

Nature of quantum criticality in the Ising ferromagnet TbV_6Sn_6

Tianxiong Han^{1,2}, R. D. McKenzie^{1,2}, Joanna Blawat³, Tyler J. Slade¹, Bing Li^{1,4}, Y. Lee¹, D. M. Pajerowski^{1,4}, John Singleton³, Paul C. Canfield^{1,2}, Liqin Ke^{1,5}, Ross McDonald³, Rebecca Flint^{1,2}, and R. J. McQueeney^{1,2,*}

¹*Ames National Laboratory, U.S. DOE, Iowa State University, Ames, Iowa 50011, USA*

²*Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA*

³*National High Magnetic Field Laboratory, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

⁴*Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

⁵*Department of Materials Science and Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*



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TbV_6Sn_6 is a topological metal where ferromagnetic Tb ions with strong uniaxial magnetic anisotropy interact with V kagome layers. Inelastic neutron scattering (INS) measurements show that the Tb ions adopt an Ising doublet ground state. Here, we consider whether a transverse magnetic field can drive TbV_6Sn_6 toward a quantum critical point, providing a rare example of transverse-field Ising criticality in a metallic compound. High-field magnetization measurements reveal a first-order-like spin-reorientation transition at 25.6 T. Our INS-based magnetic model finds that this is caused by an avoided crossing of an excited-state singlet with the ground-state doublet. Surprisingly, our model predicts that quantum critical and tricritical points are accessible within the range of experimentally determined model parameters and may be reached by varying the direction of an applied magnetic field.

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Kagome metals provide a promising platform for discovering topological and flat band physics with recent reports of charge-density wave order [1–4], superconductivity [5–7], itinerant magnetism [8–12], and Chern insulating phases [13–16]. Among the many kagome metal systems, RT_6X_6 (166) systems with R = rare-earth metal, T = V, Cr, Mn, and X = Sn, Ge are unique for their combination of magnetic R ions that interact with metallic kagome T layers [17–19].

Particularly noteworthy is TbV_6Sn_6 : While the vanadium atoms do not generate magnetic moments, the strong spin Berry curvature associated with the kagome Dirac cones leads to a significantly enhanced orbital Zeeman effect [20]. The high-spin Tb moments ($J = 6$) order ferromagnetically below $T_C = 4.4$ K and have strong Ising anisotropy inferred from low-field magnetization data [21,22] and magnetic heat capacity data, which are consistent with an isolated crystal electric field (CEF) doublet ground state [23]. Ferromagnetic (FM) order and fluctuations are known to impact transport of conduction electrons in the kagome V layers [22,23]. Thus, TbV_6Sn_6 may be a good candidate for investigating transverse-field Ising quantum criticality and its interaction with kagome electrons.

In the transverse-field Ising model (TFIM) [24], a transverse magnetic field leads to a quantum critical point (QCP) between Ising FM and quantum paramagnetism. Strong quan-

tum fluctuations near a QCP lead to highly nonclassical behavior and exotic phases of matter [25]. There are only a few FM Ising materials, like LiHoF_4 [26], that are described by an effective spin-1/2 TFIM up to the critical transverse field, and weakly correlated metallic versions have not been studied.

It is an open question whether TbV_6Sn_6 will exhibit TFIM quantum criticality where a continuous (second-order) transition is maintained as $T \rightarrow 0$. In a high-spin Ising FM, the effect of a transverse field may stabilize other CEF levels that compete with the Ising doublet, leading to a first-order metamagnetic transition [27]. Even if the doublet remains isolated, the coupling of the Tb moments to the transverse spin fluctuations of conduction electrons may eventually drive the transition first order [28,29] and generate a quantum critical wing structure, as seen in the heavy fermion URhGe [30].

In this Letter, we investigate the Ising nature of TbV_6Sn_6 using inelastic neutron scattering (INS) and high-field magnetization measurements. The INS data allow us to determine the CEF parameters, which characterize a magnetic Hamiltonian with an isolated non-Kramers quasidoublet (Ising) ground state at zero field. To search for evidence of TFIM quantum criticality, we measure the magnetization in a transverse field ($H_\perp$) and observe a first-order-like transition at $\mu_0 H_\perp^c = 25.6$ T. Our magnetic model clearly identifies that this metamagnetic (spin-reorientation) transition originates from a CEF level crossing rather than a coupling to conduction electrons.

However, our model reveals that the true nature of quantum criticality in TbV_6Sn_6 is subtle and dependent on the strength of the planar magnetic anisotropy, a parameter that is not clearly captured in our experimental data. When the planar anisotropy is small, the QCP is avoided by a first-

*Contact author: mcqueeney@ameslab.gov

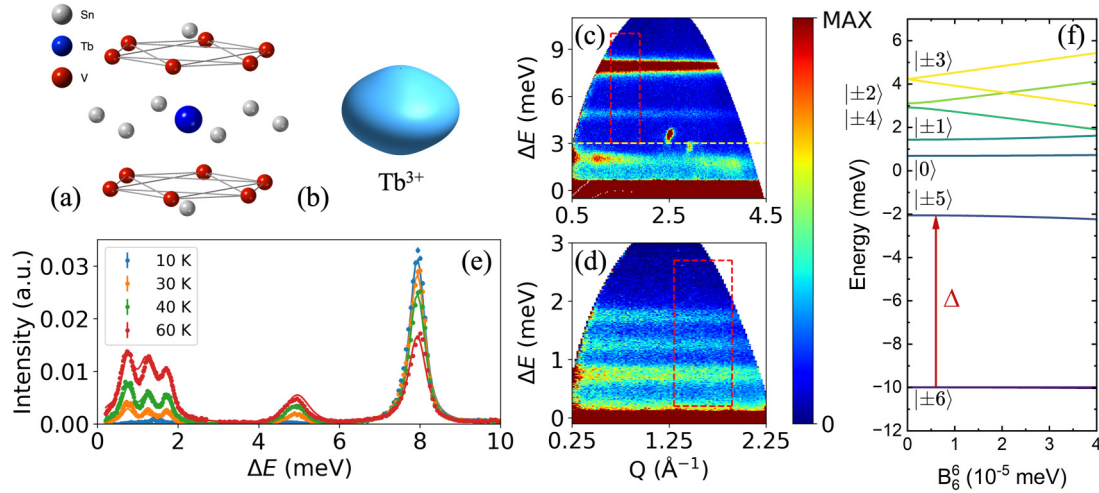


FIG. 1. Ising anisotropy in TbV_6Sn_6 . The D_{6h} point-group symmetry environment of the Tb^{3+} ion [panel (a)] and its oblate-shaped $\text{Tb}-4f$ charge density [panel (b)]. The INS data with incident neutron energy of (c) $E_i = 12$ meV and (d) $E_i = 3.32$ meV. Data near and below the yellow dashed line in panel (c) are corrupted by spurious instrumental background. (e) The energy dependence of the combined $E_i = 3.32$ and 12 meV INS spectra at various temperatures (dots). Intensities at each E_i and T are averaged over the same Q range indicated by the dashed red boxes in panels (c) and (d) and have a fitted background removed. Data for different E_i are placed on the same intensity scale using procedures described in the Supplemental Material [31]. Fits to the INS spectra from CEF calculations are plotted as solid lines in panel (e). (f) Zero-field CEF energy levels versus B_6^0 while keeping other Case I CEF parameters fixed. CEF states are pure $|\pm m\rangle$ eigenstates when $B_6^0 = 0$.

order transition exhibiting a wing structure with quantum critical end points (QCEPs) occurring in small longitudinal fields. For larger planar anisotropy, second-order QCPs nucleate along the planar easy axis and eventually expand to all transverse-field directions. In this intermediate regime, TbV_6Sn_6 is proximate to a realization of a quantum tricritical point (QTCP) that is reachable by varying the magnetic field direction.

Sample preparation. Single crystals were grown from a Sn flux similar to the previously described process [21,22]. An obstacle to growing large TbV_6Sn_6 crystals, suitable for inelastic neutron scattering, is the relatively poor solubility of V in Sn. We found that the typical reactions described in earlier reports normally yielded mixtures of small TbV_6Sn_6 crystals alongside chunks of (likely undissolved) V. We empirically found that by using the two-step process described below, larger crystals can be produced.

Elemental pieces of Tb (Ames Lab, 99.99%), V (Alfa Aesar, 99.99 %), and Sn (Alfa Aesar, 99.99%) were weighed in a 1:6:60 molar ratio and loaded into the growth side of a 5 ml alumina Canfield crucible set [32]. The crucibles were sealed under vacuum in a fused silica ampule and heated in a box furnace to 1180 °C. After holding at 1180 °C for ≈ 24 h, the samples were removed from the furnace and the liquid decanted in a metal centrifuge, upon which the liquid phase is cleanly captured in the second crucible on the top side of the crucible set. This first step allows us to cleanly separate the liquid from any undissolved V. A second sequence is then carried out, with the captured decant used in the “growth” side of another crucible set. The second crucible set was sealed under vacuum as before and warmed in a box furnace to 1195 °C. After holding at 1195 °C for 6 h, the furnace was cooled to 775 °C, upon which the ampule was removed from the furnace and the excess liquid decanted. Once cool, the ampules and crucibles were opened

to reveal relatively large (5–10 mg), single-phase, crystals of TbV_6Sn_6 .

Neutron scattering data. INS measurements on a polycrystalline sample of TbV_6Sn_6 were performed on the Cold Neutron Chopper Spectrometer at the Spallation Neutron Source at Oak Ridge National Laboratory using incident neutron energies of $E_i = 3.32$ and 12 meV. A polycrystalline sample for INS measurements was prepared by grinding the flux-grown single crystals and sieving with a diameter of 32 μm to obtain particles of uniform size. A sample weighing 1.2 g was sealed in the aluminum can filled with helium gas and mounted to a closed-cycle refrigerator for studies down to $T = 1.7$ K. The measured INS intensity is proportional to the spin-spin correlation function $S(Q, \omega)$, where Q is the momentum transfer, and $\Delta E = \hbar\omega$ is the energy transfer. The data are histogrammed with bin sizes of $\delta E = 0.06$ meV and $\delta Q = 0.02$ $\text{\AA}^{-1}$ using DAVE software [33].

Figures 1(c) and 1(d) show the INS data at $T = 30$ K, well above $T_C = 4.4$ K, with $E_i = 12$ and 3.32 meV, respectively. These data show multiple nondispersive excitations between CEF levels of the Tb^{3+} ion. The Q -averaged intensities are plotted versus energy in Fig. 1(e) for $T = 10, 30, 40$, and 60 K. The main transition out of the ground state appears at $\Delta E = 7.9$ meV at $T = 10$ K and its intensity drops as the ground state thermally depopulates when raising the temperature. Multiple transitions occur nearly concurrently above 30 K suggesting that excited CEF levels are well separated from the ground-state doublet, but lie close to each other in energy.

Detailed information about the CEF can be obtained by fitting the INS spectra. The Hamiltonian for a Tb^{3+} ion in a hexagonal CEF with D_{6h} point-group symmetry [see Fig. 1(a)] is

$$\mathcal{H}_{\text{CEF}} = B_2^0 \mathcal{O}_2^0 + B_4^0 \mathcal{O}_4^0 + B_6^0 \mathcal{O}_6^0 + B_6^6 \mathcal{O}_6^6. \quad (1)$$

TABLE I. Crystal-field parameters for TbV₆Sn₆ (in meV).

Case	B_2^0	$B_4^0 (\times 10^4)$	$B_6^0 (\times 10^6)$	$B_6^6 (\times 10^6)$	χ_v^2
I	-0.1036(4)	-5.91(2)	2.31(2)	3.3(6)	8.6
II	-0.1151(3)	-5.57(1)	2.31(2)	23.0(5)	11.6

We assume the Tb ion is in its Hund's rule ground state with total angular momentum $J = 6$ ($S = 3$, $L = 3$, and $g_J = 3/2$). $\mathcal{O}_l^m$ are the corresponding Stevens operator equivalents for $J = 6$ and B_l^m are the CEF parameters.

For the measurement of CEF excitations in powder samples with unpolarized neutrons, the neutron intensity is given by

$$\mathcal{S}(Q, \omega) \propto \sum_{j,k} \sum_{\alpha} p_j |\langle \Gamma_k | J_{\alpha} | \Gamma_j \rangle|^2 \delta(E_k - E_j - \hbar\omega). \quad (2)$$

Here, j labels an initial CEF state $|\Gamma_j\rangle$ with energy E_j and thermal population p_j , and k labels a final state $|\Gamma_k\rangle$ with energy E_k . INS transitions follow the dipole selection rules for $J_{\alpha=x,y,z}$ matrix elements ($\Delta m = 0, \pm 1$, where m is the magnetic quantum number).

Using both the PyCrystalField and LMFIT packages [34,35], we simultaneously fit INS spectra at all temperatures in Fig. 1(e) using Eqs. (1) and (2). We use the Voigt peak profile convoluting the resolution and intrinsic widths since the CEF peaks are broader than the instrumental resolution. We achieved two reasonable CEF parameter sets labeled Cases I and II, as shown in Table I and described in detail in the Supplemental Material [31]. The main difference is the size of

the B_6^6 parameter, which is nearly an order of magnitude larger in Case II. This parameter establishes the planar magnetic anisotropy and its magnitude is pivotal in the development of quantum critical behavior, as described below.

Figure 1(f) shows the calculated CEF levels in Case I for variable B_6^6 . For small B_6^6 , the CEF eigenstates can be labeled by $|\Gamma_j\rangle \approx |m\rangle$ and the $\mathcal{O}_6^6$ operator causes small mixing between $|m\rangle$ and $|m \pm 6\rangle$ states (the eigenvectors are listed in Table S1 of the Supplemental Material [31]). As expected, we find that the ground state is an Ising doublet $|\pm 6\rangle$ with a strongly anisotropic orbital wavefunction shown in Fig. 1(b). In both cases, the ground-state doublet is well separated from the first excited state $\approx |\pm 5\rangle$ by $\Delta = 7.9$ meV with the ratio $k_B T_C / \Delta \approx 0.05$, consistent with an Ising character of the FM ordered state. Thermodynamic calculations from these CEF models agree with magnetization (see below), susceptibility, and heat capacity data (see Fig. S1 in the Supplemental Material [31]).

High-field magnetization data. To search for evidence of TFIM quantum criticality, we performed magnetization measurements on TbV₆Sn₆ in pulsed magnetic fields up to 60 T at the National High Magnetic Field Laboratory at Los Alamos National Laboratory using a compensated coil extraction magnetometer. The voltage induced in the coil by the field pulse is proportional to the sample susceptibility (dM/dH). Magnetization $[M(H)]$ data are obtained by subtracting the empty coil signal from the data with the sample inside the coil and integrating with respect to the field (time). Two different crystal mountings were investigated with the field parallel to inequivalent (100) or (210) high-symmetry directions in the hexagonal plane, as shown in Fig. 2(b). Figure 2(a) compares

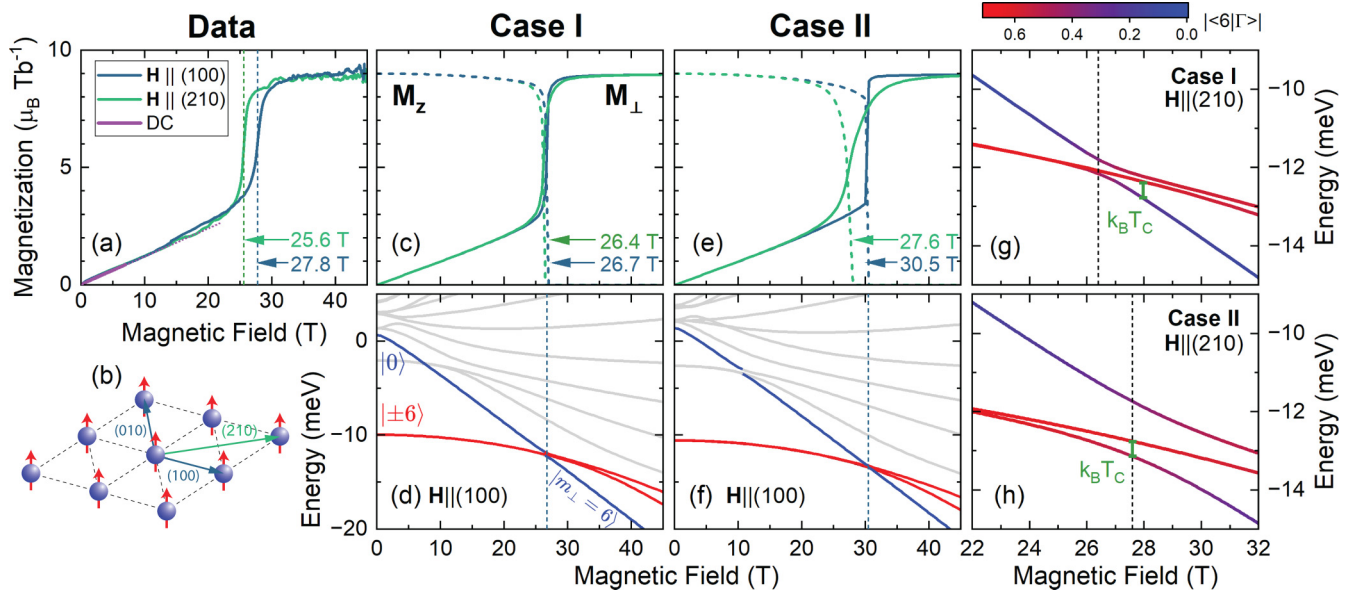


FIG. 2. Transverse-field quantum phase transition. (a) Magnetization data measured in a pulsed transverse field along (100) and (210) directions at $T = 625$ mK. Low-field dc data measured at 1.8 K are shown for comparison. (b) Triangular Tb layer showing field directions. (c) Case I mean-field model calculations of the magnetization components parallel to the (100) and (210) applied field directions ($M_{\perp}$, solid lines) and along (001) (M_z , dashed lines). (d) Evolution of crystal-field levels with transverse field applied along (100). The Ising state doublet (red lines) and the state that evolves from the zero-field $|m = 0\rangle$ state to the planar $|m_{\perp} = 6\rangle$ state (blue line) are highlighted. (e), (f) Mean-field calculations for Case II parameters. Panels (g) and (h) show the evolution of the Case I and Case II CEF levels with transverse field along (210). Lines are colored according to projection of their wavefunction $|\Gamma\rangle$ onto the $|6\rangle$ state. Dashed lines show the critical field and the green bar is $\sim k_B T_C$.

the magnetization with field applied in either direction (raw dM/dH data are shown in Fig. S3 of the Supplemental Material [31]). The magnetization initially tilts weakly toward the field due to the strong Ising anisotropy and then jumps parallel to the field in a first-order-like spin-reorientation transition. We observe critical fields of 25.6 and 27.8 T for (210) and (100) field directions, respectively, suggesting a planar easy axis along the (210) direction ($B_6^0 > 0$). The first-order-like character of the spin-reorientation transition provides clues to the nature of the quantum criticality in TbV_6Sn_6 that are revealed by theoretical analysis described below.

Independent estimates of the B_6^0 CEF parameters derived from magnetization data are described in the Supplemental Material [31] and provide values similar to INS data. The observed critical field anisotropy of 2.2 T between the two field directions allows for an independent determination of $B_6^0 \approx 30 \times 10^{-6}$ meV in support of Case II. However, high-field proximity detector oscillator (PDO) measurements described in the Supplemental Material [31] find evidence that the sample orientation is tilted by strong torques. PDO measurements were performed while varying the in-plane field direction and found a minimum critical field of 25.6 T for $\mathbf{H} \parallel (210)$, but also found a prominent twofold rotational component, rather than the expected sixfold pattern, which suggests sample misalignment. Tilting introduces a small longitudinal component of the applied field [along the (001) axis] that increases the apparent critical field by approximately 1 T per degree of tilt (see the Supplemental Material [31]). Thus, the measured critical field anisotropy is caused by misalignment and we cannot distinguish Case I from Case II.

Quantum criticality. To understand the quantum critical behavior of TbV_6Sn_6 , mean-field (MF) calculations of the magnetic Hamiltonian were performed. We extend the CEF Hamiltonian in Eq. (1) to include an applied magnetic field and an effective isotropic FM Heisenberg coupling between Tb ions with total angular momentum J ,

$$\mathcal{H} = \mathcal{H}_{\text{CEF}} - \frac{1}{2} \sum_{ij} \mathcal{J}_{ij} \vec{J}_i \cdot \vec{J}_j - g_J \mu_B \mu_0 \vec{H} \cdot \sum_i \vec{J}_i. \quad (3)$$

Hyperfine corrections and long-range dipolar interactions are estimated to be small, as discussed in the Supplemental Material [31]. The MF transition temperature is set by $\mathcal{J}_0 = \sum_j \mathcal{J}_{ij}$ and we choose $\mathcal{J}_0 = 122$ mK to reproduce the experimental $T_C = 4.4$ K. Fluctuations will decrease the critical temperature, making this value a lower bound of the interaction strength.

We first employ the Case I CEF parameters where planar anisotropy is small and find that the zero-temperature quantum phase transition is first order for all in-plane field directions indicating that the TFIM QCP is avoided. The calculated magnetization, $\vec{M} = g_J \langle \vec{J} \rangle$, as a function of transverse field is shown in Fig. 2(c), where the critical fields are weakly anisotropic: 26.4 T along the (210) easy axis and 26.7 T along (100). Figure 2(d) shows that the first-order transition is a metamagnetic spin reorientation triggered by a CEF level crossing in the transverse field. Near the critical field, the original ground-state doublet (red lines) mixes with the original $|0\rangle$ state (blue line) that evolves into the $\approx |m_\perp = 6\rangle$ state appropriate for a field-aligned planar moment.

According to our MF calculations, the spin-reorientation transition becomes second order at finite temperature, with a classical tricritical point at $T^* = 1.65$ K (1 K) along the easy (hard) axis, respectively. The tricritical point leads to a quantum critical wing structure [36] in small c -axis fields. This wing structure is very compact, with quantum critical end points at $\mu_0(H_x, H_z) \approx (27, \pm 0.2)$ T along the hard axis, as shown in Figs. 3(g) and 3(h). Similar to a QCP, quantum critical end points possess quantum fluctuations that may mediate exotic high-field superconductivity observed in URhGe [37,38] and UTe_2 [39–43].

Calculations using Case II CEF parameters [Fig. 2(e)] provide a more interesting scenario where a TFIM quantum critical (second-order) transition is observed for the field along the easy axis, while a first-order transition is observed for the hard axis. This indicates that a quantum tricritical point separating first- and second-order transitions would be accessible by varying the field direction. Close to the hard axis, Fig. 3(h) shows that a quantum critical end point appears for fields tilted out of the plane. Figures 2(f) and 2(h) show that quantum critical behavior in Case II is influenced by the same level crossing.

By comparing the two cases, we find that the nature of quantum criticality in TbV_6Sn_6 is highly sensitive to the B_6^0 CEF parameter. To illustrate this, we study how the quantum criticality is tuned by B_6^0 with other Case I parameters held fixed. The mean-field ground-state phase diagrams in a transverse field are shown in Figs. 3(a)–3(f) for evolving B_6^0 , where we find two critical values of B_6^0 . Below the lower critical $B_6^0 = 4.6 \times 10^{-6}$ meV, the transverse-field quantum transitions are all first order, but above it, a line of second-order transitions nucleates around the (210) easy axis, separated from the first-order transitions by QTCPs at finite field angles. As B_6^0 increases, the line of second-order QCPs grows until the QTCPs disappear at the upper critical $B_6^0 = 33 \times 10^{-6}$ meV, above which the transition is second order for all in-plane field directions. The phase diagram evolves from circular to hexagonal as the planar anisotropy B_6^0 increases.

The conditions for quantum critical behavior are understood by examining Figs. 2(g) and 2(h) that show the evolution of CEF levels near the critical field for Cases I and II that, respectively, host first-order and second-order transitions. In the spin-1/2 Ising model, a QCP occurs when the splitting of the doublet is large enough to overcome the longitudinal interaction strength ($\sim k_B T_C$) [44]. For small B_6^0 , Fig. 2(g) shows that a weakly avoided level crossing with the excited CEF singlet occurs before the doublet splitting is large enough to overcome the interaction strength, resulting in a first-order metamagnetic transition. In Fig. 2(h), the larger value of B_6^0 increases level repulsion between the Ising doublet and the excited singlet. The splitting of the Ising doublet is sufficient to overcome the interaction without strong mixing between the doublet and excited singlet, leading to a continuous transition. The field direction dependence of the doublet splitting then allows for the development of quantum critical end points and tricritical points (see the Supplemental Material for details [31]).

Discussion. We have shown that the kagome metal TbV_6Sn_6 is a good realization of an Ising ferromagnet. Remarkably, whether or not it exhibits TFIM quantum criticality,

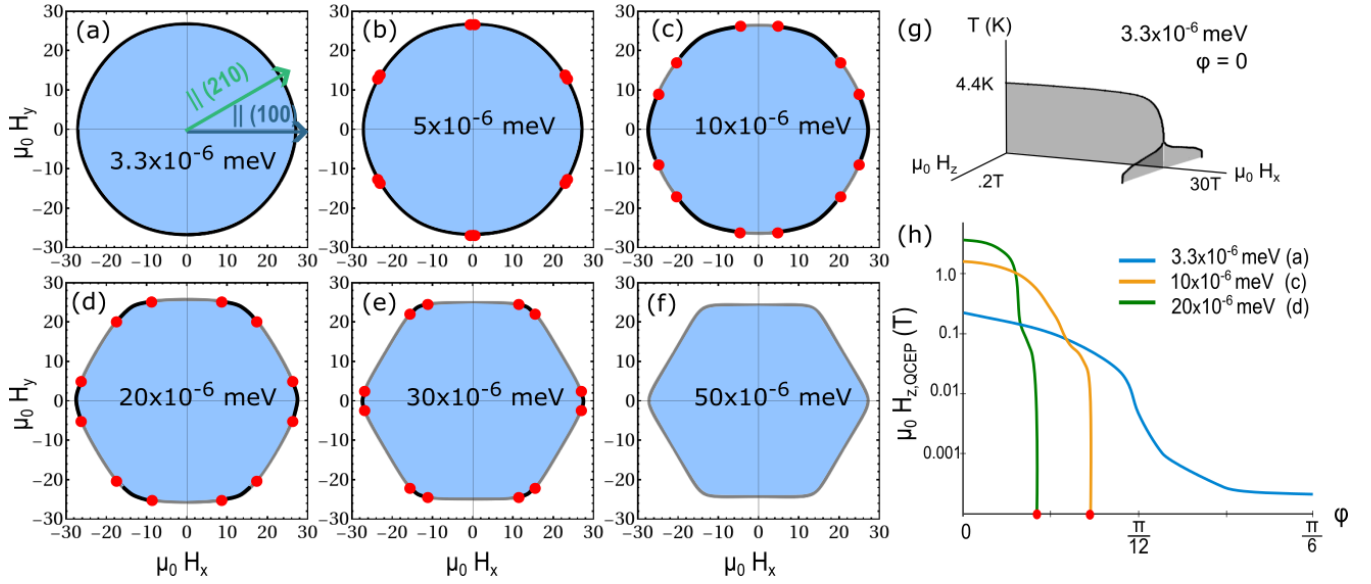


FIG. 3. Quantum criticality in TbV_6Sn_6 . (a)–(f) Mean-field phase diagrams in the H_x - H_y plane [with (100) and (210) directions shown in panel (a)] for Case I CEF parameters with variable B_6^0 values; the B_7^0 and $\mathcal{J}_0$ are held fixed. The blue regions are ferromagnetic and the white regions are quantum paramagnetic. There are two critical B_6^0 values: a lower $B_6^0 = 4.6 \times 10^{-6}$ meV, below which all transitions are first order (black), and an upper $B_6^0 = 33 \times 10^{-6}$ meV, above which all transitions are second order (gray). In-between, there are QTCPs at certain angles (red dots) that separate the first- and second-order lines. Case II behaves very similarly to $B_6^0 = 20 \times 10^{-6}$ meV. (g) Quantum critical wing structure in H_x - H_z plane for Case I parameters. (h) Evolution of the angular dependence of the QCEPs with B_6^0 , in the H_x - H_y plane. The red dots indicate where the QCEPs evolve into QTCPs for larger B_6^0 .

quantum tricriticality or a wing structure with quantum critical end points depends sensitively on the crystal fields and the angle of the transverse field in the ab plane. This discovery reveals a rare opportunity to address the nature and tunability of quantum critical Ising behavior in a non-heavy-fermion metal.

However, our measurements show that the true nature of quantum criticality in TbV_6Sn_6 is somewhat elusive. Our best set of CEF parameters (Case I) places TbV_6Sn_6 below the lower critical value of $B_6^0 = 4.6 \times 10^{-6}$ meV. The resulting first-order metamagnetic transition in a transverse field is predicted to host a wing structure with quantum critical end points in a tilted field. In this respect, the critical behavior is reminiscent of the Blume-Capel model describing an anisotropic spin-1 FM where crossing of the nonmagnetic singlet with the doublet controls the quantum criticality [45,46].

Much larger values of B_6^0 (Case II) are also consistent with the experimental data and would put TbV_6Sn_6 in a regime where the quantum critical nature varies with the in-plane field angle, including QTCPs at particular angles. Density-functional theory estimates ($B_6^0 = 21 \times 10^{-6}$ meV; see the Supplemental Material [31]) are consistent with B_6^0 values in this interesting quantum critical regime. We note that larger values of B_6^0 will not affect the Ising character of TbV_6Sn_6 , as shown in Fig. 1(f).

Thus, our analysis places TbV_6Sn_6 within the reach of field-angle-tuned quantum critical and tricritical behavior. Proximity to a QTCP has been suggested to explain unusual behavior in $\text{Sr}_3\text{Ru}_2\text{O}_7$ [47–49], URhGe [37,38], YbRh_2Si_2 , and other materials [50–54]. The capability to tune RT_6X_6 materials themselves (by chemical substitution, for example) to

increase the planar anisotropy could provide a more definitive example of quantum tricriticality.

The fields required to tune TbV_6Sn_6 into this quantum critical regime are large, but experimentally accessible, making it a potential testing ground to compare the signatures of quantum criticality or tricriticality by tuning the angle of the in-plane magnetic field. However, sample tilting in the presence of large torques would need to be very carefully avoided. One interesting possibility is to search for the finite temperature signatures of quantum criticality. For example, the longitudinal magnetic susceptibility should diverge with different power laws above the QTCPs, $\chi_{zz} \sim T^{-2\psi}$, compared to the QCPs, $\chi_{zz} \sim T^{-\psi}$ [49,55], where ψ is likely 2 for short-range three-dimensional interactions. Alternately, high-field transport measurements could reveal that the critical points are masked by the formation of low-temperature phases, as with the superconductivity in URhGe [37,38] or UTe_2 [39–43], and the nematic spin density waves in ultrapure $\text{Sr}_3\text{Ru}_2\text{O}_7$ [48,56].

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Data availability. The data that support the findings of this article are openly available [57]; embargo periods may apply.

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