Magnetic-field induced melting of long-range magnetic order in Tb$_3$Si$_3$: A possible case for Kitaev physics

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We show that the features attributable to long-range magnetic ordering of the intermetallic compound, Tb$_3$Si$_3$, ($T_N = 69$ K), containing honeycomb network of Tb ions, are completely suppressed by a critical applied field, $H_c$, in heat-capacity and magnetization data. Such a vanishing of long-range magnetic ordering features beyond $H_c$ in these bulk measurements is similar to that commonly known for Kitaev insulators. Increasing magnetic entropy as a function of $H$ with a peak in the magnetically ordered state is consistent with some kind of magnetic disorder induced by $H_c$. The neutron diffraction patterns as a function of $H$ reveal that it is an incommensurate magnetic structure that gets suppressed, leading to magnetically chaotic state with multiple wave vectors beyond $H_c$. Viewing together all the experimental results, we raise a question whether this “melting” of magnetic ordering demands an extension of Kitaev physics to a more broad-based theory, taking into account Ruderman Kittel Kasuya Yoshida indirect exchange interaction instead of superexchange interaction.

Introduction. - In the field of magnetism, there have been constant efforts to find quantum spin-liquid (QSL) behavior due to geometrical frustration [see, for instance, Refs. 1, 2], as originally proposed by Anderson [3]. While such efforts are common on materials with magnetic ions in triangular, kagome, or tetrahedral arrangement, the strongly spin-orbit coupled Mott insulators [4] in which the magnetic ions, with the effective spin ($j_{eff}$) $\frac{1}{2}$, form the (tri-coordinated) honeycomb lattice, characterized by bond-dependent anisotropic exchange interactions, was proposed to provide a promising route to achieve QSL behavior, by the pioneering theory of Kitaev [5]. Jackeli and Khaliullin [6] brought out that this Kitaev prediction might be expected in transition metal oxides, such as Ir$^4+$ and Rh$^4+$, with strong spin-orbit coupling. In conformity with this proposal, Na$_2$IrO$_3$ and α-Li$_2$IrO$_3$ [see, for instance, Refs. 7-9], and α-RuCl$_3$ (see, for instance, Refs. [10-12]) turned out to be first generation systems of this kind. Some 3d systems, e.g., Na$_2$Co$_2$TeO$_6$ [13], are also under discussion for the same in the current literature. However, the applicability of Kitaev physics to 4f magnetic moment containing systems remains largely unexplored, barring some attempts [see the articles cited in Ref. 14]. This is because the super-exchange interaction – an ingredient in Kitaev’s theory – is negligible for localized 4f/electrons. Other points to note are: (i) No metallic system in which the long-ranged Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediates intersite magnetic interaction has been proposed for Kitaev physics, though the Kitaev physics to third nearest neighbour interaction is feasible [15], and (ii) the Kitaev physics was originally proposed for $j_{eff} = \frac{1}{2}$ systems, but there are also theoretical advancements supporting Kitaev physics for larger spins [16, 17]. These advancements encouraged us to search for Kitaev-like features among RKKY-controlled rare-earth intermetallic magnetic materials with higher spin quantum numbers as well. A clue to find such a system is that the dominating non-Kitaev spin exchange channels, responsible for magnetic ordering, should be suppressed by a sufficiently strong magnetic field [see, for instance, Refs. 18-20].

We bring out here a promising case for Kitaev-like physics for a rare-earth with $S = 3$ and $L = 3$ in a metallic environment, viz., Tb$_3$Si$_3$. This compound forming in Mn$_3$Si$_3$-type hexagonal space group $P6_3/mcm$, has been known for several decades [21, 22]. The coordinates of the Tb atoms (labelled Tb1 and Tb2) at the 6g and 4d sites are (x, 0, 1/4) and (1/3, 2/3, 0) respectively, while Si occupies the 6g site (x, 0, 1/4).

![FIG. 1. Crystal structure of Tb$_3$Si$_3$. (a) Unit cell, and (b) Tb2 honeycomb layer along with adjacent Tb1 layer (omitting Si). The green line represents the unit cell boundary.](image-url)
exotic transport anomalies, even in the paramagnetic state as shown in Supplementary Material [29]. The magnetic property relevant to this letter is that there is a magnetic-field-induced transition in the entire $T$-range below 69 K, as inferred from isothermal magnetization $\{M(H)\}$ data, with the critical field ($H_c$) decreasing as $T$ is increased (say, ~58 kOe at 2 K to ~20 kOe at 65 K, as measured in polycrystals). The intriguing property [23-28], which is the motivation for the present investigation is as follows: The magnetoresistance, $MR$, defined as $\rho(H)/\rho(0)$, across this transition is large, but the sign of $MR$ upturn is positive (instead of being negative expected for conventional metamagnetic transitions which lead to ferromagnetic alignment). This surprising “positive” $MR$ jump at $H_c$, is intrinsic to this compound which has been confirmed by measurements on single crystals [28], and was speculated to arise from some kind of magnetic fluctuations, getting triggered by $H_c$. In this letter, we report the results of field-dependent heat-capacity ($C$), dc magnetic susceptibility ($\chi$), and neutron diffraction (ND) measurements to establish that there is indeed a suppression of homogeneous long-range magnetic order with the application of magnetic field, with the bulk data mimicking the behavior of commonly known Rh/Ir-based Kitaev materials mentioned earlier. We therefore raise a question whether this compound is a test case for extending Kitaev-like physics, in a metallic environment for rareearths.

The polycrystalline sample was synthesized by arc melting together high purity constituent elements Tb (> 99.9 wt. %) and Si (> 99.99 wt. %) in proper proportion in an atmosphere of high purity argon gas. The samples were characterized by x-ray diffraction (Cu $K_\alpha$) to be single phase within the detection limit (< 2%) of this technique. Scanning electron microscopic image and energy dispersive x-ray analysis were further employed to ascertain the uniformity and composition of the specimen. Heat-capacity measurements in the presence of several magnetic fields were measured by relaxation method on a piece of about 5 mg with the help of a Physical Properties Measurements System (Quantum Design) in the $T$-range 2 - 140 K; such measurements were reliable up to 60 kOe only, as the specimen tends to reorient at higher fields due to the torque (and hence not presented here). Dc magnetization measurements were also performed in the range 2 - 300 K with several fields in the range 100 Oe to 90 kOe. All these measurements were performed for zero-field-cooled conditions of the specimens. Powder neutron diffraction measurements were carried out on neutron powder diffractometer E6 at BER II reactor of the Helmholtz Zentrum in Berlin using the neutron wavelength $\lambda = 2.43$ Å. To confirm the crystal structure of Tb$_3$Si$_3$, a powder pattern was collected at 300 K in the 20 range between 7.6 - 121.5°. The field induced magnetic order was investigated up to 120 kOe using a shorter range between 3.4 to 73.9° at the temperatures 2, 5, 10, 20, 58, 72 and 110 K.

The results of heat-capacity measurements, measured at 0, 10, 20, 30, 40, and 60 kOe, are shown in Fig. 2a below 85 K. Above 80 K, no significant feature is observed in the data [see also Fig. 1 in Ref. 23]. As reported earlier, in the zero-field data, there is a sharp upturn at 69 K with decreasing temperature, followed by a peak at 67 K, attributable to the onset of a distinct long-range magnetic order, followed a gradual fall down to 2 K. This peak temperature remains almost the same for 10 kOe, but undergoes a downward shift to 66 K and 62.6 K for an application of 20 and 30 kOe respectively. There is also a gradual suppression of the peak value. For an application of 40 kOe, the peak becomes very weak, and it appears at a lower temperature (~57 K). For 50 kOe, the peak is further smeared, appearing as a broad shoulder in the range 30 - 50 K (inset of Fig. 2). For 60 kOe, the peak is completely suppressed. The decrease of ordering temperature with the application of magnetic field and its complete disappearance around 60 kOe is unusual for a strictly localized 4f system and mimics the behavior seen in other Kitaev materials, e.g., Na$_2$Co$_2$TeO$_6$ [Ref. 13] and a-RuCl$_3$, [Ref. 18].

This indicates that Tb$_3$Si$_3$ is transformed into a magnetically “molten” (in other words, a disordered) state around ~60 kOe. For the benefit of the reader, we also show the plots of $C/T$ versus $T$ in the Supplementary Material [29] to reveal that the plots look similar to that of Na$_2$Co$_2$TeO$_6$. The $C$ versus $T$ curves in all fields are found to overlap at low temperatures (e.g., below 40 K), thereby implying gapless behavior of the disordered magnetic state. Finally, it is worth mentioning that there is an additional weak and broad feature in the plot of $C$ versus $T$ in zero field at 70 K (marked by an arrow in Fig. 2a) which persists with the application of magnetic fields (but undergoing a marginal shift towards lower temperatures) and it is possible that it is due to the formation of antiferromagnetic clusters preceding 67K magnetic feature.

FIG. 2. (a) Heat capacity as a function of temperature (<85 K) in the presence of various fields for Tb$_3$Si$_3$. The curves for non-zero magnetic fields are shifted along y-axis (by multiples of 15 J/mol-K, as stated in the figure) for the sake of clarity, as these curves overlap well below the peaks. The small vertical arrow marks an additional weak feature at 70 K. Inset shows the measured data in the expanded form for $H= 50$ kOe in the range 30 to 50 K. (b) Isothermal entropy change derived from heat-capacity data below 100 K for a change of the field from zero to $H$.
In order to render support to the above conclusion on the evolution of magnetically molten state at \( H_c \), we have also derived isothermal entropy change, \( \Delta S = S(H) - S(0) \), as a function of temperature by integrating the area under the curves of \( C/T \) plots. The values thus obtained are shown in Fig. 2h. The overall features are in good agreement with those obtained from the magnetization data [27]. While the negative \( \Delta S \) is expected above 69 K due to the suppression of magnetic disorder or short-range correlations, the sign crossover below 69 K for a conventional (antiferromagnetic to ferromagnetic) metamagnetic transition is unexpected and the positive sign supports the onset of some form of magnetic disorder in high fields. Note that, as a function of \( H \), there is a peak around \( H_c \) below \( T_N \), as shown in Fig. S4 and discussed in Ref. 27, clearly providing evidence for magnetic disorder around \( H_c \), however getting suppressed gradually at further high fields.

De \( \chi \) measured in different external fields are shown in Fig. 3 in the temperature region of interest (< 120 K) to the aim of this article.

\[ \chi(T) \text{ curves obtained in different fields do not overlap over a wide } T \text{-range above 70 K, as shown in the Supplementary Material [29], deviating from the high temperature (> 220 K) Curie-Weiss region; this can be correlated to the transport anomalies in the paramagnetic state [23-28]. In fact, the previous zero-field neutron diffraction data [30, 31] as well as the present neutron diffraction studies reveal the development of magnetic Bragg peaks above 70 K, without any evidence for a well-defined magnetic phase transition temperature. Low-field \( \chi \) data including ac \( \chi \) provide evidence for a magnetic feature around 100 K [29]. We therefore tend to believe that short-range magnetic order indeed occurs well above 70 K, though we still use \( T_N \) for 69 K transition in this paper. Returning to the data below 70 K, there is a peak in the 100 Oe curve at 69 K, expected for the onset of antiferromagnetism, followed by a steep fall and a change of slope at about 64 K, as though there are subtle changes in magnetism with a lowering of temperature. The point of interest is that the features shift to the lower temperature range with increasing fields to ~68.7, ~64 K for 1 kOe, ~68.5, ~63.5 K for 5 kOe, ~67.8, ~62.7 K for 10 kOe, ~63.9, ~54 K for 30 kOe; and ~55, ~39 K for 40 kOe respectively. For 50 kOe, there is a dramatic fall in the peak temperature to ~36.8 K, but no shoulder could be resolved. The fascinating observation is that the decrease of the peak temperature for a further increase of the field does not happen at the same rate; instead, for an application of a field beyond \( H_c \), say, for 60, 70, 90 and 120 kOe, the peak is completely suppressed with the susceptibility tending to a constant value as temperature is lowered to 2 K. This trend resembles that seen in the Kitaev system, \( Na_2Co_2TeO_6 \) [13]. These findings, viewing together with the behavior of the peak in the \( C(T) \) described above, establish that the zero-field (virgin state) long-range antiferromagnetic ordering (< 69 K) is destroyed at \( H_c \) mimicking quantum critical point behavior leading to spin-liquid.

Neutron diffraction experiments reveal a chaotic magnetic state at high fields, particularly in the close vicinity of \( H_c \) with an interplay between commensurate and incommensurate magnetic structures.

\[ \text{FIG. 3. Magnetic susceptibility as a function of temperature for } Tb_2Si_3, \text{ measured in the presence of several fields below 120 K.} \]

\[ \text{FIG. 4. Neutron diffraction patterns of } Tb_2Si_3 \text{ recorded at } T = 2K \text{ under different magnetic fields are shown with Rietveld refinement (continuous lines through the data points) of structural and magnetic phases. Wave vectors, and difference spectra (blue lines) are also given in the figures. Also see Fig. S21 for patterns at more fields indexing the peaks.} \]
a strong magnetic Bragg peak at $Q \sim 0.47\text{Å}^{-1}$, which could be indexed with the incommensurate propagation vector, $k_0 = [0, 0, \pm 0.47]$. For all temperatures below this temperature, down to 2 K, the magnetic structure in zero field could be refined with this incommensurate magnetic structure only. The main point of emphasis relevant to the aim of this letter is that ND patterns with the variation of the magnetic field up to 120 kOe at different temperatures below $T_s$ reveal the appearance of new magnetic phases. At 72 K, the magnetic phase remains commensurate only, with the appearance of a small fraction of ferromagnetic phase as $H$ increases. However, at 58 K, though the zero-field state shows incommensurate magnetic structure, $H$ up to 40 and 60 kOe results in the formation of commensurate ($k_1 = [0, 0, \pm k]$) and $k_0 = [0, 0, 0]$ magnetic phases / propagation vectors at the expense of incommensurate ($k_2 = [0, 0, \pm k_0]$; $k_2 \sim 0.47$) phase. Similar observations are made in the lower temperature ND patterns with increasing $H$. That is, at all temperatures below 58 K, the external field beyond $H_c$ transforms a major part of the sample into commensurate magnetic phase. This conclusion is in fact straightforward, if one looks at the field-induced suppression of the intensity of the peak due to incommensurate magnetic structure, indexed as “$(0,0,0) + k_2$” in Fig. 4 for 2 K. The field-induced phases also show certain degree of “supercooling” effect while reversing the field for some temperatures, as in $M(H)$ curves, however with a full recovery of virgin zero field state, that is, after the field is reversed to zero from high fields (up to 120 kOe).

From the results presented above, it is amply clear that the features attributable to incommensurate long range magnetic ordering vanish in $C(T)$ and $\chi(T)$ data around a certain critical field, leading to magnetic fluctuations. However, there is no evidence for spin-glass behavior even at high fields [29] in our $ac$ measurements. Crucial experimental features offering support to magnetic disorder are: (i) The isothermal entropy change, $\Delta S = S(H) - S(0)$, derived from isothermal $M$ employing Maxwell equation increases with $H$ (below 69 K), peaking around $H_c$, and subsequently falling similar to the trend in the paramagnetic state [27]. The decrease of $\Delta S$ after the peak signals a gradual suppression of the fluctuations well beyond $H_c$; (ii) MR versus $H$ curves are hysteric below the tricritical point (around 20 K) and the MR curve shows an unusual increase below $H_c$, while reducing the field; if this hysteresis curve is extended by applying pressure or by doping by Lu [24, 26], there is a quadratic $H$-dependence of MR as $H$ tends to zero, followed by a linear variation of $M$ with $H$: such a functional form is typical of dominating paramagnetic-like fluctuations [see Figs. S5–S7]. In other words, the magnetic state with fluctuations at $H_c$ gets “supercooled” while reversing the field across the first-order field-induced transition [32]. As argued earlier [27], it is not possible to explain such observations in terms of a high-field ferromagnetic state alone. Looking at the magnetic and magneto-transport behavior of single crystals [Ref. 28, see also Figs. S8-S10], such a field-induced disorder occurs along the basal plane only, implying low-dimensional character of spin-liquid-like anomalies, expected for Kitaev physics.

We now comment on the field range where the idea of proposed Kitaev-like liquid may be dominant. A careful look at the magnetization data of the polycrystals [see Fig. 2, Ref. 23] for 2 K suggests that there are indeed additional steps beyond 60 kOe, which are smeared at higher fields. The corresponding single crystal data (see Fig. S9) reveals a distinct step at 70 kOe (along basal plane only), in addition to the one at about 55 kOe well below $T_s$, e.g., for the 5 K $M(H)$ curve, for the orientation $H$ parallel to a basal plane: such an additional step persists even at 50 K. $M(H)$ almost saturates to 8 $\mu_B$/Tb well below $T_s$, beyond this second step, as though ferromagnetic-like alignment partly occurs at very high fields. The appearance of additional stronger magnetic reflections at 120 kOe is supportive of this line of argument. MR also exhibits an additional drop at a higher field for the single crystals for the orientation along the basal plane (see Fig. 2, in Ref. 28), which is smeared for the polycrystals. In view of these, we are tempted to propose that the fluctuating magnetic state may be more prominent between two critical fields, similar to that noted for Na$_2$Co$_2$TeO$_6$ [Ref. 13].

Summarizing, we bring out that a heavy rare-earth intermetallic compound, Tb$_2$Si$_2$, exhibits magnetic-field-induced chaotic magnetic state, as revealed by heat-capacity, magnetization, and neutron diffraction results. We believe that such a chaotic state following the destruction of uniform long range magnetic order by a critical magnetic field - also consistent with the enhancement of magnetoresistance and of magnetic entropy in the positive quadrant of the respective plots as a function of field – is a special kind of spin-liquid state, induced by $H_c$. Considering that this compound contains a honey-comb layers of Tb ions with strong anisotropy in its properties and the heat-capacity and magnetization behaviors in the presence of magnetic fields are analogous to those of some Kitaev insulators as brought out in this letter, we are compelled to propose that this layered compound is a prospective case for bond-dependent anisotropic exchange interactions - an essential ingredient of Kitaev physics - due to RKKY interaction instead of superexchange interaction to lead to a frustrated magnetic state. We however confess that the presence of two sites for Tb ions makes it difficult to place our conclusions on firmer grounds. Therefore, we call for further work to learn more about this material and to debate on the validity of this idea, considering its implications for the field of frustrated magnetism, in particular, for a step-forward for extensions of Kitaev theory.

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Here we show in Figs. S1-S10 the behavior of temperature and magnetic field dependence of electrical resistivity ($\rho$), isothermal magnetization ($M$), magnetoresistance (MR) and magnetic entropy change ($\Delta S$) behaviors for polycrystalline Tb$_5$Si$_3$. Some of these data for Lu doped specimens as well as for Tb$_5$Si$_3$ under pressure are also shown. Magnetization and MR behavior of single crystalline Tb$_5$Si$_3$ are also shown. These are reproduced from the past literature [S1-S6].

It is notable that the electrical resistivity behavior in the paramagnetic state was found to be intriguing (negative $d\rho/dT$ in zero field, getting suppressed in high fields) [S1], similar to that reported for some other heavy rare-earth compounds [S7, S8]. This transport behavior appears to be a hallmark of classical spin-liquid behavior predicted theoretically [S9] to arise from the combined influence of RKKY interaction and geometrical frustration. Gradual formation of a classical spin-liquid (correlated magnetic clusters) with decreasing temperature above 70 K, sensitive to external fields, may be responsible for this. Thus, magnetic frustration effects are visible even above 69 K over a wide temperature range.

It may be mentioned that the ground state is a doublet for both the Tb sites, judged by crystal-field calculations for isostructural Tb$_5$Sb$_3$ [S10].

The shapes of heat-capacity (present data) divided by temperature obtained in various fields for Tb$_5$Si$_3$ in the present study are also shown (Fig. S11). Inverse susceptibility and ac susceptibility are also shown in Figs. S12-13 to bring out short range magnetic features at low fields near 110 K. As discussed in [S1], there is no evidence for spin-glass in ac susceptibility and the application high fields, e.g., 60 kOe, is not found to change this scenario. More details of neutron diffraction (ND) measurements of results are given for the benefit of interested readers in the form of figures and tables.

Fig. S1: Temperature dependence of electrical resistivity, normalized to 300 K value, for polycrystalline Tb$_5$Si$_3$ in zero field and in 50 kOe. This brings out negative temperature coefficient above 69 K in zero field over a wide temperature range (unusual for an intermetallic compound), which gets suppressed by the external field.
Fig. S2: Isothermal magnetization hysteresis curves for polycrystalline Tb$_5$Si$_3$ at different temperatures to show the behavior of field-induced magnetic transitions for increasing and decreasing directions of magnetic fields in the magnetically ordered state. Hysteresis and supercooling effect following cycling across field-induced transitions can be distinctly seen at 5 K. The hysteresis loop around the critical field gradually vanishes with increasing temperature.

Fig. S3: The behavior of isothermal magnetoresistance, MR = (ρ(H) - ρ(0))/ρ(0), for polycrystalline Tb$_5$Si$_3$ at different temperatures to show the positive MR feature across field-induced magnetic transitions, for increasing and decreasing directions of magnetic fields below 69 K. It is apparent that MR values keep increasing with decreasing field (in the supercooled region), with the critical field becoming lower in this reverse direction. The curves for two temperatures above 69 K, at which there is no field-induced transition, are also shown.
Fig. S4: Isothermal entropy change, $\Delta S = S(H) - S(0)$, for polycrystalline Tb$_5$Si$_3$ at different temperatures, derived from isothermal magnetization data. Note that $\Delta S$ is positive and initially increases, attains a peak around the transition field and then decreases towards negative values, below 67 K. The nature of these curves implies initial magnetic disorder at the respective critical fields, following which this magnetic disorder tends to get gradually depressed at higher fields.

Fig. S5: Magnetoresistance behavior of a Lu doped composition of Tb$_5$Si$_3$ to show that the high-field disordered magnetic phase (and hence its transport behavior) continues to persist down to zero field after a field-cycling (“supercooling”). The red line represents quadratic variation, typical of paramagnets.

Fig. S6: Isothermal magnetization behavior of polycrystalline Lu-doped Tb$_5$Si$_3$ at 1.8K to show that the supercooled high-field phase (as mentioned in Fig. S5 caption) behaves like a paramagnet undergoing essentially linear variation with the field.
Fig. S7: Magnetoresistance behavior of high pressure Tb$_5$Si$_3$ to show that the high-field disordered magnetic phase (and hence its transport behavior) continues to persist down to zero field (in the supercooled region) after a field-cycling, as in Lu-doped specimen.

Fig. S8: Magnetic susceptibility as a function of temperature for single crystalline Tb$_5$Si$_3$ for two geometries. Huge anisotropy is distinctly clear.

Fig. S9: Isothermal magnetization (that is, easy axis) of single crystalline Tb$_5$Si$_3$ for the parallel orientation of magnetic field with respect to b-axis to show additional critical field below $T_N$. 
Fig. S10: Isothermal magnetization (for the hard axis) of single crystalline Tb$_5$Si$_3$ for the parallel orientation of magnetic field with respect to c-axis. Magnetoresistance behavior at 50 K is also shown. These reveal the absence of magnetic field induced transitions for this (hard axis) orientation.

Fig. S11: Heat capacity divided by magnetic field, obtained in this study is plotted to mainly reveal that the curves overlap at low temperatures, for (polycrystalline) Tb$_5$Si$_3$, apart from showing how the peak shifts with increasing field.
Fig. S12: Inverse susceptibility of Tb$_5$Si$_3$, measured with different magnetic fields for Tb$_5$Si$_3$. A broad feature around 120 K at low fields is clearly visible, which is suppressed by a gradual increase of field, similar to Griffiths phase.

Fig. S13: Ac susceptibility of polycrystalline Tb$_5$Si$_3$ measured with 1.3 Hz in different dc magnetic fields to reveal a feature around 110 K at low fields, which is suppressed for dc fields beyond 500 Oe. There is no frequency dependence of the curves.
Neutron Diffraction:

For measuring neutron diffraction patterns, the powdered sample (~ 3g) was packed in a vanadium container and loaded on a liquid helium filled vertical field magnet with variable temperature inset. In order to avoid reorientation of grains under the application of an external magnetic fields, a mixture of deuterated methanol and deuterated ethanol was added to the sample at room temperature and cooled rapidly. On doing so, this mixture freezes around 100 K and locks the sample grains randomly, preventing any preferential orientation by magnetic field.

Neutron diffraction measurements were done in two stages. In the first study (2009 data set), magnetic fields up to 60 kOe were applied (T = 10 K, 58 K, 72 K and 110 K) and in the second study (2010 data set), fields up to 120 kOe were applied (T = 2 K, 5 K and 20 K). Apart from this difference in the maximum field applied, other difference was in the tighter collimation in the second study, which helped in getting better resolution in the diffraction data. However, it may be noted that both the data sets (at T = 2K) yield similar results with respect to magnetic structure and structural parameters.

ND measurements from room temperature (~ 300 K) down to 110 K offer information about the crystallographic (nuclear) and magnetic structure under applied magnetic fields. In-field neutron diffraction measurements were carried out as a function of field, i.e., in the forward cycle of H = 0, 15kOe, 40 kOe, 60 kOe, (80 kOe, 120 kOe in a few cases as described above) and then in the downward cycle of (80, and 60 kOe in some cases) 40, 15 and 0 kOe. The in-field ND measurements were carried out from 110 K down to 2K.

In the ND pattern of Tb, though there is no apparent signature of long-range magnetic order in zero field, in in-field measurements (particularly for H ≥ 40 kOe), a weak ferromagnetic phase is observed which disappears once the field is removed.

In the vicinity of 72 K, just above 69 K, magnetic features appear in the ND pattern of zero field (strong magnetic Bragg peak at Q ~ 0.25 Å⁻¹), which could be ascribed to commensurate magnetic structure with the propagation vector, k1 = [0, 0, ±4]. At T (58K) < TN, the ND pattern appears totally different from that at 72 K, with a strong magnetic Bragg peak appearing at Q = -0.47 Å⁻¹ which is a clear indication of an incommensurate magnetic structure. Attempts to refine the ND pattern with a commensurate magnetic phase or with a combination of incommensurate and commensurate magnetic phases did not yield a satisfactory fitting, and hence for T < TN, for zero field case, a fitting with an incommensurate magnetic phase only was found satisfactory. The strong magnetic peak at 20 ~ 10.5° (Q ~ 0.47 Å⁻¹) can be accounted for by choosing a magnetic structure with incommensurate antiferromagnetic wave vector, k2 = [0, 0, ±k2], where k2 ~ 0.47. A helical magnetic structure is chosen with phase angles constrained for each Tb site. Moments are given in spherical coordinates and all magnetic ions in the unit cell were considered. The atomic positions of all the atoms of Tb6⁴⁺ and Tb4⁴⁺ were given, and the moments were constrained - that is, all atoms at Tb1 (6 sites) were given the same moment, and similarly all atoms at Tb2 (4 sites) were given the same moment. On further decrease in T, ND patterns exhibit only incommensurate antiferromagnetic structure (see tables), with the values of the moments tending to attain saturation moment values for both Tb sites (Tb6⁴⁺ and Tb4⁴⁺) at T = 2K.

Now, turning to the in-field neutron diffraction studies, let us describe how the patterns evolve with decreasing temperature. As stated above, at 72 K, the sample is just above TN, and has a commensurate magnetic structure having k = [0, 0, ±4] in zero field. As the field strength is increased up to 15 kOe, no observable change in the ND pattern is observed. However, for H = 40 kOe, to account for the increase in the intensity of magnetic Bragg peaks at higher angles, ferromagnetic phase (k0 = [0, 0, 0]) is also required to index and fit all the peaks. The combination of two commensurate magnetic phases exists for higher fields (40kOe and 60 kOe) only, essentially in the critical field (Hc) region at this temperature. However, as the field is decreased the high-field structure is retained and, only when field is reduced to zero, the pattern almost matches the virgin ND pattern, essentially tracking isothermal magnetization features, attributable to ‘supercooling effect’ across disorder-broadened first-order transition.

At T = 58 K, the main difference with respect to the pattern for T = 72 K is the observation of a strong magnetic Bragg peak at 20 ~ 10.5° (Q ~ 0.47Å⁻¹) in the ND pattern measured in zero field. The ND pattern could be refined successfully considering an incommensurate antiferromagnetic phase with the propagation vector, k2 = [0, 0, ±k2] where k2 ~ 0.47. As the field strength increases to ~ 40 kOe, another peak at lower Q (~ 0.25 Å⁻¹) is also observed which can be accounted for by introducing a commensurate antiferromagnetic phase with k1 = [0, 0, ±4], which appears to be reminiscent of the high temperature magnetic structure (i.e., 72 K). A weak ferromagnetic phase is also needed for a better refinement of the ND data. Increasing the field further to 60 kOe, completely suppresses the incommensurate antiferromagnetic phase (k2) and the ND pattern can be refined only with a commensurate antiferromagnetic (k1) and ferromagnetic (k0) phases. In the down cycle (that is while reducing the field), the ND pattern is almost similar to that of the upward cycle down to H = 0.

At T= 20K, the incommensurate antiferromagnetic phase with k2 = [0, 0, ±k2], where k2 ~ 0.47] is the most dominant magnetic phase at H = 0. In 60 kOe, the incommensurate phase is largely suppressed, but the ferromagnetic
phase k0 is required along with the weakened k2 phase to refine the entire ND pattern. However, it is interesting to note that at this field, the high temperature commensurate antiferromagnetic phase (k1) appears when the field is increased to 120 kOe at the expense of the incommensurate antiferromagnetic phase (k2). Three coexisting phases are observed in the ND pattern at 120 kOe, i.e., one incommensurate antiferromagnetic phase along with a commensurate antiferromagnetic and ferromagnetic phase. While reversing the field (that is in the down cycle), the commensurate phase (k1) gives way to the incommensurate phase (k2) along with the ferromagnetic k0 phase ($H \sim 60 \text{ kOe}$). After removing the field, the ND pattern can be refined with k2 phase only.

At $T = 5 \text{ K}$, the up-cycle patterns are similar to that observed at 20 K; however, what is interesting is that on the down cycle, at $H = 60 \text{ kOe}$, to get a good fit, two incommensurate magnetic phases (k2 and k1 with refinable $z$ component, i.e., $[0, 0, \pm k_z]$ $k_z \sim 0.23$ for k1) are required. The value of k1, with refinable $z$ component is slightly different from the commensurate value, 0.25, which means that there is a slight field-induced interplay between commensurate and incommensurate magnetic structure at this temperature.

At $T = 2 \text{ K}$, the zero-field magnetic structure could be obtained with k2 incommensurate antiferromagnetic phase. With increase in the field value to 60 kOe, a new feature in the strong magnetic Bragg peak appears which could be indexed with a commensurate magnetic propagation vector, $k_3 = [0, \pm \frac{1}{2}, 0]$. Therefore, the ND pattern at $H = 60 \text{ kOe}$ in the forward cycle at 2 K can be indexed with a combination of ferromagnetic phase, k0, along with two antiferromagnetic phases - one commensurate (k3) and another incommensurate (k2) phase. The high field data could be refined with k0, k1 and k2 phases. In the downward cycle, at $H = 60 \text{ kOe}$ a weak feature is observed in the ND pattern which could be refined by invoking the incommensurate antiferromagnetic k1 phase (i.e., by refining the $k_z$ component of $[0, 0, k_z]$, where $k_z \sim 0.23$). Along with these, two more phases are required to refine the data at this field, i.e., k0 and k2. The zero-field magnetic structure in the down cycle is similar to the up cycle one, which means that there is no major influence of field cycling.

However, it is amply clear from in-field neutron diffraction patterns that, as the field is increased beyond 60kOe, the strong magnetic Bragg peak at $Q = 0.465 \text{ Å}^{-1}$ is completely suppressed, and newer features are observed at higher angles. These new features could be indexed using multiple phases corresponding to coexisting commensurate and incommensurate magnetic phases. The magnetic structures for 60 kOe and beyond are to be described by a combination of incommensurate magnetic phase (with k2 = 0, 0, ±0.47), and commensurate ferromagnetic phases (with k0 = (0,0,0); k3 = (0, $\frac{1}{2}$, 0), or k1 = [0, 0, ±$\frac{1}{4}$].

![Fig. S14](image-url): Room temperature (300 K) neutron diffraction pattern for Tb5Si3 measured on E6 diffractometer ($\lambda = 2.451 \text{Å}$). The good agreement between observed ($I_o$) and calculated ($I_c$) profile confirms the phase formation of the compound. The vertical green tick marks show the Bragg peak positions as per the structural model used for Rietveld refinement. The coordinates of the Tb atoms are $Tb^{\text{dy}} 6g$ ($x$Tb, 0, $1/4$) and $Tb^{\text{dy}} 4d$ ($1/3$, $2/3$, 0), respectively while the silicon atoms occupy the $6g$ ($x$Si, 0, $1/4$) site.
Fig. S15: Rietveld refinement of ND patterns recorded at $T = 110$K in the presence of applied external magnetic fields up to $H = 60$ kOe. Though no structural changes are observed, a small ferromagnetic component with $k_0 = [0,0,0]$ is observed at higher fields ($H = 40$ & $60$ kOe, in both up and down cycles of field variation).

Fig. S16: Rietveld refinement of ND patterns recorded at $T = 72$K, which is close to the magnetic ordering temperature ($T_N \approx 69$ K) as per the magnetization measurements, in the presence of applied magnetic fields up to $H = 60$ kOe. It can be clearly seen that, for $H = 0$, a strong magnetic peak at $20 \approx 5^\circ (Q \approx 0.248 \text{ Å}^{-1})$ is observed, which can be accounted for by choosing a magnetic structure with commensurate antiferromagnetic wave vector, $k_1 = [0,0,\pm \frac{1}{2}]$. The magnetic structure was chosen with moments given in spherical coordinates and all magnetic ions in the unit cell were given.

The atomic positions of all the atoms of Tb$_1^{6g}$ and Tb$_2^{4d}$ were given, and their moment were constrained (all atoms at Tb1 (6 sites) were given same moment, and similarly all atoms at Tb2 (4 sites) were given same moment). At this temperature also, with increasing magnetic field a small ferromagnetic component with $k0 = [0,0,0]$ is observed for $H \geq 40$ kOe.
Fig. S17: Rietveld refinement of ND patterns recorded for $T=58$ K, which is just below $T_N \sim 69$ K, in the presence of applied magnetic fields up to $H = 60$ kOe. It can be clearly seen that, for $H = 0$, a strong magnetic peak at $2\theta \sim 10.5^\circ$ ($Q \sim 0.467$ Å$^{-1}$) is observed, which can be accounted for by choosing a magnetic structure with incommensurate antiferromagnetic wave vector, $k_2 = [0, 0, \pm k_z]$, where $k_z \sim 0.47$. A helical magnetic structure is chosen with phase angles constrained for each Tb site. Moments are given in spherical coordinates and all magnetic ions in the unit cell were given. The atomic positions of all the atoms of Tb$^{16g}$ and Tb$^{24d}$ were given, and their moment were constrained (all atoms at Tb1 (6 sites) were given same moment, and similarly all atoms at Tb2 (4 sites) were given same moment). As field strength increases, there is a coexistence of multiple $k$ vectors, and for high fields ($\geq 40$ kOe), both strong antiferromagnetic peaks at $Q \sim 0.25$ and $\sim 0.47$ Å$^{-1}$ appear and with further increase in field the commensurate phase dominates at the expense of the incommensurate phase.
Fig. S18: Rietveld refinement of ND patterns recorded at $T=20$K under fields up to 120 kOe. Up to $H \sim 60$ kOe, only incommensurate antiferromagnetic phase with $k_2 = [0, 0, \pm k_z]$ persists. On further increase of field to and beyond 60 kOe, a strong ferromagnetic phase ($k_0$) persists with weakened incommensurate antiferromagnetic phase ($k_2$). At $H = 120$ kOe, the incommensurate $k_2$ phase is largely suppressed but the high temperature commensurate antiferromagnetic phase ($k_1$) is observed again.

Fig. S19: Rietveld refinement of ND patterns recorded for $T=10$K under fields up to 60 kOe. For $H = 0$, only incommensurate antiferromagnetic phase ($k_2$) is observed and with increase in $H$, coexistence of a ferromagnetic phase ($k_0$) with ($k_2$) is observed. The increase in $H$ decreases intensity of the strong antiferromagnetic peak at $Q \sim 0.47 \text{ Å}^{-1}$, but strong antiferromagnetic Bragg peaks at higher angles corresponding to this phase persists along with the ferromagnetic phase up to $H = 60$ kOe. However, during the down cycle only incommensurate antiferromagnetic phase is sufficient to get a decent fit to the ND pattern.
Fig. S20: Rietveld refinement of ND patterns recorded at $T=5$K under fields up to 120 kOe. In the upward cycle up to $H \sim 60$ kOe, incommensurate antiferromagnetic phase $k_2$ persists along with the ferromagnetic $k_0$ phase. On further increase of field to and beyond 60 kOe incommensurate antiferromagnetic phase ($k_1$) develops as seen during the measurements at $H = 20$ K. At $H = 120$ kOe, the incommensurate $k_2$ phase is largely suppressed but the high temperature commensurate antiferromagnetic phase ($k_1$) is observed along with the ferromagnetic phase, $k_0$. The high field phase $k_1$ persists and reasonable fitting parameters are observed with this phase is considered as an incommensurate phase and refined. In this case, a $k$ vector of $[0, 0, -0.23]$ gives a good fit and fitting parameters. The $k$ value of 0.23 is not too distant from the commensurate $(\frac{1}{4}) = 0.25$ value, indicating that the magnetic unit cell is close to 4 times the nuclear cell along the $c$-direction. The zero-field incommensurate antiferromagnetic structure with $k_z \sim 0.47$ recovers completely on reducing the field to 0.
Fig. S21: Rietveld refinement of ND patterns recorded at \( T = 2K \) under fields up to 120 kOe. At \( H = 0 \), the incommensurate antiferromagnetic phase \( k_2 \) is observed, as in the case of patterns for higher temperatures (but below \( T_N \)). However, as \( H \) is increased to 60 kOe, the strong antiferromagnetic peak intensity not only decreases, but the width of the peak broadens as if another magnetic structure develops partially at this field and temperature. This additional phase could be indexed using additional propagation vector, \( k_3 = [0, \frac{1}{2}, 0] \). On further increasing the field strength, this additional phase melts and is not recovered again while reversing the field, but at 120 kOe, the profile can be fitted with three propagation vectors, two commensurate phases (\( k_0 \) and \( k_1 \)) and one incommensurate antiferromagnetic phase \( k_2 \). On reducing the field to \( H = 60 \) kOe, the phase \( k_1 \) when treated as an incommensurate phase (\( k = [0, 0, \pm 0.23] \) gives good fit and reliable refinement parameters. On reducing the field to 0, the zero-field incommensurate antiferromagnetic phase, \( k_2 \), is recovered almost completely. In the right figure, the ND diffraction patterns shown in Fig. 4 are replotted in an expanded region in the range 32 to 64\(^\circ\), indexing the peaks.
Table A (Zero field ND analysis results)

Magnetic structure of Tb₅Si₃: \(ab\)-plane \(\mathbf{AF}_{ab}^{K_1}\) (\(K_1 = [0, 0, \pm 1/4]\)) and \(\mathbf{AF}_{ab}^{K_2}\) incommensurate \(K_2 = [0, 0, \pm K_z]\)

| T (K) | Unit cell (Å) | \(x_{\text{Tb}_{bg}}\) | \(x_{\text{Si}_{bg}}\) | \(M_{\text{Tb}_{bg}}^{K_1}\) (\(\mu_B\)) | \(M_{\text{Tol}_{d}}^{K_1}\) (\(\mu_B\)) | \(M_{\text{Tol}_{d}}^{K_2}\) (\(\mu_B\)) | \(K_2\) | \(R_F\) (%) | \(R_F^m\) (%) |
|-------|---------------|-----------------|-----------------|-------------------|-------------------|-------------------|---------|-----------|-----------|
| 300   | \(a = 8.4568(11)\) \(c = 6.3574(8)\) | 0.244 | 0.608 | | | | | 4.9 | |
| 110   | \(a = 8.4616(25)\) \(c = 6.3496(32)\) | 0.238 | 0.599 | | | | | 2.9 | |
| 72    | \(a = 8.4591(35)\) \(c = 6.3180(35)\) | 0.251 | 0.627 | 2.9(2) | 2.4(4) | | | 1.8 | 2.5 |
| 58    | \(a = 8.4418(8)\) \(c = 6.3247(10)\) | 0.246 | 0.626 | 6.7(1) | 7.2(1) | 0.471(1) | | 3.4 | 1.5 |
| 20    | \(a = 8.4461(12)\) \(c = 6.3587(12)\) | 0.238 | 0.568 | 8.3(1) | 8.2(1) | 0.464(1) | | 9.5 | 2.4 |
| 10    | \(a = 8.4403(13)\) \(c = 6.3511(17)\) | 0.240 | 0.591 | 8.0(1) | 8.8(1) | 0.466(1) | | 3.9 | 1.1 |
| 5     | \(a = 8.4442(9)\) \(c = 6.3474(9)\) | 0.240 | 0.579 | 8.7(1) | 8.0(1) | 0.466(1) | | 4.3 | 2.8 |
| 2     | \(a = 8.4528(14)\) \(c = 6.3534(14)\) | 0.239 | 0.592 | 8.8(2) | 8.7(2) | 0.469(2) | | 14.1 | 3.6 |
| H (kOe) | Unit cell (Å) | \( M_{Tb6g}^{k_0} \) (\( \mu_B \)) | \( M_{Tb4d}^{k_0} \) (\( \mu_B \)) | \( M_{Tb6g}^{k_1} \) (\( \mu_B \)) | \( M_{Tb4d}^{k_1} \) (\( \mu_B \)) | \( R_f^m \) | \( K_1 \) (\% ) | \( R_f^m \) | \( K_2 \) (\% ) | \( R_f^m \) | \( K_3 \) (\% ) | \( R_f \) (\% ) |
|--------|---------------|-----------------|-----------------|-----------------|-----------------|-------|-------|-------|-------|-------|-------|-------|
| 0      | \( a = 8.4528(14) \) \( c = 6.3534(14) \) | 8.8(2) | 8.7(2) | 0.469 | 3.6 | 14.1 |
| 15     | \( a = 8.4535(14) \) \( c = 6.3532(14) \) | 8.7(2) | 8.7(2) | 0.469 | 3.7 | 13.1 |
| 40     | \( a = 8.4535(12) \) \( c = 6.3535(12) \) | 8.5(2) | 8.3(2) | 0.469 | 3.7 | 6.2 |
| 60     | \( a = 8.4515(12) \) \( c = 6.3485(15) \) | 3.0(2) | 4.3(4) | 7.1 | 4.0(2) | 3.4(2) | 0.467 | 12.7 | 3.6 | (5) | 1.9 | (4) | 31.3 | 8.9 |
| 80     | \( a = 8.4449(12) \) \( c = 6.3475(13) \) | 4.6(3) | 5.0(4) | 4.4 | 0.3(3) | 2.9(5) | 13.1 | 0.4(2) | 0.2(2) | 0.467 | 24.9 | 4.3 |
| 120   | \( a = 8.4375(9) \) \( c = 6.3504(10) \) | 8.1(9) | 7.3(9) | 6.6 | 0.6(7) | 2.3(1) | 31.6 | 0.2(3) | 0.1(2) | 0.467 | 24.6 | 4.1 |
| 80     | \( a = 8.4412(11) \) \( c = 6.3482(12) \) | 6.2(6) | 6.2(6) | 6.4 | 1.2(5) | 2.7(8) | 16.5 | 0.2(2) | 0.1(1) | 0.467 | 21.3 | 5.0 |
| 60     | \( a = 8.4453(12) \) \( c = 6.3458(13) \) | 4.5(3) | 4.8(4) | 5.0 | 1.2(3) | 3.9(5) | 10.3 | 0.2(2) | 0.1(1) | 0.467 | 20.4 | 5.3 |
| 40     | \( a = 8.4488(13) \) \( c = 6.3585(18) \) | 4.2(1) | 4.0(1) | 0.459 | 6.9 | 2.8 |
| 15     | \( a = 8.4530(12) \) \( c = 6.3559(11) \) | 8.6(1) | 7.7(1) | 0.463 | 3.2 | 1.4 |
| 0      | \( a = 8.4530(12) \) \( c = 6.3562(11) \) | 8.6(1) | 7.6(2) | 0.463 | 3.1 | 1.4 |

Table B1 (Field scan ND analysis results): \( T = 2 \text{K} \) (2010 Data Set)

Tentative magnetic structure of Tb₅Si₃ as a sum of \( ab \)-plane \( \text{AF}_{ab}^{k_1} \) and \( \text{AF}_{ab}^{k_2} \) incommensurate (\( K_1 = [0, 0, \pm 1/4] \), \( K_2 = [0, 0, \pm K_z] \)), \( a \)-axis \( \text{AF}_{a}^{k_3} \) antiferromagnet (\( K_3 = [0, 1/2, 0] \)) and \( a \)-axis ferromagnet \( \text{F}_{a}^{k_0} \) (\( K_0 = [0, 0, 0] \)).
Table B2 (Field scan ND Analysis results): $T = 5K$ (2010 Data Set)

Magnetic structure of Tb$_5$Si$_3$ at 5 K

as a sum of $ab$-plane $\mathbf{AF}_{ab}^{k_1}$ and $\mathbf{AF}_{ab}^{k_2}$ incommensurate ($\mathbf{K}_1 = [0, 0, \pm l/4]$, $\mathbf{K}_2 = [0, 0, \pm K_z]$), and $a$-axis ferromagnet $\mathbf{F}_{a}^{k_0}$ ($\mathbf{K}_0 = [0, 0, 0]$).

| H (kOe) | Unit cell (Å) | $M_{\mathrm{TMG}}^{K_1}$ (μB) | $M_{\mathrm{TMG}}^{K_2}$ (μB) | $R_f^m$ | $M_{\mathrm{TMG}}^{K_1}$ (μB) | $M_{\mathrm{TMG}}^{K_2}$ (μB) | $R_f^m$ | $M_{\mathrm{TMG}}^{K_2}$ (μB) | $R_f^m$ | $K_z$ | $R_f$ (%) |
|---------|---------------|------------------|------------------|-------|------------------|------------------|-------|------------------|-------|-------|-----------|
| 0       | $a = 8.4442(9)$ | $c = 6.3474(9)$ | 8.7(1) | 8.0(1) | 0.466 | 2.9 | 4.3 |
| 15      | $a = 8.4446(10)$ | $c = 6.3461(10)$ | 8.7(1) | 8.2(1) | 0.466 | 2.9 | 3.9 |
| 40      | $a = 8.4444(10)$ | $c = 6.3460(10)$ | 8.3(1) | 7.9(1) | 0.466 | 2.8 | 4.5 |
| 50      | $a = 8.4439(10)$ | $c = 6.3463(11)$ | 7.9(1) | 7.5(1) | 0.467 | 3.3 | 4.7 |
| 60      | $a = 8.4414(12)$ | $c = 6.3438(15)$ | 3.0(2) | 3.3(4) | 6.6 | 3.2(1) | 2.7(1) | 0.465 | 22.5 | 6.6 |
| 80      | $a = 8.4342(11)$ | $c = 6.3427(11)$ | 4.9(4) | 5.1(5) | 6.9 | 0.0 | 1.7(6) | 32.5 | 1.9(2) | 1.4(2) | 0.473 | 17.0 | 3.2 |
| 120     | $a = 8.4278(13)$ | $c = 6.3441(11)$ | 7.6(10) | 7.9(11) | 12.7 | 0.5(4) | 3.5(7) | 30.1 | 1.6(4) | 1.7(4) | 0.483 | 30.4 | 8.3 |
| 80      | $a = 8.4337(11)$ | $c = 6.3410(11)$ | 5.4(4) | 5.2(4) | 11.1 | 2.0(4) | 4.1(4) | 5.1 | 1.7(2) | 0.9(2) | 0.480 | 24.9 | 7.7 |
| 60      | $a = 8.4374(13)$ | $c = 6.3391(12)$ | 4.1(2) | 3.9(3) | 9.6 | 2.2(6) | 6.4(4) | 5.7 | 4.9(3) | 1.6(4) | 0.471 | 33.5 | 7.6 |
| 40      | $a = 8.4470(17)$ | $c = 6.3342(23)$ | 2.3(2) | 2.4(3) | 9.7 | 1.3(2) | 2.5(3) | 18.4 | 1.9(1) | 1.1(1) | 0.460 | 30.7 | 6.4 |
| 0       | $a = 8.4443(10)$ | $c = 6.3469(10)$ | 8.5(1) | 8.0(1) | 0.465 | 3.1 | 4.1 |
Table B3 (Field scan ND analysis results): \( T = 10 \text{ K} \) (2009 Data Set)
Magnetic structure of Tb$_5$Si$_3$ at 10 K
as a sum of \( ab\)-plane \( \text{AF}_{ab}^{K_1} \) and \( \text{AF}_{ab}^{K_2} \) incommensurate \((K_1 = [0, 0, \pm 1/4], K_2 = [0, 0, \pm K_z])\), and \( a\)-axis
ferromagnet \( \text{F}_{a}^{K_0} \) \((K_0 = [0, 0, 0])\).

| \( H \) (kOe) | Unit cell (Å) | \( M_{\text{Tb}g}^{K_0} \) \((\mu_B)\) | \( M_{\text{Tb}d}^{K_0} \) \((\mu_B)\) | \( R_{f}^{m} \) \(K_0\) (%) | \( M_{\text{Tb}g}^{K_1} \) \((\mu_B)\) | \( M_{\text{Tb}d}^{K_1} \) \((\mu_B)\) | \( R_{f}^{m} \) \(K_1\) (%) | \( M_{\text{Tb}g}^{K_2} \) \((\mu_B)\) | \( M_{\text{Tb}d}^{K_2} \) \((\mu_B)\) | \( K_z \) | \( R_{f}^{m} \) \(K_2\) (%) | \( R_{f} \) (%) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 0 | \( a = 8.4403(13) \) \( c = 6.3651(16) \) | | | | 8.0(1) | 8.8(1) | 0.466 | 1.1 | 3.8 |
| 15 | \( a = 8.4403(13) \) \( c = 6.3652(17) \) | | | | 8.0(1) | 8.8(1) | 0.466 | 1.0 | 4.0 |
| 40 | \( a = 8.4401(14) \) \( c = 6.3650(18) \) | | | | 7.7(1) | 8.6(2) | 0.466 | 1.0 | 3.1 |
| 60 | \( a = 8.4378(22) \) \( c = 6.3446(28) \) | 2.7(5) | 2.7(5) | 8.0 | 3.7(8) | 0.0 | 15.4 | 3.3(3) | 3.4(4) | 0.458 | 5.5 | 7.3 |
| 40 | \( a = 8.4391(17) \) \( c = 6.3667(19) \) | | | | 7.7(1) | 8.5(2) | 0.460 | 1.0 | 3.6 |
| 15 | \( a = 8.4404(14) \) \( c = 6.3662(17) \) | | | | 8.1(1) | 8.9(1) | 0.462 | 1.5 | 3.4 |
| 0 | \( a = 8.4413(13) \) \( c = 6.3660(16) \) | | | | 8.1(1) | 8.9(1) | 0.462 | 1.3 | 3.4 |
Table B4 (Field scan ND analysis results): T = 20K (2010 Data Set)

Magnetic structure of TbSi3 at 20 K
as a sum of ab-plane AF_{ab K1} and AF_{ab K2} incommensurate (K1 = [0, 0, ±1/4], K2 = [0, 0, ±Kz]), and a-axis ferromagnet F_{a K0} (K0 = [0, 0, 0]).

| H (kOe) | Unit cell (Å) | M_{ab Tb6 g K0} (µB) | M_{ab Tb4 d K0} (µB) | M_{ab Tb6 g K1} | M_{ab Tb4 d K1} | M_{ab Tb6 g K2} | M_{ab Tb4 d K2} | Kz | Rf (%) |
|---------|---------------|-----------------------|-----------------------|-----------------|-----------------|-----------------|-----------------|-----|--------|
| 0       | a = 8.4461(12) | c = 6.3587(12) | 8.3(1) | 8.2(1) | 0.464 | 2.4 | 9.5 |
| 15      | a = 8.4462(13) | c = 6.3568(13) | 8.2(1) | 8.3(2) | 0.464 | 2.7 | 8.5 |
| 40      | a = 8.4454(12) | c = 6.3583(12) | 8.0(1) | 8.0(1) | 0.464 | 2.1 | 9.5 |
| 60      | a = 8.4493(11) | c = 6.3413(11) | 4.2(2) | 4.9(3) | 5.7 | 8.2(3) | 6.2(4) | 0.469 | 13.6 | 6.5 |
| 80      | a = 8.4390(10) | c = 6.3462(11) | 5.4(4) | 5.1(4) | 6.4 | 2.3(15) | 4.3(14) | 29.0 | 6.8(4) | 5.6(6) | 0.474 | 14.9 | 5.0 |
| 120     | a = 8.4329(10) | c = 6.3479(11) | 6.7(7) | 6.4(6) | 10.3 | 0.5(11) | 1.2(16) | 29.0 | 4.2(5) | 4.2(9) | 0.479 | 25.9 | 5.4 |
| 80      | a = 8.4383(10) | c = 6.3453(11) | 5.3(3) | 5.0(3) | 7.6 | 1.6(13) | 4.0(12) | 17.9 | 8.2(9) | 8.4(10) | 0.457 | 24.0 | 7.2 |
| 60      | a = 8.4463(15) | c = 6.3423(16) | 4.7(4) | 5.4(4) | 6.7 | 8.2(5) | 6.1(6) | 0.465 | 23.0 | 8.4 |
| 40      | a = 8.4443(17) | c = 6.3553(17) | 7.7(2) | 7.6(2) | 0.456 | 3.0 | 8.3 |
| 15      | a = 8.4455(15) | c = 6.3564(14) | 8.2(2) | 8.3(2) | 0.462 | 2.7 | 8.9 |
| 0       | a = 8.4462(13) | c = 6.3559(13) | 8.3(1) | 8.4(2) | 0.463 | 2.8 | 12.0 |
Table B5 (Field scan ND analysis results): $T = 58$ K (2009 Data Set)

Magnetic structure of Tb$_5$Si$_3$ at 58 K as a sum of $ab$-plane AF$_{ab}^{K_1}$ and AF$_{ab}^{K_2}$ incommensurate ($K_1 = [0, 0, \pm 1/4]$, $K_2 = [0, 0, \pm K_z]$), and $a$-axis ferromagnet F$_a^{K_0}$ ($K_0 = [0, 0, 0]$).

| $H$ (kOe) | Unit cell (Å) | $M_{Tb6g}^{K_0}$ (µB) | $M_{Tb4d}^{K_0}$ (µB) | $Rm^m$ | $M_{Tb6g}^{K_1}$ (µB) | $M_{Tb4d}^{K_1}$ (µB) | $Rm^m$ | $M_{Tb6g}^{K_2}$ (µB) | $M_{Tb4d}^{K_2}$ (µB) | $K_z$ | $Rm^m$ | $R_F$ (%) |
|-----------|---------------|------------------------|------------------------|--------|------------------------|------------------------|--------|------------------------|------------------------|--------|--------|-----------|
| 0         | $a = 8.4416(8)$ | $c = 6.3246(10)$       |                        |        |                        |                        |        |                        |                        |        |        |           |
| 15        | $a = 8.4416(9)$ | $c = 6.3251(10)$       |                        |        |                        |                        |        |                        |                        |        |        |           |
| 40        | $a = 8.4450(34)$ | $c = 6.3408(42)$       | 1.9(2)                 | 0.3(3) | 6.9                    | 2.1(5)                 | 1.9(9) | 6.3                    | 3.5(1)                 | 0.4(2) | 0.418   | 21.2      |
| 60        | $a = 8.4504(27)$ | $c = 6.3362(25)$       | 3.0(2)                 | 3.6(3) | 6.0                    | 2.3(3)                 | 3.1(6) | 6.2                    |                        |        |        |           |
| 40        | $a = 8.4449(33)$ | $c = 6.3426(41)$       | 2.1(2)                 | 0.6(3) | 7.0                    | 2.1(5)                 | 2.0(9) | 5.6                    | 3.2(1)                 | 0.0    | 0.413   | 24.4      |
| 15        | $a = 8.4421(9)$ | $c = 6.3246(10)$       |                        |        |                        |                        |        |                        |                        |        | 0.469   | 1.0       |
| 0         | $a = 8.4424(9)$ | $c = 6.3241(10)$       |                        |        |                        |                        |        |                        |                        |        | 0.470   | 1.0       |
Table B6 (Field scan ND analysis results): $T = 72$ K (2009 Data Set)

Magnetic structure of Tb$_5$Si$_3$ at 72 K as a sum of $ab$-plane $\mathbf{AF}_{ab}^{K_1}$ and $\mathbf{AF}_{ab}^{K_2}$ incommensurate ($K_1 = [0, 0, \pm 1/4]$, $K_2 = [0, 0, \pm K_z]$), and $a$-axis ferromagnet $\mathbf{F}_{a}^{K_0}$ ($K_0 = [0, 0, 0]$).

| $H$ (kOe) | Unit cell (Å) | $M_{\text{at6g}}^{K_0}$ ($\mu_B$) | $M_{\text{at6d}}^{K_0}$ ($\mu_B$) | $R_f^m$ ($\mu_B$) | $K_0$ (%) | $R_f^m$ K$_0$ (%) | $R_f$ (%) |
|----------|--------------|-------------------------------|-------------------------------|------------------|---------|-----------------|--------|
| 0        | $a = 8.4590(34)$, $c = 6.3179(35)$ | 2.9(2)                        | 2.4(4)                        | 2.5              | 1.8     |                 |        |
| 15       | $a = 8.4565(34)$, $c = 6.3199(36)$ | 2.6(2)                        | 2.1(4)                        | 6.3              | 1.8     |                 |        |
| 40       | $a = 8.4488(23)$, $c = 6.3284(20)$ | 2.7(1)                        | 3.6(3)                        | 8.2              | 2.5(3)  | 2.5(6)          | 6.3    |
| 60       | $a = 8.4442(15)$, $c = 6.3319(13)$ | 2.7(1)                        | 4.3(2)                        | 4.8              | 1.7(3)  | 2.2(5)          | 7.5    |
| 40       | $a = 8.4488(23)$, $c = 6.3284(20)$ | 2.7(1)                        | 3.8(2)                        | 7.2              | 2.5(3)  | 2.5(6)          | 6.0    |
| 15       | $a = 8.4554(32)$, $c = 6.3210(33)$ | 2.5(2)                        | 2.1(4)                        | 4.5              | 1.4     |                 |        |
| 0        | $a = 8.4563(35)$, $c = 6.3200(36)$ | 2.7(2)                        | 2.2(4)                        | 5.5              | 2.2     |                 |        |
Table B7 (Field scan ND analysis results): $T = 110$ K (2009 Data Set)
Magnetic structure of Tb$_5$Si$_3$ at 110 K
Field induced changes to $a$-axis ferromagnet $F_a^0$ ($K_0 = [0, 0, 0]$).

| $H$ (kOe) | Unit cell (Å) | $M_{Tb0y}^K$ ($\mu_B$) | $M_{Tb0x}^K$ ($\mu_B$) | $R_{Fm}^\alpha$ ($\%$) | $RF$ ($\%$) |
|-----------|---------------|-------------------------|-------------------------|------------------------|-------------|
| 0         | $a = 8.4616(26)$ |                         |                         |                        | 2.9         |
|           | $c = 6.3495(32)$ |                         |                         |                        |             |
| 15        | $a = 8.4623(25)$ |                         |                         |                        | 2.7         |
|           | $c = 6.3511(31)$ |                         |                         |                        |             |
| 40        | $a = 8.4616(20)$ |                         |                         |                        | 2.7         |
|           | $c = 6.3529(20)$ | 1.0(1)                  | 0.0                     | 20.8                   | 3.4         |
| 60        | $a = 8.4682(16)$ |                         | 0.7(1)                  |                         | 9.8         |
|           | $c = 6.3464(15)$ |                         | 1.2(1)                  |                         | 3.1         |
| 40        | $a = 8.4619(20)$ |                         | 1.0(1)                  | 0.0                     | 26.8        |
|           | $c = 6.3530(19)$ |                         |                         |                        | 3.0         |
| 15        | $a = 8.4678(22)$ |                         |                         |                        | 8.1         |
|           | $c = 6.3448(22)$ |                         |                         |                        |             |
| 0         | $a = 8.4694(22)$ |                         |                         |                        | 11.0        |
|           | $c = 6.3435(27)$ |                         |                         |                        |             |
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