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Anomalous pressure dependence of magnetic ordering temperature in Tb to 141 GPa: comparison with Gd and Dy

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In previous studies the pressure dependence of the magnetic ordering temperature T_o of Dy was found to exhibit a sharp increase above its volume collapse pressure of 73 GPa, appearing to reach temperatures well above ambient at 157 GPa. In a search for a second such lanthanide, electrical resistivity measurements were carried out on neighboring Tb to 141 GPa over the temperature range 3.8 - 295 K. Below Tb's volume collapse pressure of 53 GPa, the pressure dependence $T_o(P)$ mirrors that of both Dy and Gd. However, at higher pressures $T_o(P)$ for Tb becomes highly anomalous. This result, together with the very strong suppression of superconductivity by dilute Tb ions in Y, suggests that extreme pressure transports Tb into an unconventional magnetic state with an anomalously high magnetic ordering temperature.

I. INTRODUCTION

The magnetic ordering temperatures T_o of Gd and Dy have been recently shown to track each other in a highly non-monotonic fashion as a function of pressure to $\sim$ 70 GPa; at higher pressures they deviate markedly, T_o for Dy rising rapidly to temperatures well above ambient at 157 GPa.[1] Parallel experiments on dilute magnetic alloys of Gd and Dy with superconducting Y suggest that for pressures above $\sim$ 70 GPa Dy is transformed from a magnetically conventional lanthanide into one with an unconventional magnetic state with marked deviations from de Gennes scaling,[2] a state perhaps governed by Kondo physics, indicating that the Dy ion is nearing a magnetic instability.[1] An alternate explanation is that the strong enhancement of T_o in Dy arises through changes in the crystalline electric field at extreme pressure.[1] Analogous studies on additional lanthanides are recommended to help identify the origin of this anomalous behavior.

The lanthanide Tb, which lies between Gd and Dy in the periodic table, has one fewer $4f$ electron than Dy, and is probably less stable magnetically than Dy due to its direct proximity to Gd, by far the most stable of all magnetic lanthanides. Tb orders antiferromagnetically (AFM) at $T_o \simeq 230$ K followed by a ferromagnetic (FM) transition at $T_o \simeq 220$ K.[3] Both transition temperatures initially decrease rapidly with pressure at the rate -10 to -12 K/GPa, but above $\sim$ 7 GPa neither transition can be clearly detected in either the ac or dc magnetic susceptibility.[4–6] The disappearance of the ordered moment in the susceptibility measurement indicates a transition to either an AFM or paramagnetic state above 7 GPa. Electrical resistivity studies should reveal which scenario is correct since both FM and AFM order normally lead to a distinct kink in the temperature dependence of the resistivity. However, recent resistivity and neutron diffraction experiments on Tb find that the FM transition decreases with pressure at the rate -16.7 K/GPa to 3.6 GPa;[7] that the transition could no longer be resolved above 3.6 GPa may be due to appreciable pressure-gradient broadening in the cell which contained no pressure medium. That magnetic order in Tb disappears above 7 GPa seems highly unlikely since both x-ray absorption near-edge structure (XANES) and non-resonant x-ray emission spectroscopy (XES) measurements detect no change in Tb's valence to 65 GPa and 70 GPa, respectively.[8] In fact, the XES studies show that Tb retains its strong, highly localized magnetic moment ($J = 6$) to at least 70 GPa.[8]

In this paper we present the results of dc electrical resistivity measurements on Tb over the temperature range 3.8 - 295 K to pressures as high as 141 GPa, well above the pressure of 53 GPa where Tb suffers a 5% volume collapse at the phase transition from hexagonal hR24 to body-centered monoclinic (bcm).[9] Magnetic order is indeed observed in Tb for pressures above 7 GPa. In fact, to 53 GPa $T_o(P)$ follows nearly the same highly non-monotonic pressure dependence found earlier in Gd and Dy,[1] but deviates markedly at higher pressures. As the applied pressure passes through 53 GPa, $T_o(P)$ for Tb first decreases, but then begins to increase rapidly above 80 GPa. As suggested for Dy,[1] extreme pressure appears to transport Tb into an unconventional magnetic state with an anomalously high magnetic ordering temperature, well above that anticipated from conventional de Gennes scaling.

II. EXPERIMENTAL TECHNIQUES

Resistivity samples were cut from a Tb ingot (99.9% pure, Material Preparation Center of the Ames Laboratory[10]). To generate pressures well beyond the volume collapse pressure of Tb at 53 GPa, a diamond anvil cell (DAC) made of CuBe alloy was used.[11] Two separate high-pressure experiments were carried out where pressure was generated by two opposing diamond anvils (1/6-carat, type Ia) with 0.35 mm diameter culets beveled at 7° to 0.18 mm central flats.

The Re gasket (6 - 7 mm diameter, 250 μm thick) was preindented to 30 μm and a 80 μm diameter hole electro-spark drilled through the center. The center section of the preindented gasket surface was filled with a 4:1 cBN-epoxy mixture to insulate the gasket and serve as pressure medium. The thin Tb sample was then placed on top of four thin (4 μm) Pt leads for a four-point dc electrical resistivity measurement. In an attempt to minimize the effect of the pressure gradient across the sample in this non-hydrostatic pressure environment, in run 1 an elongated sample (dimensions $\sim$8$\times$80$\times$3 μm^3) was used with the two voltage leads spaced only 5 μm apart (see inset to Fig. 1(a)). In run 2 all four Pt leads were placed near the corners of the square-shaped sample (dimensions $\sim$30$\times$30$\times$5 μm) (see inset to Fig. 1(b)), as in the previous resistivity measurements on Dy.[1] However, from the temperature-dependent resistivity data the pressure gradient was estimated to be approximately the same in both runs. Further details of the non-hydrostatic high pressure resistivity technique are given in a paper by Shimizu *et al.*[12]

A He-gas driven membrane was utilized to change pressure at any temperature.[13] The value of the pressure was determined using both the fluorescence[14] from a small ruby sphere positioned at the edge of the sample and the frequency shift of the diamond vibron via Raman spectroscopy.[15] The ruby pressure was determined at both ambient temperature and a temperature within 20 K of $T_{\rm o}$; the vibron pressure was determined only at ambient temperature. The values of the pressure given are averaged over the sample to an estimated accuracy of $\pm$10%. In these experiments temperatures from 3.8 K to 295 K were available using an Oxford flow cryostat. All measurements shown in this paper were carried out with increasing pressure; diamond anvil failure at the highest pressure ended the experiment. Further experimental details of the DAC and cryostat are given elsewhere.[8,11,16,17]

III. RESULTS OF EXPERIMENT

The present resistivity studies on Tb were carried out in two separate experiments. In Fig. 1(a) the electrical resistance $R(T)$ from run 1 is plotted versus temperature at 18 different pressures to 135 GPa. The results from run 2 are shown in Fig. 1(b) and span the pressure range 2 - 141 GPa with 17 values. The onset of magnetic ordering is identified by the kink in the $R(T)$ dependence clearly seen near 200 K at 2 GPa, the lowest pressure in each run. The kink in $R(T)$ upon cooling marks the beginning of the suppression of spin-disorder scattering $R_{sd}(T)$ as magnetic ordering sets in.[18] At higher pressures this kink broadens somewhat into a "knee" due to an increasing pressure gradient across the sample, but remains clearly visible to $\sim$115 GPa.

In Fig. 2 selected data from Fig. 1(b) are replotted but shifted vertically for clarity so that no curves intersect. In this graph the red line through the data above the knee gives the phonon contribution $R_{ph}(T)$ to the total measured resistance $R(T)$ estimated in the same manner as in our previous work on Dy,[1] as outlined in the next paragraph. The paramagnetic state of Tb yields the relatively flat region of $R(T)$ at higher temperature where the red (phonon) line overlaps the data. Where the red line begins to separate from the data marks the onset of magnetic ordering in some region of the sample. Because of the pressure gradient across the sample, other regions of the sample will have a lower onset temperature, thus broadening the kink into a knee. The intersection of the phonon resistance (red curve) with the red low-temperature tangent curve defines the temperature $T_{\rm x}$ in Fig. 2.

The total measured resistance is the sum of three terms, $R(T) = R_d + R_{ph}(T) + R_{sd}(T)$, where R_d is the temperature-independent defect resistance, $R_{ph}(T)$ the temperature-dependent phonon resistance, and $R_{sd}(T)$ the temperature-dependent spin-disorder resistance. At temperatures where there is no magnetic ordering in the sample, $R_{sd}(T)$ is independent of temperature. Above the onset temperature of the knee, the temperature dependence of $R(T)$ is, therefore, due solely to that of $R_{ph}(T)$. The temperature dependence of the phonon resistance is visible over the widest temperature range at that pressure (16 GPa in Fig. 2) where the knee begins at the lowest temperature. We extrapolate this dependence to 0 K in the temperature region below the knee to yield the temperature-dependent function $R_{ph}^{16}(T)$, the estimated phonon resistance at 16 GPa in run 2. In run 1 the data at 18 GPa were used in the same way to obtain $R_{ph}^{18}(T)$. Since the functional dependence of $R_{ph}(T)$ on temperatures above $T_{\rm x}$ is seen in Fig. 2 to change only slowly with pressure, we estimate $R_{ph}(T)$ for the other pressures in run 2 by simply multiplying the function $R_{ph}^{16}(T)$ by a "phonon factor" α chosen such that for temperatures above the knee the quantity $R(T) - \alpha R_{ph}^{16}(T)$ becomes *temperature independent* for $T > T_{\rm x}$. The values of α required are listed in Table 1 at all pressures in run 2 to 141 GPa. For pressures of 120 GPa and above, the knee in $R(T)$ apparently begins above 295 K, so that α can no longer be estimated directly from the resistance data. For $P \geq 120$ GPa, therefore, the value $\alpha = 0.41$ is assumed in Table 1 for run 2 and $\alpha = 0.69$ in run 1. This is admittedly an oversimplified way to estimate the phonon contribution, but is superior to the assumption made in an earlier study that for many lanthanides R_{ph} is simply a linear function of temperature to 0 K.[19]

In Fig. 3 the extracted spin-disorder resistance $R_{sd}(T) = R(T) - \alpha R_{ph}^{16}(T) - R_d$ is plotted for pressures 5, 35, and 86 GPa in run 2. The saturation (maximum) value of the so obtained spin-disorder resistance $R_{sd}^{\rm max}$ in the paramagnetic phase at each pressure is listed in Table 1 for run 2. At 86 GPa, for example, $R_{sd}^{\rm max} \simeq 152$ mΩ as seen in Fig. 3. A similar procedure was used to obtain $R_{sd}^{\rm max}$ in run 1.

As illustrated in Fig. 3, the average magnetic order-

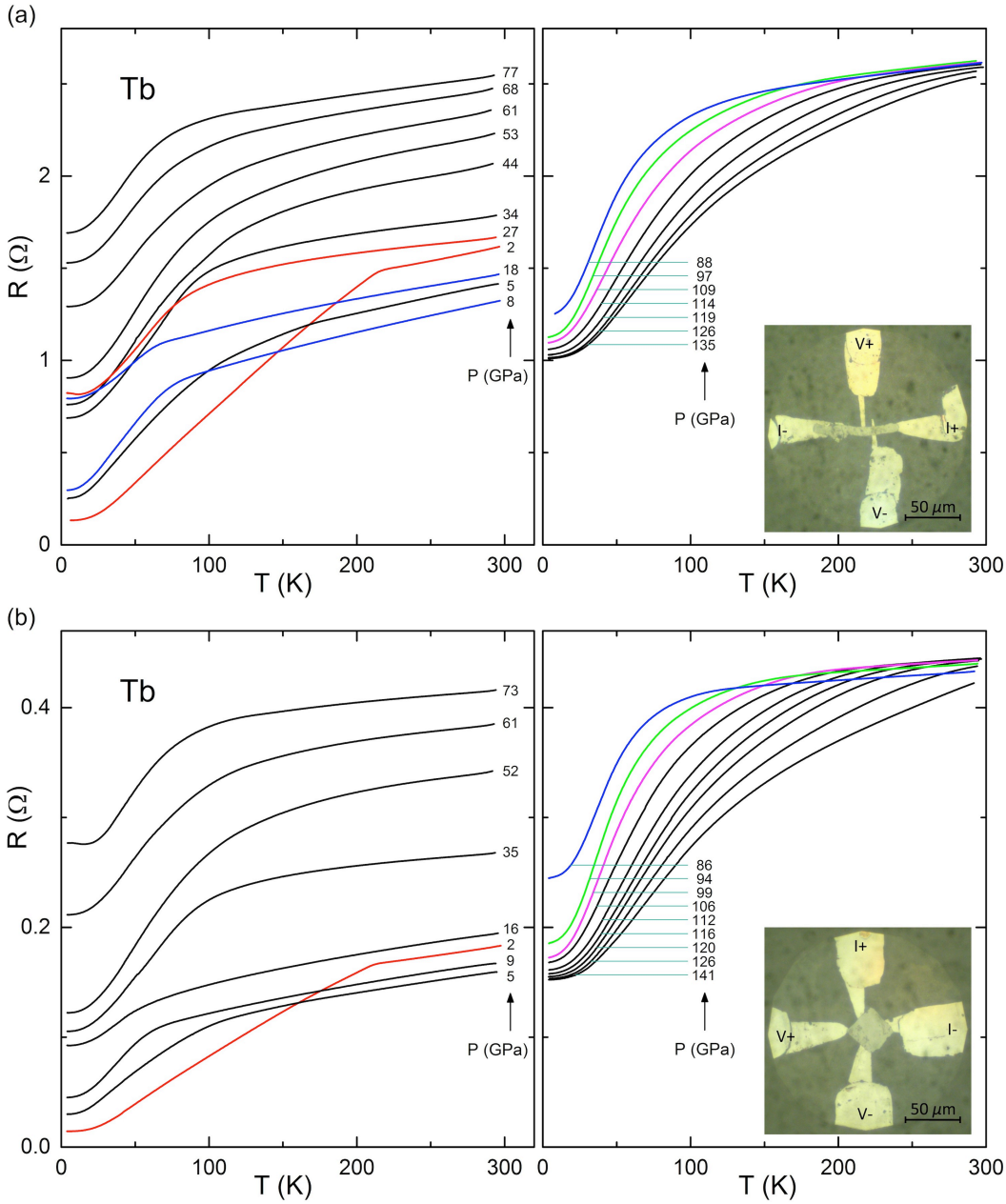

FIG. 1. (color online) Resistance of Tb versus temperature to 295 K for (a) run 1 and (b) run 2 at various pressures. Insets show photograph of (a) elongated sample in run 1 and (b) square sample in run 2.

ing temperature T_o in the Tb sample is estimated from the point of intersection of two straight (red) lines, a horizontal line for temperatures above the onset of the knee, and a line tangent to $R_{sd}(T)$ at lower temperatures. For 86 GPa it is seen that $T_o \simeq 69$ K. This temperature differs by only 1 K from $T_x \simeq 68$ K, the intersection point of the phonon resistance and the low-temperature tangent lines in Fig. 2 at the same pressure. Here we regard T_o to be the average magnetic ordering temperature (in our previous paper on Dy, T_x was used as the ordering temperature[1]). Since the pressure gradient leads to a variation in the value of the magnetic ordering temperature across the sample, we define the "maximum"ordering temperature T_o^{max} as the temperature at which the spin-disorder resistance has decreased by 1%. In Fig. 3 it is seen that $T_o^{max} \simeq 113$ K at 86 GPa. If $dT_o/dP > 0$, T_o^{max} gives the value of the magnetic ordering temperature at the center of the cell (sample) where the pressure is highest. In Fig. 3 it is seen that T_o^{max} lies 44 K higher than T_o at 86 GPa. All values of T_o and T_o^{max} in run 2 are listed in Table 1.

In Fig. 4 T_o and T_o^{max} are plotted versus pressure to 141 GPa for runs 1 and 2 on Tb; values for $P \gtrsim 120$ GPa are estimated using a procedure from Ref. 1, as outlined

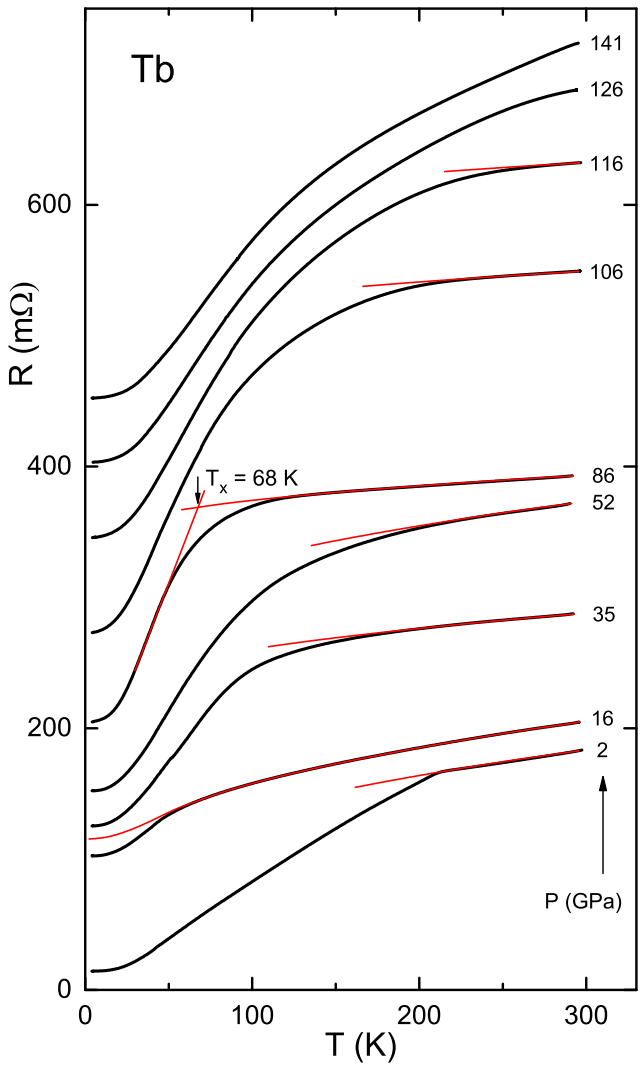

FIG. 2. (color online) Selection of resistance versus temperature curves for Tb from run 2 in Fig. 1(b) where, except at 2 GPa, the curves have been shifted vertically for clarity. Red lines with small positive slope give temperature dependence of phonon resistance for $T \gtrsim T_x$ except at 16 GPa where the phonon resistance extends to 0 K (see text).

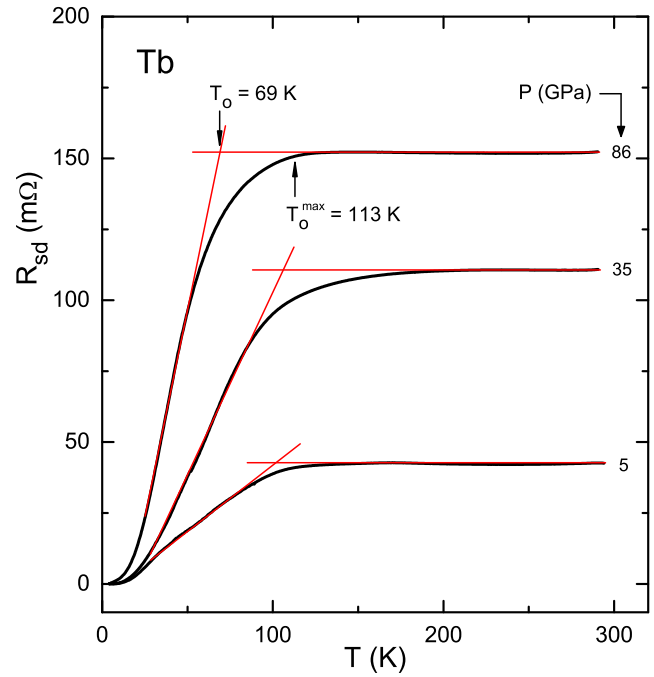

FIG. 3. (color online) Spin-disorder resistance $R_{sd}(T)$ versus temperature at three pressures from run 2. The phonon $R_{ph}(T)$ and defect R_d resistances have been subtracted off. The average magnetic ordering temperature T_o is defined by intersection point of two tangent lines. T_o^{max} gives temperature at which spin-disorder resistance has decreased by 1% (see text).

below. Where they can be compared, the present results are in reasonable agreement with earlier ac magnetic susceptibility measurements of Jackson *et al.* to 6.3 GPa.[5] The pressure dependence $T_o(P)$ at higher pressures is seen to be highly non-monotonic, presumably in response to multiple structural phase transitions[9] (see top of the graph). Note that the phase boundaries were determined from x-ray diffraction studies at ambient temperature and may shift somewhat as the temperature is lowered.

A comparison of $T_o(P)$ for Tb from Fig. 4 to comparable graphs for Gd and Dy in Ref. 1 reveals a remarkable similarity to 53 GPa, the pressure at which the 5% volume collapse in Tb occurs.[9] Also plotted in Fig. 4 are the values of T_o^{max} for Tb given by the upper error bars connected to the values of T_o at each pressure by a light (blue) vertical line. Particularly intriguing is the decrease in T_o following the hR24 to body-centered monoclinic (bcm) transition at 53 GPa,[9] followed by a rapid increase above 80 GPa. In contrast to the findings for $P \leq 53$ GPa, at higher pressures $T_o(P)$ for Tb thus differs significantly from that found earlier for either Gd or Dy.[1] Plotted versus relative volume V/V_o, the increase of T_o above 80 GPa for Tb is found to be much more rapid than the initial decrease of T_o to 6.3 GPa. A similar result was found for Dy.[1] Extrapolating T_o versus V/V_o for Tb linearly to $V/V_o = 0.40$ (141 GPa), yields the values $T_o \approx 250$ K and $T_o^{max} \approx 350$ K.

We now attempt a more quantitative estimate of the pressure dependence of T_o, T_o^{max}, and R_{sd}^{max} in the pressure range above 116 GPa where the onset of the knee appears to lie at or above ambient temperature. We first consider the spin-disorder resistance $R_{sd}(T)$ at pressures $P < 120$ GPa. The first step is to normalize $R_{sd}(T)$ to its value at 295 K, yielding the relative spin-disorder resistance $R_{sd}(T)/R_{sd}^{max}$ plotted versus $\log T$ for data at 106, 112, and 116 GPa in Fig. 5. Since at the higher pressures of 120, 126, and 141 GPa the onset of magnetic ordering appears to lie above the temperature range of the present experiments (295 K), one cannot determine the value of R_{sd} in the paramagnetic phase, nor T_o or T_o^{max}, directly from the resistance data. However, noticing that over much of the temperature range the $R_{sd}(T)$ curves

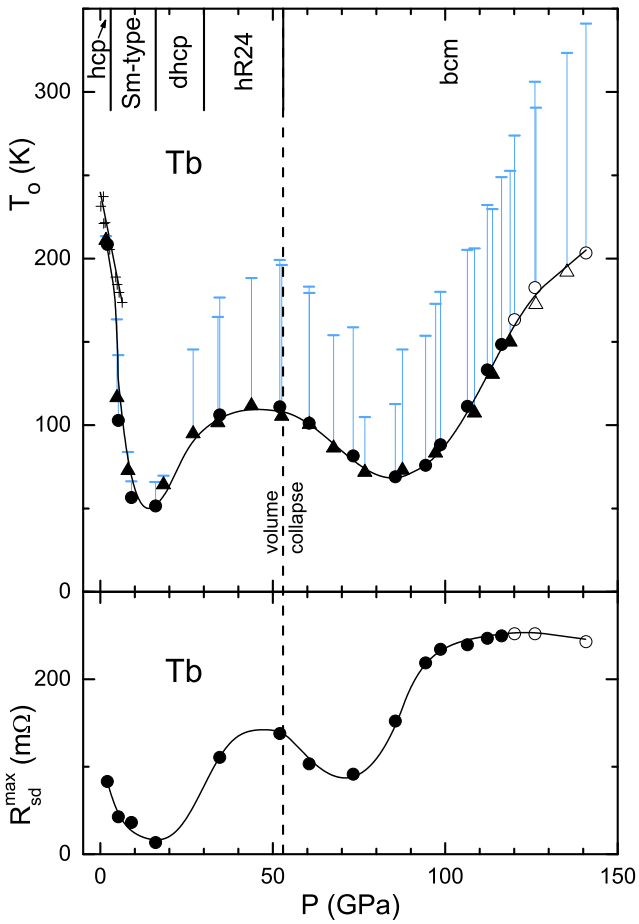

FIG. 4. (color online) Average magnetic ordering temperatures T_o of Tb versus pressure: (+) earlier susceptibility studies to 6.3 GPa with slope $dT_c/dP = -11$ K/GPa;[5] present resistance measurements to 141 GPa from (▲) run 1, (•) run 2. "error bar" connected vertically to each value of T_o gives maximum ordering temperature T_o^{max} at that pressure. Open symbols indicate extrapolated values (see text). Vertical dashed line marks pressure of volume collapse for Tb at 53 GPa. Crystal structures for Tb are given at top of graph.[9] R_{sd}^{max} versus pressure is plotted in lower part of figure from run 2 where it is seen to roughly track T_o(P).

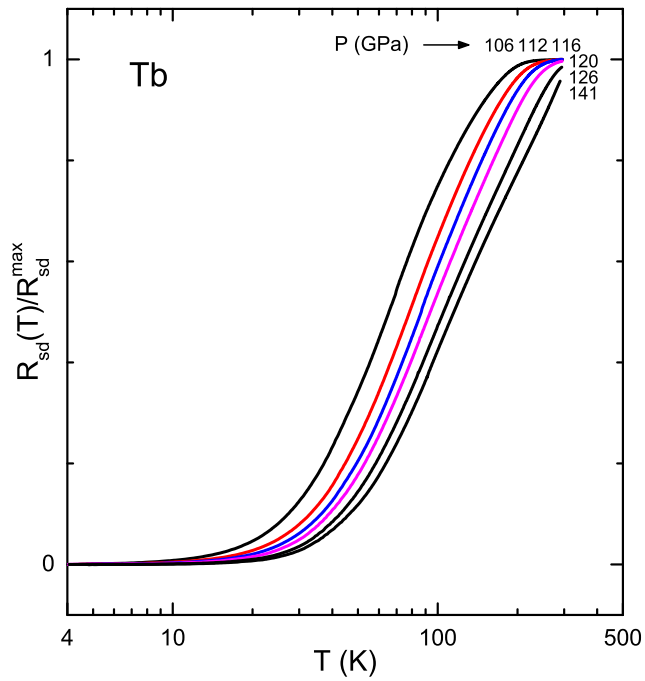

FIG. 5. (color online) Relative spin-disorder resistance $R_{sd}(T)/R_{sd}^{max}$ versus $\log T$. For data at 120, 126, and 141 GPa, R_{sd}^{max} is estimated by adjusting slope of temperature dependence to match that at 106, 112 and 116 GPa (see text). From relative horizontal shifts of the curves the pressure-dependence of the magnetic ordering temperature T_o is estimated for 120, 126, and 141 GPa (see text and Table 1).

for 106, 112, and 116 GPa are approximately parallel on the $\log T$ plot in Fig. 5, we divide the $R_{sd}(T)$ data for $P \geq 120$ GPa by that factor which results in curves parallel to those at the lower pressures, as seen in Fig. 5. We identify this factor as the value of the temperature-independent spin-disorder resistance R_{sd}^{max} in the paramagnetic phase, as listed in Table 1. This procedure is tantamount to assuming that $R_{sd} = R_{sd}(T/T_o)$ for $P \geq 106$ GPa. The change in the value of the magnetic ordering temperatures T_o and T_o^{max} can now be estimated from the shift of the $R_{sd}(T)/R_{sd}^{max}$ curves along the $\log T$ axis. The resulting values of $T_o(P)$ and $T_o^{max}(P)$ are given in Table 1 and in Fig. 4 for all pressures in runs 1 and 2 as the open triangles and circles, respectively. From this analysis we infer that from 116 to 141 GPa the average magnetic ordering temperature T_o has increased from 148 K to 203 K and the maximum ordering temperature T_o^{max} from 249 K to 341 K, values close to those obtained in the linear extrapolation above.

In our previous work on Dy the spin-disorder resistance in the paramagnetic state R_{sd}^{max} was found to approximately track the magnetic ordering temperature T_o as a function of pressure. This same result is seen in Fig. 4 to hold for Tb.

IV. DISCUSSION

We now seek to identify the mechanism(s) responsible for the highly non-monotonic dependence of Tb's magnetic ordering temperature T_o on pressure. First we focus on the pressure region below 53 GPa, the pressure at which Tb suffers a 5% volume collapse. Since the pressure dependence of T_o is so similar for Tb, Gd and Dy in this pressure range, a common mechanism seems likely.

For a conventional lanthanide metal with a stable magnetic moment, the magnetic ordering temperature T_o is expected to scale with the de Gennes factor $(g-1)^2 J_t(J_t+1)$, modulated by the prefactor $J^2 N(E_F)$, where J is the exchange interaction between the $4f$ ion and the conduction electrons, $N(E_F)$ the density of states at the Fermi energy, g the Landé-g factor, and J_t the to-

tal angular momentum quantum number.[2] Since the de Gennes factor is constant under pressure, unless the magnetic state becomes unstable and/or a valence transition occurs, the marked similarity between the highly non-monotonic pressure dependences of T_o for Tb, Dy and Gd to 53 GPa likely originates in the pressure dependence of the prefactor $J^2N(E_F)$, facilitated by a series of nearly identical structural phase transitions in Tb,[9] Dy,[20] and Gd.[21,22] These phase transitions are likely driven by increasing $5d$-electron occupation with pressure.[23] Indeed, electronic structure calculations for Dy suggest that its large negative initial pressure derivative $dT_o/dP \simeq -6.7$ K/GPa results from a strong decrease in $J^2N(E_F)$.[5,24]

We now consider the pressure region $P > 53$ GPa where the pressure dependence $T_o(P)$ for Tb is highly anomalous, deviating markedly from that of the model conventional lanthanide Gd to at least 127 GPa.[1] The absence of magnetic instabilities in Gd, even at extreme pressures, is expected since the local magnetic state of Gd with its half-filled $4f^7$ shell is the most stable of all elements, its $4f^7$ level lying ~ 9 eV below the Fermi level.[26] Why is $T_o(P)$ in Tb anomalous for $P > 53$ GPa? A long-standing strategy[27,28] to probe the magnetic state of a given ion is to alloy this ion in dilute concentration with a host superconductor and determine ΔT_c, the degree of suppression of the host's superconducting transition temperature. Yttrium (Y) is the ideal host superconductor for Tb since the character of its *spd*-electron conduction band closely matches that of the heavy lanthanides, Y even exhibiting nearly the same sequence of structural transitions under pressure.[29] One may thus anticipate that changes in the magnetic state of the Tb ion in the dilute alloy will be mirrored in the changes occurring in the magnetic state of Tb metal.

The efficacy of this strategy is supported by previous studies of the pressure dependences $T_o(P)$ for Dy metal and $\Delta T_c(P)$ for Y(Dy) where both experience a dramatic enhancement beginning just above the pressure of Dy's volume collapse at 73 GPa.[1] It was argued that this anomalous behavior might be the result of the Dy ion exhibiting Kondo physics at elevated pressures where both T_o and ΔT_c are proportional to $|J_-|^2$, the square of the negative exchange parameter leading to the Kondo effect. Dy's volume collapse itself has been suggested to have its origin in the Kondo volume collapse model of Allen and Martin.[25]

Does perhaps the same scenario apply for Tb? In Fig. 6 the pressure dependence of the superconducting transition temperature $T_c(P)$ of the dilute magnetic alloy Y(0.5 at.% Tb) from our previous work[8] is compared to that for elemental Y metal.[30] To a pressure of ∼50 GPa, T_c for the dilute magnetic alloy is seen to increase with pressure at the same rate as for Y. However, just above the pressure of Tb's volume collapse at 53 GPa, the $T_c(P)$ dependence for the alloy begins to pull away rapidly from that of Y, reaching a maximum suppression $\Delta T_c \approx 5$ K at 81 GPa, the highest pressure of the experiment. This strong suppression of Y's superconductivity by dilute Tb ions points to giant Kondo pair breaking, as has previously been observed in high pressure studies on the dilute magnetic alloys La(Ce),[31] La(Pr),[32] Y(Pr),[8,33] and, most recently, Y(Dy).[8] In contrast, as seen in the inset to Fig. 6, $T_c(P)$ for Y(0.5 at.% Gd) does *not* begin to deviate markedly from that of Y metal near 59 GPa, where Gd's volume collapse occurs, but rather faithfully tracks Y's value of T_c to 127 GPa, the maximum pressure of the experiment. Unlike for Tb, the magnetic state for Gd ions in Y remains stable to 127 GPa, so that no Kondo phenomena are expected. We thus suggest that

TABLE I. Values for Tb of the average T_o and maximum T_o^{max} magnetic ordering temperatures, spin-disorder resistance R_{sd}^{max} for $T > T_o^{max}$, and phonon factor α as a function of pressure from runs 1 and 2 (see text).

run	P(GPa)	T_o(K)	T_o^{max}(K)	R_{sd}^{max}(mΩ)	α
1	2	211	214	881	1.16
1	5	117	163	523	1.24
1	8	73	84	380	1.24
1	18	64	70	154	1.0
1	27	95	145	513	0.66
1	34	101	165	721	0.72
1	44	112	188	769	1.02
1	53	105	196	746	1.11
1	61	101	179	518	1.05
1	68	86	154	479	0.89
1	77	72	105	469	0.74
1	88	73	145	1001	0.69
1	97	83	173	1140	0.69
1	109	107	206	1165	0.69
1	114	131	230	1189	0.69
1	119	150	253	1201	0.69
1	126	172	291	1211	0.69
1	135	192	323	1214	0.69
2	2	208	210	83	0.98
2	5	103	142	43	0.98
2	9	57	66	36	0.98
2	16	52	66	13	1.0
2	35	106	177	111	0.58
2	52	111	199	138	0.92
2	61	101	183	103	0.79
2	73	82	159	91	0.56
2	86	69	113	152	0.41
2	94	76	154	219	0.41
2	99	88	180	234	0.41
2	106	111	205	239	0.41
2	112	133	232	247	0.41
2	116	148	249	250	0.41
2	120	163	274	252	0.41
2	126	183	306	252	0.41
2	141	203	341	243	0.41

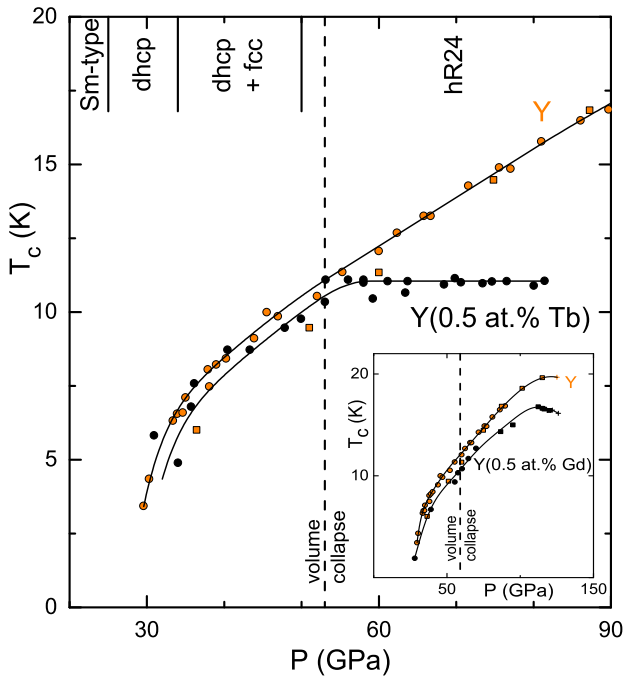

FIG. 6. (color online) T_c versus pressure for Y(0.5 at.% Tb) compared to that for Y, inset showing similar graph for Y(0.5 at.% Gd).[8] Vertical dashed line marks pressure of volume collapse for Tb at 53 GPa[9] and in inset for Gd at 59 GPa.[21,22] At top of graph are crystal structures taken on by superconducting host Y.[29]

the anomalous pressure dependences $T_o(P)$ and $\Delta T_c(P)$ in Tb and Y(Tb) alloy, respectively, have their origin in Kondo physics, as does Tb's volume collapse itself. In support of these suggestions we point out that XANES and XES experiments on Tb to extreme pressure reveal that neither a change in valence nor a magnetic local-itinerant transition occur to a pressure of ~ 70 GPa, well above the volume collapse pressure for Tb at 53 GPa.[8]

Could perhaps an alternative explanation for the anomalously high magnetic ordering temperatures T_o in Tb be the effect of crystalline electric fields? It has been argued that such fields are likely responsible for the significant enhancement of T_o over de Gennes scaling in a series of $R\mathrm{Rh_4B_4}$ compounds, where R is a lanthanide.[34,35] If the magnetic anisotropy is strong, it has been shown[34,35] that the crystal field enhancement can be as large as the factor $3J_t/(J_t+1) = 2.6$ for trivalent Tb where $L = 3$, $S = 3$, and $J_t = 6$. No crystal field effects are possible for Gd since it carries no orbital moment ($L = 0$). The lack of a sharp upturn or other anomalies in T_o and ΔT_c in the pressure region 60 - 127 GPa would be consistent with the certain absence of crystal field effects in Gd. The fact that the pressure dependence of T_o is very similar for both Gd and Tb to 53 GPa indicates that crystal field effects in Tb, if present, are only significant for pressures above 53 GPa where the $T_o(P)$ dependence becomes anomalous. In a crystal field scenario, however, it would be difficult to understand the sharp upturn in the suppression of superconductivity ΔT_c in the dilute magnetic alloy Y(0.5 at.% Tb) for pressures above 53 GPa. This strong suppression of superconductivity points rather to a Kondo physics scenario with strong Kondo pair breaking.

Further experimentation is necessary to unequivocally establish the origin of the anomalous behavior of T_o and ΔT_c in Tb and Y(Tb) alloy, respectively, for the pressure region above 53 GPa. Such experiments could include an extension of the pressure range to 2 Mbar to search for the characteristic "Kondo sinkhole behavior" in $T_c(P)$ observed for Y(Pr),[8,33] La(Ce),[31] and La(Pr)[32] where the T_c-suppression ΔT_c reaches a maximum as the Kondo temperature T_K passes through the experimental temperature range, but falls off again at higher pressures where T_K far exceeds T_c. Inelastic neutron or x-ray scattering studies to extreme pressures would help establish whether crystal-field splittings play a role in the anomalously high values of T_o for Tb.

In summary, measurements of the electrical resistivity of Tb metal to extreme pressures reveal that the magnetic ordering temperature T_o exhibits a highly non-monotonic pressure dependence, appearing to rise for $P > 80$ GPa to anomalously high values. Parallel experiments on Gd and dilute magnetic alloys of Gd and Tb with Y suggest that under extreme pressures Tb is transformed from a magnetically conventional lanthanide into one with an unconventional magnetic state, perhaps involving Kondo physics, with anomalously high values of T_o. In contrast, Gd remains a magnetically conventional lanthanide to pressures of at least 127 GPa.

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