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Stabilization of type-I antiferromagnetism in mixed-valence TmTe at $P = 6\,\mathrm{GPa}$

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Abstract

Previous investigations of magnetic order in the pressure-induced mixed-valence state of TmTe have been extended to a maximum pressure of $P = 6.0\,\mathrm{GPa}$. At this pressure, the same type-I antiferromagnetic state as in TmSe at $P = 0$ is formed below $T_N = 2.55\,\mathrm{K}$, with a comparable magnetic moment of $\mu = 1.9 \pm 0.2\,\mu_B$. The fact that this value is about 50% larger than at 5.4 GPa in the ferromagnetic phase suggests that the change in the magnetic structure is associated with an instability of the Tm 4f moment.

Keywords: Magnetic transition; Mixed valences; Neutron diffraction; High pressure; Pressure effect

TmTe belongs to the family of cubic (NaCl structure) monochalcogenides TmX (X: S, Se, Te), which feature unique magnetic and transport properties reflecting the instability of the Tm 4f shell [1] (for an extensive review on these systems, see Ref. [2]). At ambient pressure, TmTe is a rather conventional divalent magnetic semiconductor, with a band gap of about 350 meV from optical and transport measurements. The $4f^{13}$ level of Tm^{2+} is located inside the gap, slightly below the bottom of the empty 5d6s conduction band. The compound orders antiferromagnetically [AF II, $\boldsymbol{k} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$] below $T_N \approx 0.4\,\mathrm{K}$ [3], a rather low value which can be ascribed to weak superexchange interactions. It was first shown by Jayaraman et al. [4] that the transport gap closes with pressure at a rate of approximately $-210\,\mathrm{meV/GPa}$, leading to a MV state above $P \approx 2\,\mathrm{GPa}$. The question naturally arises of the relationship between this new high-pressure phase and the well-known magnetic MV state occurring in TmSe at ambient pressure. To answer this question, we have investigated the pressure–temperature magnetic-phase diagram of TmTe using neutron diffraction. In the first series of experiments reported in Ref. [5], it was found that at $P = 2.7\,\mathrm{GPa}$, i.e., just after the MV regime sets in, the ordering temperature jumps to $T_C = 14\,\mathrm{K}$ and the structure becomes ferromagnetic with a magnetic moment $\mu_{Tm} = 2.1 \pm 0.1\,\mu_B$. At higher pressure, T_C decreases again to reach 4.5 K at 5.4 GPa, while the ordered moment drops to $1.2\,\mu_B$. This pronounced reduction of the magnetic moment was tentatively ascribed to magnetic fluctuations, both in view of the anomalous scattering that develops in the $\rho(T)$ curves over the same range of pressures [6] and because the ambient-pressure properties of TmSe had been previously interpreted in terms of a Kondo-lattice state [7]. However, despite the rather high pressures achieved in those experiments, evidence for the recovery of the AF I structure corresponding to the magnetic order in TmSe was still eluding, and the pressure variation of μ_{Tm} seemed to extrapolate to zero, rather than to the relatively large moment ($\mu = 1.7 \pm 0.2\,\mu_B$ [8] or $1.9\,\mu_B$ [9]) reported for the latter compound.

This prompted us to extend our neutron measurements to a maximum pressure of 6.0 GPa. The experimental conditions were the same as those previously used

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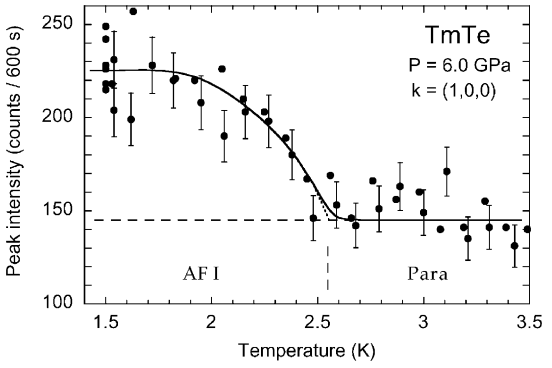

Fig. 1. Temperature dependence of the peak intensity of the 1 0 0 magnetic reflection in TmTe at $P = 6.0\,\mathrm{GPa}$ (measured on cooling).

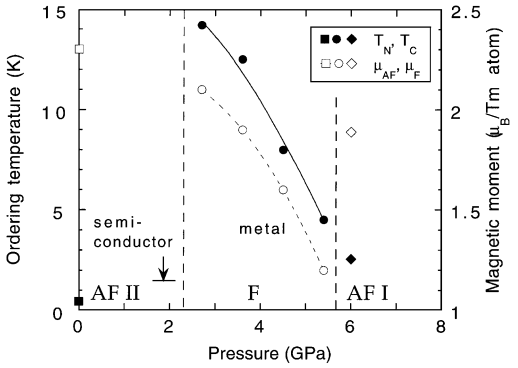

Fig. 2. Pressure–temperature magnetic-phase diagram of TmTe (left scale); closed symbols represent the ordering temperatures for the AF II (■), F (●), and AF I (◆)phases, respectively. Open symbols represent the ordered magnetic moment μ_{Tm} measured at $T_{\min} \approx 1.5\,\mathrm{K}$ (right scale).

and the single-crystal material, prepared by induction-melting of high-purity components inside a vacuum-sealed tungsten crucible, had a lattice constant $a_0 \approx 6.21\,\mathrm{Å}$, characteristic of stoichiometric TmTe. Although the increase in pressure with respect to Ref. [5] was rather limited, the data indicate that the system switches to a completely different type of magnetic structure. No ferromagnetic intensity is found superimposed on the weak 1 1 1-type nuclear peaks down to the lowest temperature but, on the other hand, satellites corresponding to the AF I wavevector $\boldsymbol{k} = (1,0,0)$ are clearly observed. From the temperature dependence of the peak intensity for $\boldsymbol{Q} = (1,0,0)$ Fig. 1, we deduce a Néel temperature of $2.55 \pm 0.10\,\mathrm{K}$. The observation of a finite intensity for the 1 0 0-type reflections implies that the magnetic moment is *not* parallel to $\boldsymbol{k}$ [only the moment component perpendicular to the scattering vector $\boldsymbol{Q} = (1,0,0)$ produces magnetic scattering]. It is consistent with previous results for TmSe, in which the moments were shown to lie along one of the four-fold axes perpendicular to the $\boldsymbol{k}$ vector [8]. Under the same assumption, we can estimate the Tm magnetic moment at 1.5 K to be $\mu_{\mathrm{Tm}} = 1.9 \pm 0.2\,\mu_{\mathrm{B}}$. The calculation takes into account differences in magnetic domain populations due to non-hydrostatic pressure components ($p_x/p_y \approx 2/3; p_z \approx 0$, z being parallel to the axis of the pressure cell), as deduced from the intensities of the 100, 101, and 110 reflections. In TmSe, the Néel temperature at $P = 0$ is $T_{\mathrm{N}} = 3.46\,\mathrm{K}$ in the best stoichiometric samples [7], but it was found to increase rather steeply with pressure [9] and the smaller value found here for TmTe may correspond to TmSe upon a slight expansion of the lattice ("negative pressure").

The Néel temperature or the new AF I phase at 6.0 GPa is plotted on the magnetic phase diagram in Fig. 2, together with the corresponding value for the magnetic moment. The previous neutron data points for the low-pressure AF II phase and the high-pressure F phase are displayed as squares and circles, respectively. The present observation of a magnetic phase transition at 2.55 K for $P = 6.0\,\mathrm{GPa}$ is consistent, within uncertainties in experimental pressure scales, with the resistivity anomaly observed recently at about 2.5 K for $P = 5.5\,\mathrm{Gpa}$ [10]. The steep rise in $\rho(T)$ following the onset of AF I order in the latter measurements provides additional evidence that the electronic and magnetic state achieved in TmTe at these pressures is essentially the same as in TmSe at $P = 0$. However, the sudden positive jump in μ_{Tm} between 5.4 and 6.0 GPa, which is not reflected by to a corresponding increase of the ordering temperature, shows that the change in the magnetic structure with pressure must be accompanied by a more subtle instability of the 4f ground state, and therefore cannot be reduced to a simple competition between different terms in the exchange interactions.

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