

Magnetic structures of $R_3T_4X_4$ (R-rare earth, T = Mn, Cu, Pd, X=Ge, Sn)

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In this work we report the results of crystal and magnetic structure as well as magnetic properties investigations of the intermetallic rare earth $R_3T_4X_4$ (R-rare earth, T=Mn, Cu, Pd; X=Ge, Sn) compounds.

X-ray and neutron diffraction patterns recorded in paramagnetic state confirmed that all of the investigated compounds crystallize in the orthorhombic structure of $Gd_3Cu_4Ge_4$ -type, described by the $Immm$ space group. In this structure the rare earth atoms occupy two sites: 2d ($\frac{1}{2}, 0, \frac{1}{2}$) and 4e ($x, 0, 0$); the transition metal atoms are situated at the 8n ($x, y, 0$) positions and the p-electron atoms are at the 4f ($x, \frac{1}{2}, 0$) and 4h ($0, y, \frac{1}{2}$) positions in the crystal unit cell. The rare earth 2d and 4e sites have different point symmetries: $2mm$ and mmm , respectively. Our aim is to determine the magnetic properties, including the magnetic structures, of these compounds and to obtain information on the magnetic interactions from these data.

Magnetic susceptibility and magnetization were measured in the temperature range 4.2-300 K by the use of a vibrating-sample magnetometer and an ac-susceptometer. Neutron diffractograms collected at different temperatures between 1.5 K and the room temperature, were obtained on the E6 instrument at the BERII reactor, Hahn-Meitner Institut, Berlin and processed with a Rietveld-type programme: FULLPROF.

Temperature dependence of the magnetization and magnetic susceptibility indicate antiferromagnetic properties of these compounds and further phase transitions below the Néel temperatures in some of them.

In the neutron diffraction patterns collected at low temperatures additional peaks of magnetic origin appear. The analysis of topology and intensities of these reflections reveals existence of distinct regions of different magnetic ordering types in different series of the investigated compounds.

In case of the $R_3Mn_4Sn_4$ (R=La, Pr, Nd) compounds the magnetic moment is localized in the manganese and rare earth sublattices. The Mn moments order at T_N of about 300 K and form a noncollinear antiferromagnetic structure. The rare earth moments order independently at low temperatures forming collinear magnetic structures.

In the $R_3Cu_4X_4$ (X=Ge, Sn) and $R_3Pd_4Ge_4$ compounds magnetic moments are localized only on the rare earth atoms. In some of the $R_3T_4Ge_4$ compounds the magnetic moments at the

high-symmetry 2d sites order at relatively high temperatures whereas the magnetic moments at the low-symmetry 4e sites order at lower temperatures.

Phase transitions from commensurate collinear magnetic structures to the incommensurate sine-wave modulated ones are observed in $R_3Cu_4Ge_4$ ($R=Tb$ and Er) below the Néel temperatures.

The stannides exhibit a different scheme of the magnetic ordering. Below T_N in $R_3Cu_4Sn_4$ ($R=Pr$, Tb and Dy) the rare earth moments in both sites order simultaneously and form collinear magnetic structures. More complex, modulated magnetic structure was found for $Ho_3Cu_4Sn_4$. At 1.5 K the Ho moments in both sites form collinear magnetic structure. The moments at the 2d and 4e sites are parallel to the b- and a-axes, respectively. With increasing temperature a change of the magnetic structure is observed.

In $Er_3Cu_4Sn_4$ the Er magnetic moments at the 2d sites form collinear structure whereas those at the 4e sites a modulated one. With increasing temperature the Er moments at the 4e sites disappear.

Large $R^{3+}-R^{3+}$ ion separation suggests that direct magnetic interactions are highly improbable. Thermal dependence of resistivity indicates metallic character of these compounds. These data indicate that stability of the observed magnetic ordering schemes may thus be considered as being due to interactions via conduction electrons (the RKKY model). In this model the Néel temperature is proportional to the de Gennes factor $G=(g_J-1)^2J(J+1)$. However, the Néel temperatures of the investigated compounds do not follow de Gennes scaling. This suggests that the main interaction leading to the magnetic ordering in these systems is not purely of the RKKY-type but is by the crystalline electric field effect, which can significantly influence on the magnitude of the Néel temperature.

Disappearance of the magnetic moment at the 4e sites below the Néel temperature observed in some of the investigated compounds indicates weaker coupling between the magnetic moments at the 2d and 4e sites.

It is likely that the observed moment reduction at the two nonequivalent 2d and 4e sites occupied by the rare earth ions is caused by the CEF behaviour.