0.67 meV (≈ 12 K). With an applied field, this becomes even smaller due to the level crossing in $\mathbf{B} \parallel c$, causing a finite thermal population in the excited states. Figure 6(a) shows a simulated spectrum calculated using the transition probabilities from Fermi's golden rule. The experimentally observed peak positions extracted from the FIR (triangles) and Raman shift (crosses) measurements in Figs. 2(b) and 2(c) are overlaid as symbols for comparison. We find that almost all observed peaks are well captured by transitions with a probability $\rho_c = 0.001$. To make a more explicit comparison, in Fig. 6(b), we plot all transitions from the three lowest states that have a probability higher than ρ_c as a function of field with the same set of peak position data in panel (a), which exhibits excellent agreement. We also note that the discontinuities and abrupt changes in curvature of the transition lines originate from level crossings, where the corresponding eigenstates switch abruptly at the crossings. These changes in turn produce discontinuous variations in the transition probabilities.

We find that most transitions with $E > 160$ cm^{-1} have calculated probabilities below $\rho_c = 0.001$, and are therefore not expected to be experimentally detectable. In Fig. 7(a), we show Raman shift spectra in extended energy up to 540 cm^{-1} under applied field as an intensity colormap. The red lines indicate transitions with probability $\rho_c \geq 0.001$, identically to Fig. 6(b). Panel (b) displays the intensities as a function of E at every 2 T increment of field. Each curve is offset by a constant for clarity.

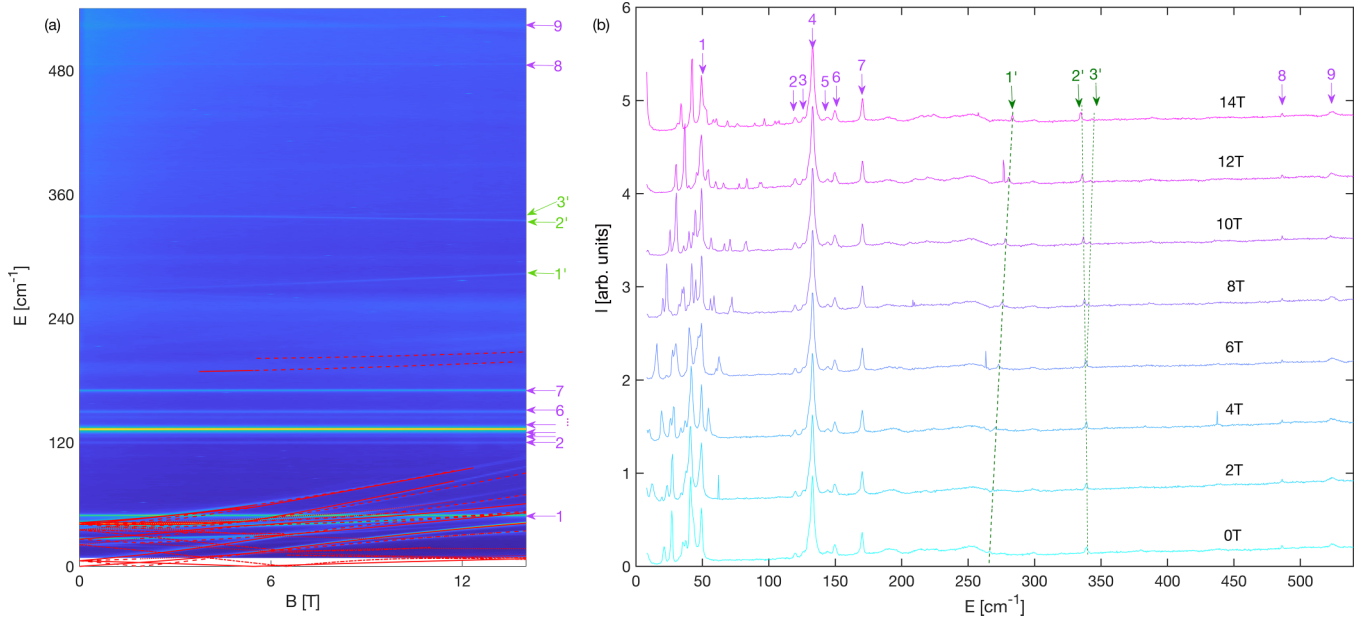


FIG. 7. (a) Raman shift (E) as a function of field in the larger range of Raman shift up to $E = 540 \text{ cm}^{-1}$. Red lines indicate transitions with probability $\rho > 10^{-3}$, including transitions from the ground, first, second, and third excited states, the same as Fig. 6(b). The detailed features in $E < 120 \text{ cm}^{-1}$ remain identical to Fig. 6(b). Green arrows mark field-dependent peaks, whereas purple arrows indicate field-independent peaks assigned to the phonon modes in Table II. Note the intensities of any field-dependent spectra lying above 120 cm^{-1} are significantly smaller than those below. (b) The Raman spectra intensity is shown as a function of energy under magnetic field, varying from 0 T (bottom) to 14 T (top) by 2 T increment. The data are plotted with an offset for clarity. Purple and green arrows mark the same locations as panel (a).

The green arrows of $1'$, $2'$, and $3'$ mark only field-dependent features observed in $E > 120 \text{ cm}^{-1}$. We find that none of these peak locations and intensities are matched by the transition probability as low as 10^{-3} , and their origins remain unclear. Based on studies on free Er^{3+} 's energy spectra [43,44], we speculate that these are related by-products of higher-order Raman processes, activated by resonance with the incoming laser energy (532 nm). Both energies of the transitions for ${}^2\text{H}_{11/2} \rightarrow {}^4\text{I}_{15/2}$ (533 nm) and ${}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{15/2}$ (547 nm) lie very close to the incoming laser energy and their intermediate transitions may cause the observed weak field dependence. We exclude contributions from the next $J = 13/2$ manifold, as it has been reported to lie

approximately 6000 cm^{-1} ($\approx 744 \text{ meV}$) above the $J = 15/2$ manifold [44].

The purple arrows 1–9 in Fig. 7 indicate field-independent peaks, the locations of which are listed in Table II. These modes show reasonable agreement with Γ point energies for phonon modes calculated from density functional theory. Details of the calculations are presented in the Appendixes.

Raman spectroscopy and far-infrared magneto-optical spectroscopy under intense magnetic fields offer a powerful and versatile approach to determining CEF physics in rare-earth materials. While these techniques have long been used to probe magnetic couplings and magnonic behavior in magnetic systems [45–49], here they are used to directly resolve the field-dependent CEF excitations in a $4f$ compound. This enables a precise determination of the CEF Hamiltonian in CsErSe_2 , while simultaneously revealing field-induced level crossings and associated magnetic properties.

Furthermore, these methods are particularly effective for probing magnetoelastic interactions enhanced by strong spin-orbit coupling. A prominent example is the formation of vibronic bound states through hybridization between phonons and CEF levels [50,51]. In such cases, the resulting hybridized modes inherit the field dependence of the underlying CEF levels [45,47]. Consequently, new quasiparticles from this hybridization exhibit field-dependent spectra and dispersion, which turns out to be a natural microscopic mechanism for unusual field dependence of thermal transport phenomena [52].

More broadly, our results demonstrate that field-dependent spectroscopy is not only a tool for determining CEF Hamiltonians, but also a powerful probe of the coupled spin-lattice

TABLE II. The field-independent peaks indicated with the numbered purple arrows [Figs. 7(a) and 7(b)] are listed here and compared to the phonon DFT calculation. The predicted locations of the symmetric phonon peaks are shown in the parentheses in the unit of cm^{-1} .

	Observed (cm^{-1})	Assignment (calculated)
1	49.3(1)	E_{2g}^1 (46.1)
2	119.6(1)	Unknown
3	125.9(1)	E_{1g} (125.9)
4	132.7(1)	E_{2g}^2 (127.3)
5	142.8(1)	Unknown
6	149.6(1)	$3E_{2g}^1$
7	170.3(1)	A_{1g} (164.8)
8	486.3(1)	Unknown
9	523.8(1)	Unknown

physics that drives complex and unexpected magnetic behavior in rare-earth magnets.

V. SUMMARY

The main result of this work is the identification of a field-induced phase transition in CsErSe_2 and the associated unusual magnetic behavior emergent under field in $\mathbf{B} \parallel c$. This transition originates from a level crossing and subsequent ground-state inversion and occurs at $B_C \approx 5.4$ T. Such a transition is expected to produce a steplike change in magnetization and corresponding peak in magnetic susceptibility. Consistent with this expectation, our ac magnetic susceptibility measurements revealed a peak at B_C , providing experimental evidence for the transition. We further show that the nonmonotonic evolution of the Zeeman gap below B_C is responsible for characteristic magnetic behavior, including the plateaulike field dependence of the magnetization.

This discovery was enabled by the accurate determination of our model Hamiltonian. As in typical $4f$ electron systems, the magnetic properties of CsErSe_2 are governed by single-ion physics; hence, reliably resolving the CEF scheme is essential for predicting the magnetic response. By directly tracking the magnetic-field dependence of the CEF level splittings using magneto-optical spectroscopy, we obtain a robust determination of the CEF Hamiltonian.

We also find that CsErSe_2 undergoes spontaneous magnetic ordering at $T_N = 110$ mK, which predominantly exhibits a stripe antiferromagnetic ordering, the characteristics of which are partly attributed to dominant contributions from $\mathcal{H}_{\text{CEF}}$.

The key findings of this work provide a framework for understanding unusual magnetic properties that arise from complex single-ion physics, rather than from other mechanisms such as interactions between magnetic ions. This distinction is essential for disentangling single-ion effects from collective behavior and, ultimately, for more clearly identifying the signatures of exotic spin states such as quantum spin liquids.

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DATA AVAILABILITY

The data that support the findings of this article are openly available at Zenodo [53].

APPENDIX A: DENSITY FUNCTIONAL THEORY CALCULATIONS FOR CsErSe_2

1. Methods

All DFT calculations are performed using the VASP version 5.4.4 [19–22] using the PAW [23,24] method and the PBE exchange-correlation functional [25]. A Γ -centered Monkhorst-Pack [26] k -mesh is used for all calculations. Phonons are calculated using the Phonopy software [27,28].

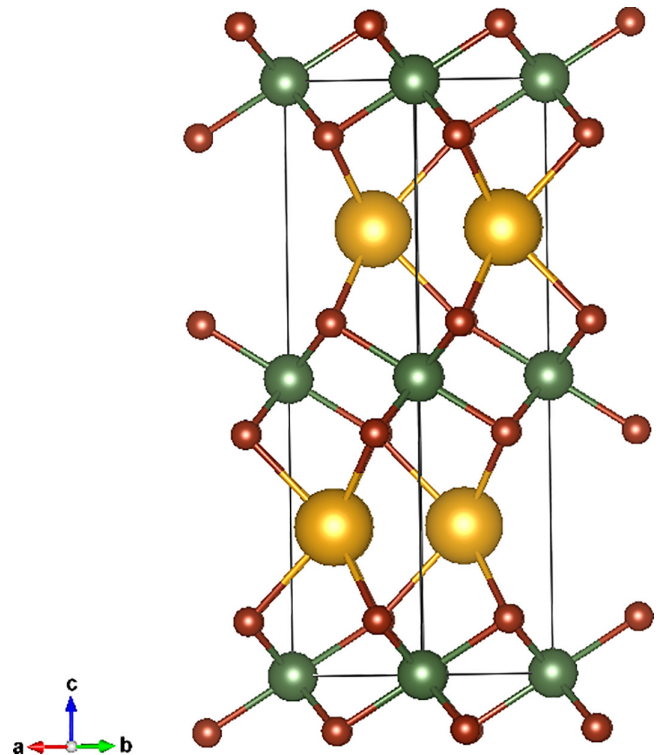


FIG. 8. Crystal structure of CsErSe_2 in the primitive cell, space group $P6_3/mmc$ (No. 194). Color scheme: gold = Cs, green = Er, and red = Se.

2. Crystal structure details

We perform DFT calculations of CsErSe₂, with space group $P6_3/mmc$ (No. 194) and a hexagonal primitive cell shown in Fig. 8. The lattice parameters are $a = b = 4.1799 \text{ \AA}$, $c = 16.5798 \text{ \AA}$, $\alpha = \beta = 90^\circ$, and $\gamma = 120^\circ$. The Wyckoff positions for Cs, Er, and Se are $2c$, $2a$, and $4f$, respectively. We use supercells of this primitive cell for phonon calculations.

3. Total energy convergence

The PAW potentials used for all calculations (Cs_sv_GW, Er_3, and Se_GW) have ENMAX values 148.575, 116.278, and 158.666 eV, respectively, so that 200 eV should be the minimum plane wave energy cutoff (E_{cut}) used. We test convergence of the total energy with respect to E_{cut} and k -point mesh for the eight-atom primitive cell, shown in Fig. 9. The results indicate that to converge the total energy to within 0.5 meV/atom, $E_{\text{cut}} = 400 \text{ eV}$ and a linear k -mesh density of $30 \text{ \AA}^{-1}$ (corresponding to an $8 \times 8 \times 2$ mesh) are needed. Note that $\Delta E_{\text{tot}} = E_{\text{tot}}(E_{\text{cut}}) - E_{\text{tot}}(1000 \text{ eV})$ and $\Delta E_{\text{tot}} = E_{\text{tot}}(N_k) - E_{\text{tot}}(N_k = 60)$, respectively.

4. Electronic density of states

The electronic density of states (DOS) is shown in Fig. 10. The top panel shows the atom-projected DOS, while the bottom panel shows the l -projected DOS. Note that because we use the Er_3 PAW potential, in which the Er f electrons are placed in the core, we do not have f character in the valence DOS. Also note that spin-orbit coupling is not included, which would further split the Er and Cs p states.

5. Phonon convergence studies

To test convergence of phonons, we explore the role of supercell size using the converged values of $E_{\text{cut}} = 500 \text{ eV}$ and $N_k = 30 \text{ \AA}^{-1}$. In Fig. 11, we show phonon dispersions for $3 \times 3 \times 1$ (72-atom) and $4 \times 4 \times 1$ (128-atom) supercells. The close agreement shows that either supercell size could be used, though we use the $4 \times 4 \times 1$ cell for all final results. Note

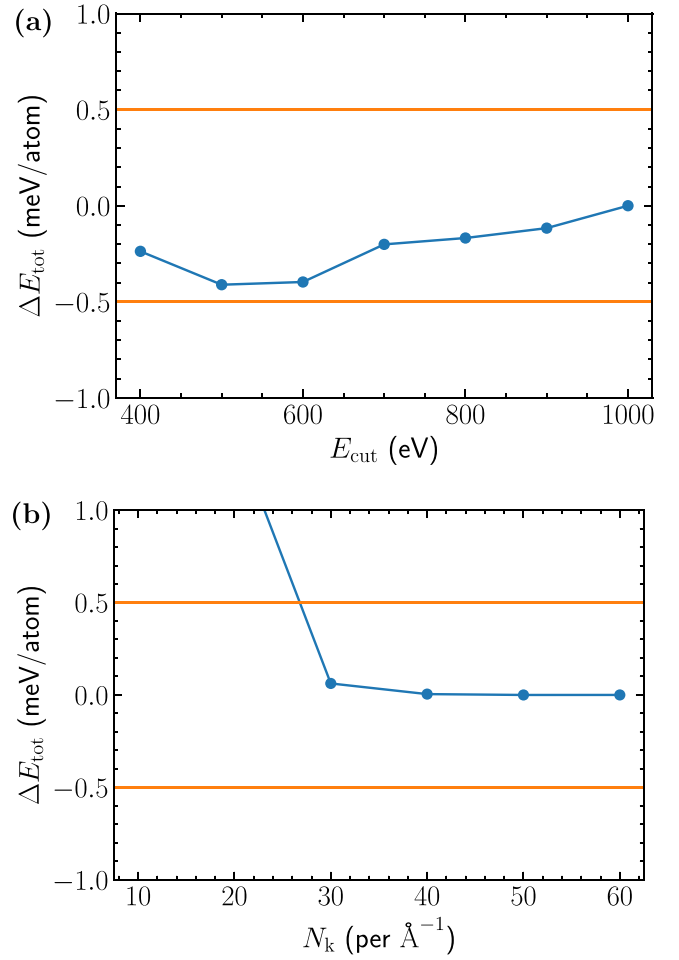


FIG. 9. Total energy convergence with respect to (a) plane-wave energy cutoff and (b) k -mesh size. Orange lines indicate ± 0.5 meV/atom with respect to the converged values.

that longitudinal optical-transverse optical (LO-TO) splitting is included in these calculations through the nonanalytic term correction as implemented in Phonopy.

TABLE III. Γ point mode frequencies in cm^{-1} .

Mode No.	Mull. symb.	Rel. lat ν (cm^{-1})	Expt. lat ν (cm^{-1})	IR active	Raman active
1	N/A	0.00	0.00	—	—
4–5	E_{2u}	30.50	30.30	—	—
6–7	E_{2g}	47.12	46.72	n	y
8–9	E_{1u}	56.64	56.41	y	n
10	B_{2u}	59.11	59.01	—	—
11	B_{1g}	82.38	82.25	—	—
12	A_{2u}	91.84	91.34	y	n
13–14	E_{2u}	117.28	118.15	—	—
15–16	E_{1u}	118.48	119.25	y	n
17–18	E_{1g}	125.27	125.91	n	y
19–20	E_{2g}	126.64	127.17	n	y
21	A_{1g}	164.54	164.70	n	y
22	A_{2u}	168.40	168.56	y	n
23	B_{1g}	170.50	170.46	—	—
24	B_{2u}	192.31	192.37	—	—

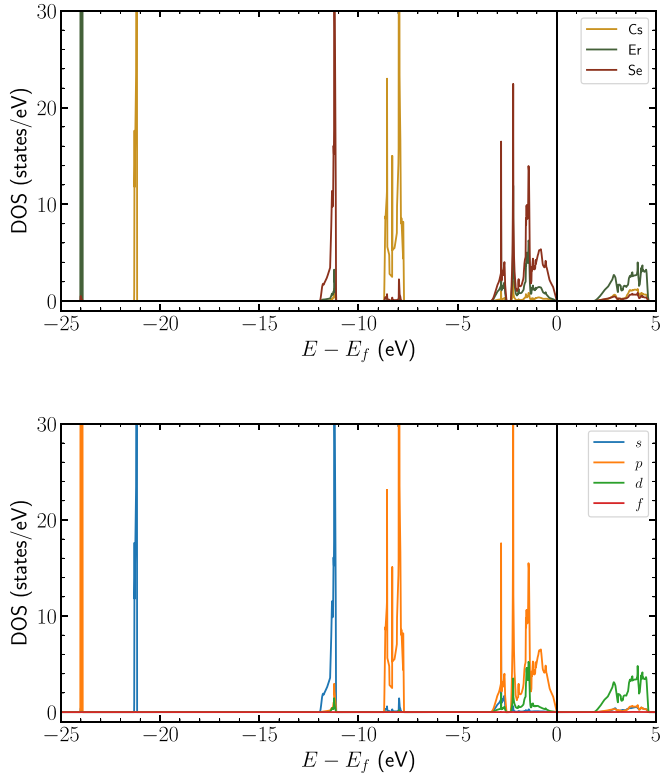


FIG. 10. Electronic density of states for CsErSe₂, projected onto atom types (top) and l quantum number (bottom).

6. Phonon density of states

The atom-projected phonon density of states for the $4 \times 4 \times 1$ (128-atom) supercell is shown in Fig. 12. Cs and Er, due to their higher mass, dominate the lower-frequency spectrum, while lighter Se character dominates the higher-frequency spectrum.

7. Γ point frequencies

In Table III, we report the Γ point mode frequencies along with their corresponding Mulliken symbols, in both THz and cm^{-1} , for the $4 \times 4 \times 1$ supercell, and whether each mode is IR or Raman active. Note that LO-TO splitting is not included. In each table, we report calculated frequencies for the experimentally determined atom positions and DFT-relaxed (with fixed lattice constants) atom positions, which are in very close agreement.

TABLE IV. Basis vectors and irreducible representations (“irreps”) for magnetic Er order with $k = (1/2, 0, 1)$ order. Irreps Γ_1 and Γ_3 have only one basis vector, while irreps Γ_2 and Γ_4 have two. Images of the best fits are provided in Fig. 14.

Irrep	Γ_1	Γ_2	Γ_3	Γ_4	
Basis vector	ψ_1	ψ_1	ψ_1	ψ_1	ψ_2
Site 1 (0, 0, 0)	(0, -1, 0)	(1, 1/2, 0)	(0, -1, 0)	(1, 1/2, 0)	(0, 0, 1)
Site 2 (0, 0, 1/2)	(0, 1, 0)	(-1, -1/2, 0)	(0, -1, 0)	(1, 1/2, 0)	(0, 0, -1)

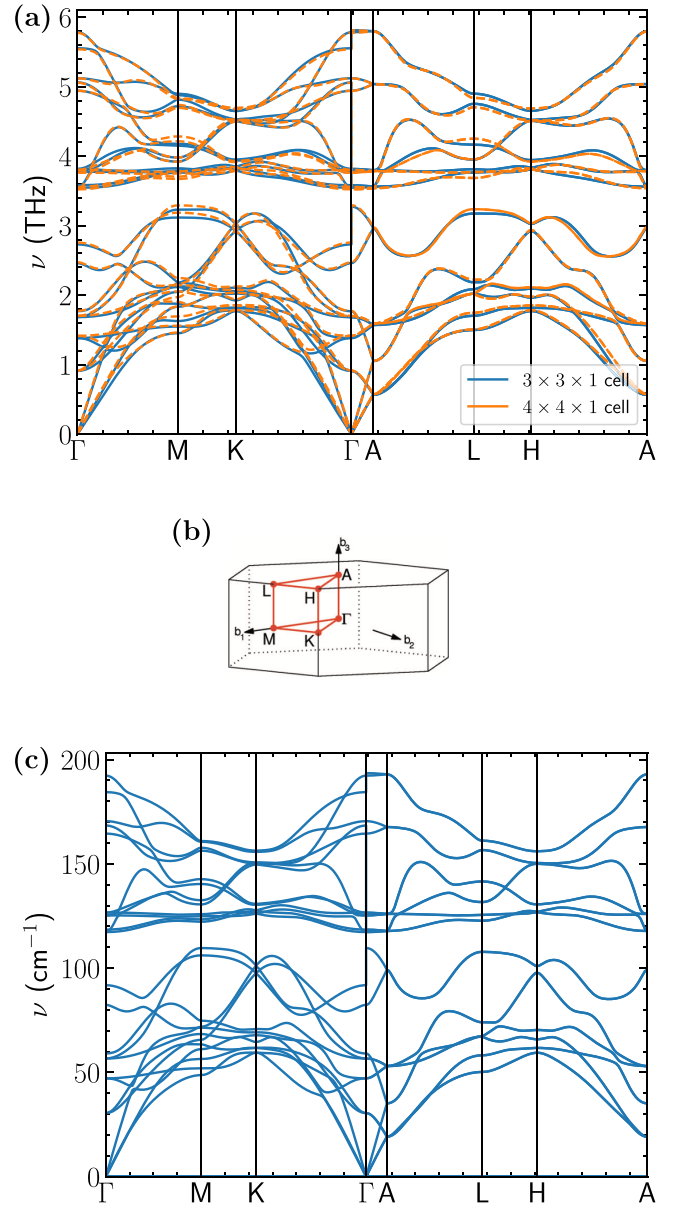


FIG. 11. (a) Convergence of phonons with respect to cell size, using a $3 \times 3 \times 1$ (72-atom) and a $4 \times 4 \times 1$ (128-atom) supercell of the primitive cell. (b) The Brillouin zone path, obtained from the AFLOW convention. (c) Phonon dispersions calculated using density functional theory with a $4 \times 4 \times 1$ supercell.

APPENDIX B: NEUTRON REFINEMENTS

The nuclear structure of CsErSe₂ was refined to the $T = 0.5$ K scattering data and is shown in Fig. 13.

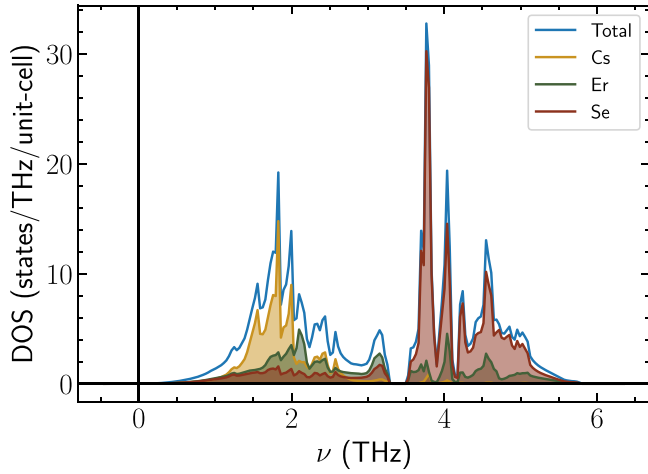


FIG. 12. Atom-projected phonon density of states for the $4 \times 4 \times 1$ (128-atom) supercell, including LO-TO splitting.

We get an excellent fit of the data in all temperature ranges.

We refined the magnetic structure of CsErSe_2 by performing irreducible representational analysis for the $P6_3/mmc$ space group and the $(1/2, 0, 1)$ propagation vector. This yielded four irreducible representations Γ_1 – Γ_4 , listed in Table IV. The magnetic structures and associated powder diffraction patterns are shown in Fig. 14. The coplanar alternating antiferromagnetic structure Γ_1 is clearly the best fit to the data. However, the fit is not perfect: There is a small but

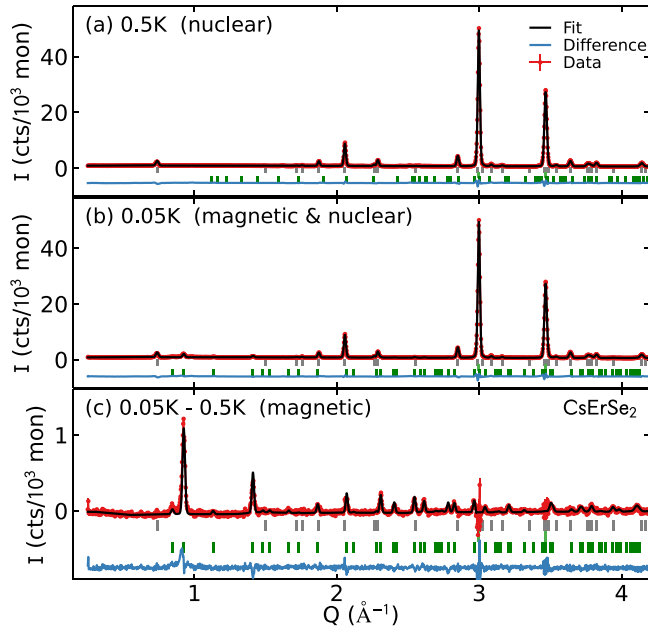


FIG. 13. Rietveld refinements of the CsErSe_2 neutron diffraction at (a) 0.5 K, (b) 0.05 K, and (c) temperature-subtracted 0.05–0.5 K to isolate the magnetic diffraction signal.

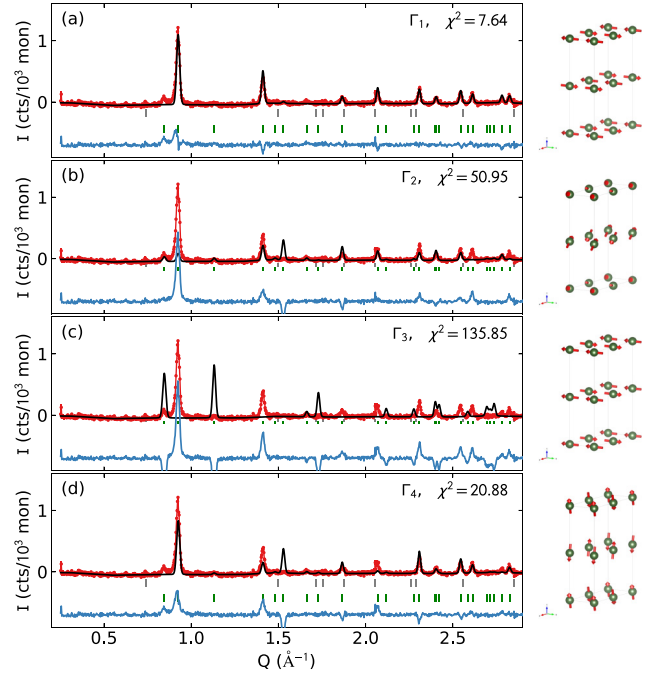


FIG. 14. Irreducible representations of CsErSe_2 magnetic structure with $(1/2, 0, 1)$ propagation vector. The best-fit diffraction pattern is on the left and the magnetic structure is on the right. The only structure that matches the data is Γ_1 , which has coplanar alternating antiferromagnetic order.

clear temperature-dependent peak at $Q = 0.863 \text{ \AA}^{-1}$, which is not accounted for by the magnetic structure, highlighted in Fig. 15 [Fig. 5(b)]. This corresponds to the $(1/2, 0, 0)$ wave vector, which indicates a weak magnetic correlation that does not alternate along the c axis. This means that an additional irreducible representation is necessary to fully account for the data, and by Landau theory indicates that the magnetic transition will be first order. There are many possible combinations of irreducible representations that will account for this

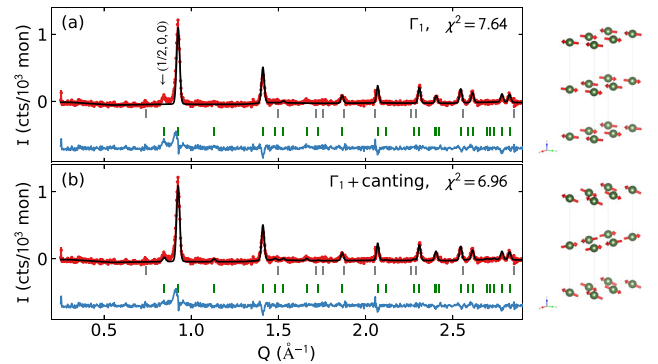


FIG. 15. Refinements of the CsErSe_2 magnetic structure with $(1/2, 0, 1)$ Γ_1 order and with an added $(1/2, 0, 0)$ canting along the c axis. This additional canting accounts for the $(1/2, 0, 0)$ Bragg peak, and noticeably lowers the reduced χ^2 .

magnetic structure, but we here show the simplest option that preserves uniform moment size: alternating canting along the c axis, shown in Fig. 15. In the language of magnetic space groups, this corresponds to P_a2_1/m (whereas the uncanted

structure corresponds to P_{6mm}). There are other possible combinations of irreps, but they generically have nonuniform moment sizes. This is a logical possibility, but would be unusual for Er sites that are symmetry equivalent.

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