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The effect of the 4f-quadrupole charge distribution on the electrical resistivity of TmSb

N. Hessel Andersen
Physics Laboratory I, H.C. Ørsted Institute, University of Copenhagen, Denmark

and O. Vogt
Laboratorium für Festkörperphysik, ETH, Zürich, F.R.G.

Résumé. — Nous avons étudié les interactions entre électrons de conduction et les électrons localisés 4f dans le composé TmSb par la mesure de la variation en température de la résistivité électrique. On montre que la diffusion due à la distribution de charge quadrupolaire des électrons 4f donne une contribution à la résistivité du même ordre de grandeur que le processus d’échange habituel. On trouve un bon accord entre théorie et expérience quand cette contribution est prise en compte.

Abstract. — The interaction between the conduction electrons and the localized 4f-electrons in TmSb has been investigated by measuring the temperature variation in the electrical resistivity. It is shown that the scattering by the 4f-quadrupole charge distribution in Tm gives a contribution to the resistivity of the same order of magnitude as the usual exchange scattering processes. Good agreement is obtained between theory and experiment when this contribution is taken into account.

The interaction between conduction electrons and 4f-electrons has a special significance in metallic rare earth systems, since the dominating interionic couplings are established indirectly via the conduction electrons. In this paper we show from resistivity investigations on TmSb that, in some rare earth ions, the conduction electron interaction with the 4f-quadrupole charge distribution is as important as the usual exchange interaction.

TmSb is from experimental investigations [1, 2] known to be a crystal field only model system. The same conclusion may be drawn if the de Gennes scaling is applied to the known value of the interionic exchange interaction of the isostructural Tb\(_{0.06}\)Y\(_{0.94}\)Sb system. Both ions have \(J = 6\) and a cubic crystal field which is well described by the fourth order parameter only. The only difference in the crystal field level scheme is a change in the overall energy splitting. With an energy separation between the \(I_1\) (singlet) ground state level and the first excited \(F_4\) (triplet)-level of 25.7 K in TmSb and 14.4 K in Tb\(_{1-x}\)Y\(_x\)Sb, we may conclude that the ratio of interionic exchange to crystal field is the same in TmSb as in Tb\(_{0.06}\)Y\(_{0.94}\)Sb.

It has previously been shown that a single-ion model calculation based on the exchange interaction between the conduction electrons and the 4f-electrons may account very well for the resistivity measurements on Tb\(_{0.06}\)Y\(_{0.94}\)Sb [3]. The expression for this resistivity contribution may be expressed in a condensed form as:

\[
\rho_{\text{ex}} = c \rho_0^0 (g - 1)^2 \text{tr} (PQ),
\]

Here \(\rho_0^0\) is a constant which contains the common properties of the conduction electrons and different rare earth ions, \(c\) is the rare earth concentration in diluted alloys, \(g\) is the Landé factor and the matrices \(P_{ij}\) and \(Q_{ij}\), from which the trace is derived, are defined from the crystal field energies \(E_i\) and states \(|i\rangle\) as:

\[
P_{ij} = \frac{\exp(-E_i/k_BT)}{\sum_l \exp(-E_l/k_BT)} \frac{(E_i - E_j)/k_BT}{1 - \exp(-(E_i - E_j)/k_BT)}
\]

and

\[
Q_{ij} = \langle i | J_z | j \rangle |^2 + \frac{1}{2} \langle i | J_+ | j \rangle |^2 + \frac{1}{2} \langle i | J_- | j \rangle |^2.
\]

In figure 1 we have shown our experimental resistivity data on TmSb and YSb in the temperature range from 1.5 K to 30 K. Since Y\(_3^+\) is non-magnetic, the YSb data should represent the electron-phonon resistivity. The previous results from Tb\(_{1-x}\)Y\(_x\)Sb [4], however, reveal that the interchange of Y with Tb causes almost a doubling in the low temperature electron-phonon resistivity. This enhancement may be qualitatively accounted for as a result of the large mass difference between Y and Tb. In any case we estimate the electron-phonon resistivity to be negligible below 10 K and choose this temperature as a fitting point in the comparison between the theoretical calculations from Eq. (1) and our experimental results. The agreement we obtain is unsatisfactory, especially when the value of the fitting parameter \(\rho_{\text{ex}}^0\) is compared to that of Tb\(_{0.06}\)Y\(_{0.94}\)Sb.
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The theoretical resistivity model for the interaction with the 4f-quadrupole charge distribution may be established from a microscopic effective coulomb interaction by use of the operator equivalent method. The derivation follows the lines leading to Eq. (1) and the result has a similar form:

\[ \rho_{\text{qu}} = c \rho_{\text{qu}}^0 \alpha_j^2 \text{tr} (PO) \]  

(4)

\[ \rho_{\text{qu}}^0 \] is like \( \rho_{\text{ex}}^0 \) a basic constant, \( \alpha_j \) is the Stevens multiplicative factor for the quadrupole operators and the matrix \( O_{ij} \) is defined from the Racah operators \( \tilde{O}_{2m} \) as:

\[ O_{ij} = \sum_{m=-2}^{2} |<i|\tilde{O}_{2m}|j>|^2 \]  

(5)

\( \tilde{O}_{20} = \frac{1}{2}(3J_z^2 - J(J + 1)) \),

\[ \tilde{O}_{2 \pm 1} = \pm \frac{\sqrt{3}}{2}(J_0 J_\pm + J_\pm J_0) , \]

\[ \tilde{O}_{2 \pm 2} = \frac{\sqrt{3}}{2} J_\pm ^2 \].

In figure 1 we have also fitted the theoretical results of Eq. (4) to the experimental data at 10 K. The agreement is still unsatisfactory, but the average sum of the two theoretical curves gives excellent agreement with the experimental data below 10 K and deviates above with a contribution which, as already discussed, we may attribute to the electron-phonon scattering.

In our final fit the value chosen for the parameter \( \rho_{\text{qu}}^0 = 0.58 \, \mu \Omega \text{cm} \) is equal to that obtained for the TbcYl, Sb-system [4]. For the quadrupole contribution we have chosen \( \rho_{\text{qu}}^0 = 9.17 \, \mu \Omega \text{cm} \). Since \( \alpha_j \) is equal for \( \text{Tb}^{3+} \) and \( \text{Tm}^{3+} \) we should estimate from Eq. (4) the same quadrupole contribution in TbSb as in TmSb. The large exchange contribution in TbcYl, Sb (\( g = \frac{3}{2} \) in \( \text{Tb}^{3+} \) and \( g = \frac{7}{6} \) in \( \text{Tm}^{3+} \) ), however, totally dominates the quadrupole term.

We may therefore conclude that the electrical resistivity of TmSb reveals that in some cases the quadrupole scattering is of equal importance as the usual exchange scattering. Among the other rare earth ions, by using the scaling laws in Eq. (1) and Eq. (4) and our values for \( \rho_{\text{qu}}^0 \) and \( \rho_{\text{qu}}^0 \), we may estimate that the quadrupole scattering will dominate in CeSb and YbSb and give significant contributions in PrSb. The significance of the conduction electron interaction with the 4f-quadrupole charge distribution should also be reflected in the interionic couplings. This means that the interionic quadrupole couplings should be large compared to the exchange interaction in YbSb, CeSb, TmSb and PrSb. Although our conclusions are based on experiments on the antimonides we suggest that they might have more general validity.

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References