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Structurally tuned magnetic properties of $Tb_mRh_nIn_{3m+2n}$ ($m = 1, 2$; $n = 0, 1$) intermetallic antiferromagnets

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Abstract

We report the evolution of the magnetic properties of a new series of intermetallic compounds $Tb_mRh_nIn_{3m+2n}$ ($m = 1, 2; n = 0, 1$) which are structurally related to a class of Ce-based heavy-fermion superconductors (HFS). Measurements of temperature-dependent magnetic susceptibility, specific heat and X-ray resonant magnetic scattering (XRMS) were performed on single crystalline samples of $TbIn_3$, $TbRhIn_5$ and Tb_2RhIn_8. The tetragonal materials $TbRhIn_5$ and Tb_2RhIn_8 both order antiferromagnetically with higher ordering temperatures ($T_N = 45.5$ and 42.8 K, respectively) than their cubic relative $TbIn_3$ ($T_N = 32.7$ K). Their commensurate magnetic structure and the direction of Tb magnetic moments will be discussed in terms of crystalline electrical field (CEF) effects and Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction with effects of magnetic frustration being released by lower dimensionality. A more general picture about the role of CEF in determining the magnetic properties of other R-based members of these series is discussed.
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1. Introduction

The existence of series of structurally related compounds is a great opportunity to explore how the evolution of dimensionality and/or anisotropy effects along the series can affect the ground state of their members. For instance, a very important open issue in condensed matter physics is the relationship between superconductivity and crystal structure. Do certain crystal structures favor superconductivity? The family of highly correlated materials $CeMIn_5$ (the so-called 1-1-5) (M = Rh, Ir, Co) which are tetragonal variants of $CeIn_3$ has allowed further investigation of the possibility of magnetically mediated superconductivity in HFS and its relationship with dimensionality and crystal structures. The $CeRh_{1-x}Ir_xIn_5$ compounds have revealed a linear dependence between T_C and the ratio of the tetragonal lattice parameters c/a indicating that the increase of the quasi-2D character is favoring the unconventional superconductivity (USC) [1,2]. These Ce-based materials belong to the family R_mMIn_{3m+2} (R = rare earth; $m = 1, 2$) for which the tetragonal structures can be viewed as m layers of RIn_3 units stacked sequentially along the c-axis with intervening layers of MIn_2 [3].

As the HFS in this family are presumably magnetically mediated superconductors, to understand f-electron magnetism along these series is an important task. In this regards, the Nd- and Gd-based structurally related compounds have been investigated in detail and their magnetic properties were found to mainly depend on the interplay between CEF effects and the RKKY interaction [4–7]. Furthermore, when comparing the magnetic properties of the tetragonal variants $RMIn_5$

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and R_2MIn_8 with their cubic relatives RIn_3, T_N is significantly enhanced for R = Nd [4] and, as will be showed, also for Tb compounds.

In this work we add new members to these series for which the trends observed in the magnetic properties along these series can be further explored.

2. Experiment

Single crystals of $TbRhIn_5$, Tb_2RhIn_8 and $TbIn_3$ were grown in In flux in the ratio Tb : Rh : In = 1 : 1 : 20, 2:1:8 and 1:20, respectively. Typical crystal sizes were 1 cm× 1 cm × several mm. The tetragonal $Ho_mCo_nGa_{3m+2n}$ ($m = 1,2; n = 1$) structure types and phase purity were confirmed by X-ray powder diffraction, and the crystal orientation was determined by the usual Laue method. Magnetization measurements as a function of temperature were performed using a quantum design (QD) SQUID magnetometer, for specific-heat measurements the QD PPMS small-mass calorimeter was used. XRMS measurements were carried out at the bending magnet beamline XRD2 of the Brazilian Synchrotron Laboratory (LNLS) in Campinas, Brazil.

3. Results and discussion

Fig. 1 presents the temperature dependence of the magnetic susceptibility χ (a) and the specific heat C/T (b) for $TbRhIn_5$, Tb_2RhIn_8 and $TbIn_3$ compounds. For the tetragonal variants, the magnetic susceptibility is higher for the field applied along the *c*-axis in agreement to what was found for all other non-S R-members of these series [4]. For $TbIn_3$ besides the well-defined antiferromagnetic (AFM) transition at $T_N = 32.7$ K, an additional low-T anomaly around 25 K can be seen in both measurements of Fig. 1, probably associated with a secondary magnetic transition within the ordered state. This result is consistent with the data reported by Galera et al. [8] who have studied this compound using neutron diffraction experiments under applied magnetic field and constructed a phase diagram mapping these two magnetic transitions. For the data in Fig. 1(b), all three curves were corrected for the phonon contribution using non-magnetic $YRhIn_5$, Y_2RhIn_8 and YIn_3 for $TbRhIn_5$, Tb_2RhIn_8 and $TbIn_3$, respectively. From both sets of data in Fig. 1, one can observe the increase in the T_N value while going from the $TbIn_3$ to the tetragonal $TbRhIn_5$ and Tb_2RhIn_8. This result is very similar to what was found for the Nd-analogous and it is in contrast to what is seen for the Ce-based compounds [4,6].

In Fig. 2 we show the magnetic structures of $TbRhIn_5$ and Tb_2RhIn_8 determined by magnetic scattering. The magnetic structure for the $TbIn_3$ parent compound is already in the literature [8]. $TbIn_3$ has a complex magnetic phase diagram with two magnetic transitions with different magnetic structures which is reminiscent of the behavior found for $NdIn_3$ [9]. These successive magnetic transitions and complex phase diagrams may be an indicative of competing magnetic interactions and frustration. The magnetic structure of $TbRhIn_5$ was resolved from the commensurate modulation vector $\tau = (\frac{1}{2}0\frac{1}{2})$ and the magnetic moments direction was determined by comparing observed magnetic peak intensities with calculated ones using the resonant cross-section for a dipolar resonance (magnetic cell to the right) [10,11]. For the Tb_2RhIn_8, the same kind of measurements were done, leading to the magnetic structure displayed to the left in Fig. 2, with a modulation vector $\tau = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$. The fact that the tetragonal compounds appear to have a more simple and robust magnetic structure than

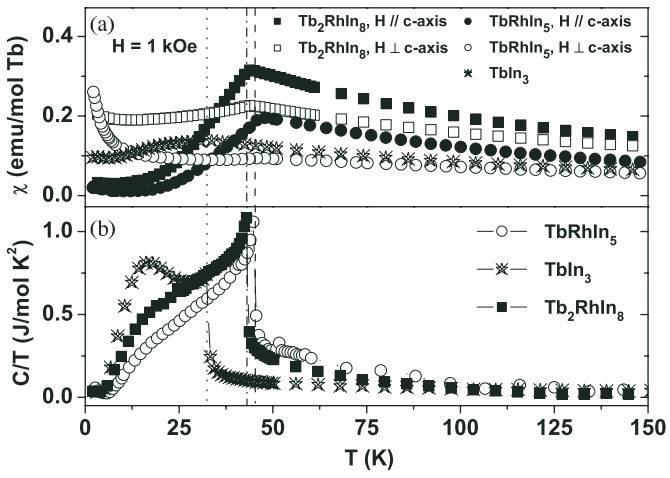

Fig. 1. (a) Temperature dependence of the magnetic susceptibility for applied magnetic field $H = 1$ kOe along *c*-axis (filled symbols) and perpendicular to *c*-axis (open symbols) for $Tb_mRh_nIn_{3m+2n}$ materials for $m = 2$, $n = 1$ (squares); $m = 1$, $n = 1$ (circles) and for $m = 1$, $n = 0$ (crossed symbol). (b) Specific-heat data divided by temperature for $TbRhIn_5$, Tb_2RhIn_8 and $TbIn_3$. The vertical lines (from right to left) indicate the T_N's for each compound in this sequence.

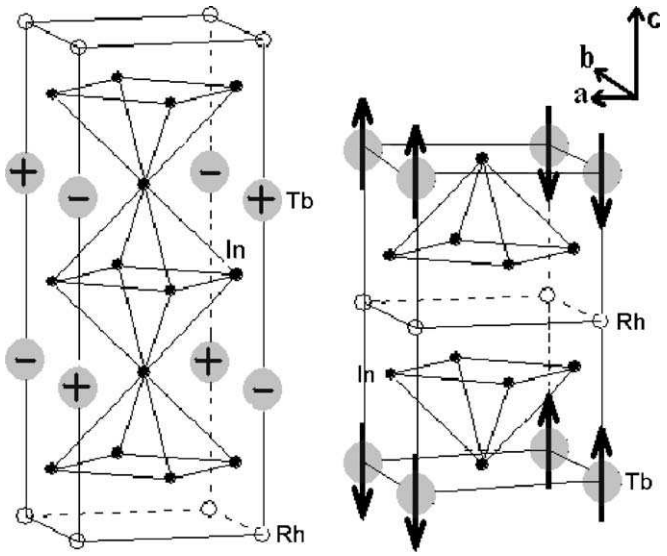

Fig. 2. The magnetic structures of the Tb_2RhIn_8 (left) and the $TbRhIn_5$ (right) as determined using XRMS. Our preliminary analysis show that the 2-1-8 structure follows the (+, −, −, +) ordering, instead of the (+, +, −, −) one, for Tb-atoms in the [0 0 1] direction.

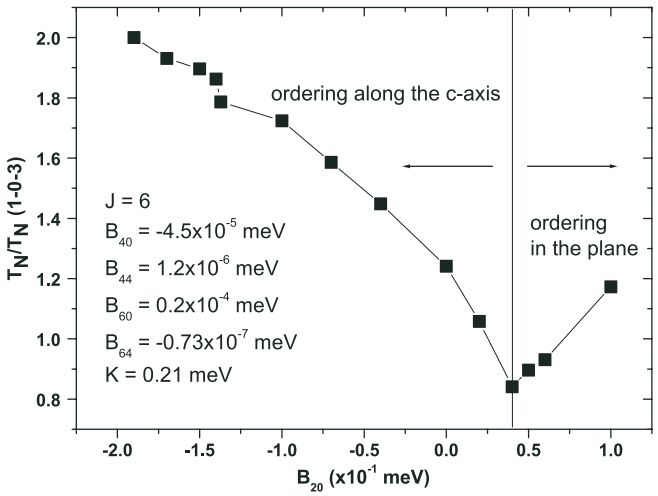

Fig. 3. T_N (Normalized by $T_{N,1\text{-}0\text{-}3} = 32.7$ K) as a function of the crystal field parameter B_{20}. To the left of the vertical line, all B_{20} values correspond to AFM order along c-axis, while to the right are related to an in-plane AFM order. The parameter K is the first-neighbors isotropic exchange interaction.

their cubic relative is also consistent (as in the Nd case) with the fact that ordering along the c-axis comes with a significantly enhanced T_N when compared to that of their cubic RIn_3. All other complementary analysis will be published elsewhere [11].

As CEF defines the 4f multiplet splitting of a given rare earth, the determination of the CEF parameters is an essential step for the understanding of its magnetic properties. Recently, a mean field model including an isotropic first-neighbors RKKY interaction and the tetragonal CEF has been developed [12] in order to study the R_mMIn_{3m+2} (R = Ce, Nd, Gd, Tb; M = Rh, Ir and $m = 1, 2$) series and to explain the ordered states seen experimentally. In Fig. 3 we show a calculation using this model for $J = 6$ of Tb^{3+} from which we extracted the behavior of a normalized order temperature T_N as a function of the tetragonal CEF parameter B_{20} for fixed cubic CEF parameters B_{40}, B_{44}, B_{60}, B_{64} and K (In Fig. 3, $K > 0$ represents an AFM interaction between nearest neighbor spins) and for $-1.9\,\text{meV} \leqslant B_{20} \leqslant 1\,\text{meV}$. Néel Temperature normalization has been made using T_N of $TbIn_3$. The sign and value of B_{20} determines two regions with respect to the direction of the ordered moments. Referring to Figs. 1 and 2, B_{20} for these compounds is negative (a more detailed determination of the CEF parameters for these new Tb-based compounds is in progress and it will be published elsewhere). Further, as one can see in Fig. 3, T_N increases as a function of B_{20} revealing a CEF driven enhancing mechanism of T_N. This result also shows that T_N can be significantly changed when the spins interact with a crystal field, which is in agreement with the results obtained for all non-S R ions and the properties of these new Tb-based series corroborate with the trends observed for the other R compounds [4,12].

4. Conclusions

In summary, we discussed the evolution of the magnetic properties of a new Tb-based series $Tb_mRh_nIn_{3m+2n}$ ($m = 1, 2; n = 0, 1$) intermetallic compounds. The tetragonal materials 1-1-5 and 2-1-8 both order antiferromagnetically with higher ordering temperatures ($T_N = 45.5$ and 42.8 K, respectively) than their cubic relative $TbIn_3$ ($T_N = 32.7$ K). The magnetic structure of $TbRhIn_5$ shows the commensurate wave-vector $\tau = (\frac{1}{2}0\frac{1}{2})$ and the magnetic moments are aligned along the c-axis. In the case of Tb_2RhIn_8, we found a commensurate modulation vector $\tau = (\frac{1}{2}\frac{1}{2}\frac{1}{2})$ with an AFM coupling between the two Tb ions in the same unit cell. Their enhanced T_N and magnetic structure can be accounted by the combination of isotropic magnetic interaction and tetragonal CEF. The results presented here are consistent with a more general picture about CEF driven enhancement or suppression of T_N in these series for other rare earths.

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