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The specific heat of TbPd₃ and ErPd₃ at low temperatures

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Résumé. — Nous avons mesuré les chaleurs spécifiques de TbPd₃ et de ErPd₃ de 2.1 à 8.6 K et de 0.2 à 0.9 K respectivement. La chaleur spécifique de TbPd₃ présente un pic au voisinage de la température 3.75 K. La chaleur spécifique du ErPd₃ ressemble à une anomalie de Schottky avec un maximum $C_v/R \sim 0.74$ qui apparaît aux environs de la température 0.75 K.

Abstract. — The specific heats of TbPd₃ and ErPd₃ have been measured from 2.1 to 8.6 K and from 0.2 to 0.9 K respectively. The specific heat of TbPd₃ shows a magnetic ordering with a typical $\lambda$ shape at 3.75 K. The specific heat of ErPd₃ resembles a Schottky anomaly with a maximum at about 0.75 K with $C_v/R \sim 0.74$.

In the REM₃ phases the strength of the exchange interaction, as shown by the transition temperatures, varies widely. The magnitude of the crystal field shows also substantial variations due to the different charge on the M ion. The symmetry of the crystal field is cubic and its effect can therefore be calculated from the results of Lea et al. [1]. We attempt here to calculate the energy levels of the low lying states for the TbPd₃ and ErPd₃ compounds.

The calorimeter where the experiments were performed, as well as the method of measurement, have been described elsewhere [2].

The samples used in the measurements were prepared by I. R. Harris [3] by arc melting stoichiometric quantities of the pure metals in an argon atmosphere.

The heat capacity of TbPd₃ (figure 1) clearly shows a magnetic transition with a typical $\lambda$ shape at 3.75 K. The heat capacity up to 8.6 K is always larger than that of LaPd₃ [4] (figure 1), revealing that above 3.75 K there is a contribution from the exchange interaction and from the crystal field. We expect that above 5 K the contribution to the heat capacity due to the exchange interaction is negligible and that the extra heat capacity above the heat capacity of LaPd₃ must be explained in terms of the crystal field splittings.

A point charge model calculation with $Z_{tb} = 3$, $Z_{pd} = 0$ predicts that a non-magnetic doublet ($\Gamma_3$) will be the ground state with a triplet ($\Gamma_5^{(3)}$) at 2.2 K, and a singlet ($\Gamma_2$) at 9 K. The other excited states, having much higher energy, need not be considered in the temperature range of this measurement. In the absence of an exchange interaction a Schottky anomaly would occur, and considering simply the two lowest states, $C_v(R)_{\text{max}}$ will be $\sim 0.61$ and $T_{\text{max}} \sim 0.87$ K. Comparing these values with the results above 5 K, we conclude that the point charge model predictions must be rejected. A fitting of the Schottky type above 5 K is very sensitive to the two low lying states and we can only achieve a good fit if $\Gamma_3$ is the ground state and the energy of $\Gamma_5^{(3)}$ is $8 \pm 0.5$ K. Some uncertainty about the energy of $\Gamma_2$ remains because the lattice heat capacity is not accurately known. If we assume that the lattice heat capacity of TbPd₃ is the same as that of LaPd₃, the energy of $\Gamma_2$ will be $60 \pm 10$ K: however a value of $33 \pm 5$ K for $\Gamma_2$ will give a good fit provided a lattice heat capacity similar to that of PrPd₃ [4] is adopted. The inelastic neutron scattering results on TbPd₃ [5] exhibit a transition peak at $\sim 6$ meV. This might well suggest that the energy of $\Gamma_2$ is indeed $\sim 60$ K as predicted above.

The magnetic ordering of TbPd₃ raises the question whether the exchange energy is sufficient to cause the...
states $\Gamma_3$ and $\Gamma_5^{(1)}$ to cross or it merely induces a moment in the $\Gamma_5$ state through its second order term. The crystal field energies suggest that the second possibility is the likely answer.

Calculations of the energies of the levels $\Gamma_3$ and $\Gamma_5^{(1)}$, within a molecular field model, show that for the direction $\langle 100 \rangle$ no magnetic field will make these levels cross, and that $\Gamma_3$ magnetises in second order due to the mixing in the $\langle 100 \rangle$ direction. In the $\langle 111 \rangle$ direction the $\Gamma_3$ state remains degenerate to second order and again the exchange field cannot cause $\Gamma_3$ and $\Gamma_5^{(1)}$ to cross; thus we deduce [6] that ordering will occur in the $\langle 100 \rangle$ direction from the $\Gamma_3$ state.

The specific heat of ErPd$_3$ from 0.2 to 0.9 K (figure 2) resembles a Schottky anomaly with a maximum $C_v/R \sim 0.74$ at about $T_{\text{max}} = 0.75$ K, revealing that magnetic ordering does not occur in ErPd$_3$ above 0.2 K. We can therefore use the point charge model to interpret the experimental results. If we take $Z_{\alpha} = 3$, $Z_{\text{pd}} = 0$, the point charge model predicts that a magnetic doublet $\Gamma_6$ will be the ground state with the four-fold degenerate level $\Gamma_8^{(3)}$ at 0.9 K and the doublet $\Gamma_7$ at 17 K. The two four-fold degenerate levels $\Gamma_8^{(2)}$ and $\Gamma_6^{(1)}$ have energies still higher than $\Gamma_7$. It can be seen from the energy level diagram [1] that $\Gamma_8^{(3)}$ and $\Gamma_6$ cross at $x \sim 0.85$, where $x$ is one of the parameters of the crystal field.

We compare the observed data with the predictions for a two level system, with $g_1/g_0 = 2$, where $g_1$ and $g_0$ are the degeneracies of $\Gamma_8^{(3)}$ and $\Gamma_6$ respectively. We obtain $(C_v/R)_{\text{max}} = 0.74$, which agrees reasonably well with the experiments. This indicates that $\Gamma_6$ is the ground state, since if $\Gamma_8^{(3)}$ and $\Gamma_6$ had crossed, $g_1/g_0$ would be 0.5 and $(C_v/R)_{\text{max}}$ would become 0.24, which would have been too small. On the other hand from $T_{\text{max}} = 0.75$ K we can derive a value of 2.0 K for the energy of the $\Gamma_8^{(3)}$ state.

The agreement between the experimental and theoretical values (figure 2) is reasonable above 0.35 K, especially as the experimental points show a lot of scatter above 0.7 K. Below 0.35 K there is a significant deviation from the theoretical curve. This additional contribution to the heat capacity could well be associated with the short range interactions present above a magnetic ordering temperature.

Values of $x \sim -0.8$ and of the second crystal field parameter $W \sim 0.65$ K and of $x \sim 0.9$ and $W \sim 0.12$ K for ErPd$_3$, would be compatible with our experimental results. In both TbPd$_3$ and ErPd$_3$, the reduced fourth-order crystal field parameter $B_4 \alpha^3 \langle r^4 \rangle / \beta$ [5] is $-0.30 \pm 0.05$ μeV.cm. Here $\alpha$ is the lattice constant [3], $B_4 = W / x$, $\beta$ is a reduced matrix element [7] and $\langle r^4 \rangle$ is a relativistic radial function tabulated by Lewis [8]. The value of this parameter in TbPd$_3$ and ErPd$_3$ agree quite well with those in PrPd$_3$, NdPd$_3$ [5] and YbPd$_3$ [5, 9] suggesting, as pointed out by Furrer et al. [5], that the reduced fourth-order parameter (together with the less reliable reduced sixth-order parameter), can be used to estimate the crystal field splittings in the REPd$_3$ compounds.

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