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Linewidths of TmSe studied by magnetic neutron scattering

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Résumé. — Nous reportons des expériences de diffusion de neutrons sur le composé à valence intermédiaire TmSe. Ses propriétés sont interprétées en supposant l’existence de 50 % Tm$^{3+}$ et 50 % Tm$^{2+}$. À hautes températures ($T > 120$ K) ces deux états sont mélangés de façon incohérente. À basses températures ($T < 75$ K) ils se séparent laissant les ions Tm$^{2+}$ dans un état fondamental magnétique alors que les ions Tm$^{3+}$ participent à un fondamental de type singulet non magnétique.

Abstract. — We report on neutron scattering experiments on the intermediate valence compound TmSe. Its properties are discussed assuming the existence of 50 % Tm$^{3+}$ and 50 % Tm$^{2+}$. At high temperatures ($T > 120$ K) both states are incoherently mixed. At low temperatures ($T < 75$ K) both states seem to separate leaving the Tm$^{2+}$ ions in a magnetic ground state while the Tm$^{3+}$ ions develop a non magnetic singlet ground state.

From lattice parameter — and XPS — measurements TmSe seems to be an intermediate valence system, which however has the particularity to show magnetic ordering effects below 3.5 K. To study the spin dynamics in the ordered and paramagnetic region we performed neutron scattering measurements with energy analysis (TOF [1] and 3-axis method) on TmSe and the isostructural diamagnetic reference compound YSe in the temperature region 1.5-300 K.

Comparing the spectra of both systems magnetic scattering could be clearly detected on the Tm-ions. For $T > 120$ K a single magnetic line is observed with a nearly temperature independent line width of about 6.5 meV. Two lines become discernible when the thermal energy $k_B T$ becomes smaller than 6.5 meV. One of these lines is quasielastic and has a width $\Gamma/2 \approx k_B T$. The second line has been identified as an inelastic transition by a recent neutron scattering experiment performed by us on a 3-axis spectrometer (IN8, I.L.L. Grenoble) operated in the energy loss configuration (see figure 1). Its position shifts from 6 meV at 60 K to 10 meV at lower temperatures ($T \leq 10$ K). Its line width decreases with decreasing temperature. We obtain $\Gamma/2 = 4$ meV at 60 K and 2 meV at 2 K.

We interpret this inelastic line as the transition from a singlet ground state of Tm$^{3+}$ ions to a magnetic excited state, while the narrow quasielastic line is attributed to the existence of the Tm$^{2+}$ ions in TmSe. However an interpretation of the inelastic magnetic spectrum of TmSe cannot be given by simple crystal field theory of Tm$^{3+}$ ions.

For temperatures below $T_N = 3.5$ K no quasielastic scattering is detectable, instead an inelastic magnetic scattering appears at 1 meV with $\Gamma/2 = 0.45$ meV and $\sigma = 4.8$ barn.

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Constant-Q scan ($Q = 1.65$ Å$^{-1}$) on TmSe-powder at 2 K in the neutron energy loss configuration ($k_q = 2.66$ Å$^{-1}$).}
\end{figure}
An additional neutron diffraction experiment at 1.8 K shows magnetic Bragg-peaks, whose angular positions and intensities can be explained by an antiferromagnetic f.c.c. cell and on effective moment of \((2.3 \pm 0.6) \mu_B\).

A consistent explanation of the magnitude of the ordered moment \([2]\), of the specific heat in the AFM-phase \([3a]\), of the saturation magnetization \([3b]\), of the effective moment obtained by the static susceptibility at low and high temperatures \([4]\) and of the neutron scattering cross sections \([1]\) can be given, if one assumes that TmSe consists of 50\% Tm\(^{3+}\) ions which become non magnetic at low temperatures and of 50\% Tm\(^{2+}\) ions which stay magnetic even at low temperatures. The characteristic fluctuation temperature between both configurations is 70 K for \(T > 120\) K, it decreases with decreasing temperature for \(T < 120\) K and is smaller than 5 K in the ordered state.

References

  a) CHOUTEAU, G. et al., p. 493.
  b) Holtzberg, F. et al., p. 487.
[4] We obtained \(P_{\text{eff}}^\text{d} = 3.53 \mu_B\) and \(P_{\text{eff}}^\text{s} = 6.3 \mu_B\), see also \([1]\).