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INFLUENCE OF THE DIRECT COULOMB INTERACTIONS ON THE RESISTIVITY IN $NdPb_3$

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In order to investigate the influence of the $4f$ electrons on the scattering processes we measured resistivity of the $NdPb_3$ in the temperature range 7–300 K. As a result, we could compare the experimentally determined magnetic contribution to the resistivity with the theoretically calculated ones for the exchange and quadrupolar scattering. The results may confirm that the direct Coulomb interactions between $4f$ and conduction electrons predominate.

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In the REX_3 families of compounds (RE = rare earth, X = Sn, In and Pb) the exchange interaction between the $4f$ shell of rare earth ions and conduction electrons is usually stronger than the direct Coulomb interaction. However, a few exceptions were found. The most spectacular one is the $PrPb_3$ where quadrupolar interactions are strong enough to drive the system to an antiquadrupolar ordering below the temperature $T_Q = 0.35$ K. In $SmIn_3$ and $SmSn_3$ transitions to the antiferromagnetic state are accompanied with three close lying anomalies; at least one of them is believed to be caused by quadrupolar ordering [1].

It is known that in intermetallic RE compounds such as REX_3 the conduction electrons and the localized $4f$ electrons are well distinguished. The influence of the conduction electrons on the $4f$ shell manifests itself through the properties of the crystalline electric field (CEF). The experimentally determined CEF parameters give information on the role of the exchange and direct Coulomb interaction in forming the environment of the $4f$ shell. Results of such investigations for the REX_3 compounds (X = Sn, Pb) were reported by Lethuiller et al. [2]. The CEF parameters obtained from the susceptibility data allowed the authors to conclude that the $4f$ shell is in $NdPb_3$ well localized and the Coulomb contribution to CEF predominates. This allows one also to expect that the direct Coulomb interaction predominates in the scattering processes of the conduction electrons on the $4f$ shells.

The aim of our work was to verify this hypothesis. For this purpose the temperature dependence of the magnetic part of the resistivity of $NdPb_3$ has been investigated.

The resistivity measurements were performed in the temperature range of 7–300 K for the $NdPb_3$ and $LaPb_3$ compounds. The single crystals of these compounds were grown by slow cooling of appropriate melts. Samples had the form of rectangular prisms with dimensions of $0.31 \times 0.53 \times 2.6$ mm^3 and $0.26 \times 0.4 \times 3$ mm^3 for $NdPb_3$ and $LaPb_3$, respectively. In order to apply the standard four probe d.c. method of measurements, the current and voltage wires were soldered to the samples with pure indium.

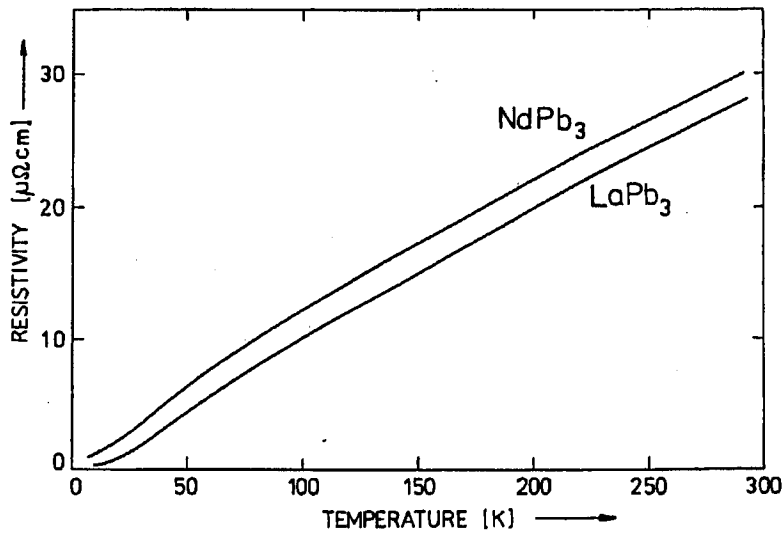

Fig. 1. Temperature dependence of the electrical resistivity for $NdPb_3$ and $LaPb_3$. The dependence for $LaPb_3$ was multiplied by factor 1.2 — see comment in text.

Figure 1 shows the measured resistivity of $NdPb_3$. The resistivity of $LaPb_3$ was used as the reference standard of the phonon contribution. In fact, the latter quantity was multiplied by 1.2 in order to fix well the data at high temperature. Note however that the phonon contribution is in principle not essential at low temperatures where the magnetic contribution predominates.

In order to estimate the magnetic contribution to the resistivity of $NdPb_3$ the validity of Matthiessen rule was assumed

$$\rho_{\mathrm{mag,NdPb_3}} = \rho_{\mathrm{NdPb_3}} - \rho_{\mathrm{imp}} - \rho_{\mathrm{phon}} \tag{1}$$

where $\rho_{\mathrm{NdPb_3}}$ is the total resistivity of $NdPb_3$, ρ_{imp} is the resistivity due to impurities and ρ_{phon} is the phonon resistivity.

The resulting magnetic contribution to the resistivity of $NdPb_3$ is shown in Fig. 2. The curve is normalized to unity at high temperatures and marked as E (experimental). In order to present an apparent contribution of the Coulomb scattering to the magnetic resistivity we made computations of ρ_{mag} using the Hirst formula $\rho_{\mathrm{mag}} \sim \mathrm{Tr}(PQ)$ [3], where the matrix P describes the CEF energy levels influence on the resistivity.

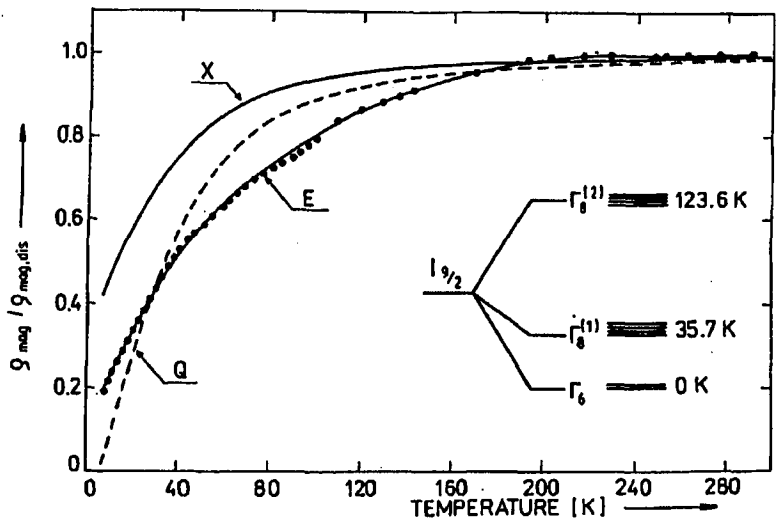

Fig. 2. Temperature variation of the magnetic resistivity found from experimental data — E; computed for quadrupolar scattering — Q; calculated for exchange scattering — X. The used crystal field splitting of the $I_{9/2}$ term of Nd^{3+} is also depicted.

The form of the second matrix Q is dependent on the type of interaction [4], as described by the relevant Hamiltonian. The exchange interaction is described by the Hamiltonian H_{ex}:

$$H_{ex} = -2J_{ex}(g-1)\boldsymbol{J}\cdot\boldsymbol{s} \tag{2}$$

and the quadrupolar interaction is described by the Hamiltonian H_q:

$$H_q = \sum_{k',k,s,\nu',\nu} \sum_{M=-2}^{+2} Q_2 I_2(k',\nu';k,\nu) Y_2^M(\boldsymbol{J}) a^+_{k',s,\nu'} a_{k,s,\nu}$$

The quadrupolar interaction is the leading contribution to the Coulomb interaction when considering the scattering of conduction electrons. In the above formula we denote $\boldsymbol{J}$ and s, respectively, as the angular momentum of $4f$ shell and the spin of a conduction electron.

Y^M are operator equivalents (as in [5]) for $L = 2$. The operator $a_{ks\nu}$ destroys a conduction electron of momentum k and spin s in band ν. $I_2(k',\nu';k,\nu)$ is an integral which we assume to be a constant. Q_2 is proportional to the quadrupolar moment of the fully aligned $4f$ shell.

The results of our computations are depicted in Fig. 2 along with the experimental data. It is clearly seen that the dependence for the exchange scattering (curve X) can hardly approximate the experimental data, especially at the temperature region of 7–50 K, where discrepancy reaches *ca.* 100%. In the same temperature region the difference between the experimental dependence and computed one for quadrupolar scattering (curve Q) is much smaller. Also at higher temperatures ($T > 50$ K) where the both calculated curves deviate from the exper-

imental one, the curve Q is still closer to the experimental points. This indicates that the scattering takes place mainly on quadrupoles.

In our opinion the observed discrepancy between E and Q curves may be caused by two reasons. Firstly, the phonon contribution used in the procedure of the extraction of the magnetic resistivity may be not the proper one. Secondly, the model used for the computations does not take into account interactions between the RE ions as well as the complicated Fermi surface of this material.

Concluding, the predomination of the quadrupolar scattering in $NdPb_3$ confirms the hypothesis that in the case when the direct Coulomb interactions dominate the exchange ones in forming of the CEF, similar effects are observed also in electron scattering processes.

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