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One impurity effect of Tm in YSe. Kondo effect and Van Vleck behavior

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Résumé. — Des mesures d’aimantation et de résistivité sur le système dilué Tm\textsubscript{y}Y\textsubscript{1−x}Se montrent à la fois des comportements de type Tm\textsuperscript{2+} et Tm\textsuperscript{3+}. Cependant, comme les résultats sont presque linéaires en concentration, ils paraissent refléter les propriétés de chaque site de thulium.

Abstract. — Low temperature magnetization and resistivity measurements in the dilute Tm\textsubscript{y}Y\textsubscript{1−x}Se system show both Tm\textsuperscript{2+} and Tm\textsuperscript{3+} like behavior. However, since the results are nearly linear with concentration, they apparently reflect the properties of each thulium site.

This paper presents a new approach to the TmSe mixed valence problem [1]. The idea is to build up the rather complex properties of the concentrated system starting from the substitution of Tm ions in non magnetic YSe, which has a lattice constant equal to that of stoichiometric TmSe (5.71 Å). We report here on the first step of this study, namely the low concentration single impurity limit.

The inverse susceptibilities of two samples of Tm\textsubscript{x}Y\textsubscript{1−x}Se with x = 0.01 and 0.052 are shown in figure 1 between 4.2 and 300 K. The temperature-independent contribution of the matrix which is responsible for the curvatures can be deduced from a χT vs T plot of the less concentrated sample. Correcting our data for this contribution

\[ χ_0 = 6.52 \times 10^{-5} \text{ emu/mole YSe} \]

results in Curie-Weiss law from which a Curie constant of 3.70 uem/mole Tm is obtained in both samples. It is remarkable that this value is intermediate between those of Tm\textsuperscript{2+} (2.57 uem) and Tm\textsuperscript{3+} (7.13 uem) ions, indicating that the situation in these dilute alloys is very similar to that already encountered in concentrated TmSe; the proportion of Tm\textsuperscript{2+} is about 75 % in the present case. The low temperature inverse susceptibilities (T < 4.2 K) shown in figure 2 can be interpreted in the same model, by assuming both Tm\textsuperscript{2+} and Tm\textsuperscript{3+} to be in their crystal field ground states (Tm\textsuperscript{3+}: Van Vleck, non-magnetic; Tm\textsuperscript{2+}: magnetic).

We have fitted our results below 4.2 K to the law:

\[ χ = \frac{C}{T + θ} + χ_{vv}. \] (1)

The Van Vleck susceptibility \( χ_{vv} \) was obtained from a χT vs. T diagram (figure 2 inset) in the range where
the effect of $\theta$ is negligible. Then, we have plotted $(\chi - \chi_{\text{Curie}})^{-1}$ as a function of temperature to deduce the Curie constant and the Curie temperature (figure 2). The Curie constants per Tm$^{2+}$ are in reasonable agreement with those of the $\Gamma_6$ or $\Gamma_7$ crystal field states of Tm$^{2+}$ (0.67 and 1.10 $\mu$em/mole respectively). The Curie-Weiss temperature can be ascribed to RKKY interactions between Tm$^{2+}$ ions and is actually lower in the less concentrated sample. If our analysis is correct, the very low temperature magnetization should write:

$$M = M^{2+} + M^{3+} = M^{2+} + \chi_{\text{Curie}} H$$

(2)

provided the admixing to the excited states of Tm$^{3+}$ by the external field is not too large. Our data for $T \approx 70$ mK are shown in figure 3. Using the values of $\chi_{\text{Curie}}$ taken from figure 2, we obtain the saturation magnetization of Tm$^{2+}$, in reasonable agreement with those expected (1.4-1.7 $\mu$B/Tm$^{2+}$).

![Fig. 3. Low temperature magnetization of Tm$_x$Y$_{1-x}$Se.](image)

We have measured the resistivities of the samples between 20 mK and 300 K. The two curves shown in figure 4 exhibit minima at 10 K for $x = 0.01$ and 15 K for $x = 0.052$ and a logarithmic slope $dp/d\ln T$ of about $-0.25 \mu\Omega\text{cm/at }\%$ below the temperature of the minimum. A strong negative transverse magnetoresistance is also observed at low temperature. These results provide the first evidence for the Kondo effect of thulium in a dilute system and prove that the samples contain a large amount of magnetic impurities. Below a few tenths of a degree, the resistivity reaches a plateau which we ascribe to RKKY interactions in agreement with the Curie-Weiss temperatures discussed above.

In conclusion, this low temperature study of Tm$_x$Y$_{1-x}$Se is consistent with the assumption that thulium exists in two different valence states, each of which is characterized by a typical set of properties: Curie-Weiss law, saturation magnetization, Kondo effect and RKKY interactions in Tm$^{2+}$; temperature-independent susceptibility, and magnetization proportional to $H$ in Van Vleck Tm$^{3+}$.

However, the fact that we observe mixed valence properties even in the single impurity limit seems to imply that a valence fluctuation on each site occurs rather than a static mixture of Tm$^{2+}$ and Tm$^{3+}$, unless some extrinsic mechanism (defects?) stabilizes a heterogeneous distribution. We thank H. Lilienthal for the high temperature susceptibility measurements.

Reference
