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MAGNETIC PROPERTIES OF RCuGe (R = Pr, Nd, Gd, Tb, Dy, Ho, Er) COMPOUNDS

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The magnetic properties including the magnetic structure were determined by magnetometric and neutron diffraction measurements of the polycrystalline samples. RCuGe compounds are antiferromagnets at low temperatures, with T_N ranging from 2.2 K for R = Pr to 17 K for R = Gd. The external magnetic field causes a change in the type of the magnetic ordering from antiferro- to ferromagnetic state. From the neutron diffraction data the magnetic structure of all RCuGe compounds are determined. In the case of ErCuGe below T_N an additional phase transition is observed. The obtained experimental data are discussed in terms of an exchange interaction of the RKKY-type and crystalline electric field.

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1. Introduction

Lanthanide ternary systems with the composition RTX, where R is a lanthanide element, T — transition metal and X = Si, Ge, and Sn have been extensively investigated in the course of recent years [1]. Continuing our studies of the magnetic properties of RTX phases we have turned recently our attention to the RCuGe family. These compounds crystallize in a hexagonal crystal structure [2, 3]. GdCuGe has been known to behave as an antiferromagnet below 17 K [4], while CeCuGe as a ferromagnet below $T_C = 10.2$ K [5]. A neutron diffraction and magnetometric studies have been undertaken for RCuGe (R = Pr, Nd, Gd, Tb, Dy, Ho, and Er) compounds in order to determine their magnetic properties.

2. Experiment and results

Measurements were carried out on polycrystalline samples, as in Ref. [6]. The X-ray and neutron diffraction data indicate that the compounds with R = Pr and Nd have the hexagonal A1B₂-type crystal structure while other compounds

show hexagonal LiGaGe-type with an ordered distribution of Cu and Ge atoms in the unit cell ($P6_3mc$ space group).

Ac magnetic susceptibility vs. temperature plots were recorded using the mutual inductance method at a frequency of 122 Hz. Dc susceptibility data were collected using a vibrating sample magnetometer in magnetic fields up to 50 kOe. A SQUID magnetometer was adopted to perform additional measurements in magnetic fields below 100 Oe.

Neutron diffraction measurements were carried out at the BER II reactor in the Hahn–Meitner Institute in Berlin. The neutron wavelength was 2.40 Å. The observed on the neutron diffraction patterns line intensities were compared with those calculated using the nuclear scattering lengths taken from Ref. [7] and the magnetic form factors for R^{3+} ions from Ref. [8]. The intensities were processed using the Rietveld type Fullprof program [9].

The temperature dependence of the magnetization at low magnetic fields and ac and dc magnetic susceptibilities give maxima typical for the transition from antiferro- to paramagnetic state for RCuGe compounds with $R = Pr, Nd, Gd, Tb, Ho,$ and Er . For DyCuGe below T_C equal to 6 K an increase in the magnetization is observed. In the case of ErCuGe below $T_N = 4.3$ K the additional phase transition at $T_C = 3.7$ K is observed. For all title compounds, above the transition temperatures the reciprocal magnetic susceptibilities obey the Curie–Weiss law with the negative value of the paramagnetic Curie temperature and effective magnetic moments near to the free R^{3+} ion values (see Table).

The magnetization curves as a function of the applied magnetic field at $T = 4.2$ K are typical for paramagnetic materials with $R = Pr$ and Nd , while for $R = Gd, Tb, Dy, Ho,$ and Er compounds are linear up to the critical field H_{cr} and then attain saturation. The values of the critical fields decrease with the rise of the number of $4f$ electrons. The determined values of the magnetic moments at the saturation state are smaller than the free R^{3+} ion values.

TABLE

Magnetic data for RCuGe compounds.

| R | $T_N(K)$ | $\Theta_p(K)$ | $\mu_{eff}(\mu_B)$ | | $\mu(\mu_B)$ | | | m.m.d. | k_x |
|----|----------|---------------|--------------------|-------|--------------|--------|-------|--------|-------|
| | | | exp. | theo. | exp. M | exp. N | theo. | | |
| Pr | 1.8 | -1.0 | 3.56 | 3.58 | 1.5 | * | 3.2 | * | 0.333 |
| Nd | 3.5 | -7.5 | 3.62 | 3.62 | 1.3 | 1.2 | 3.27 | [100] | 0.5 |
| Gd | 17.0 | -0.3 | 7.89 | 7.94 | 6.4 | - | 7.0 | - | - |
| Tb | 11.6 | -21 | 9.8 | 9.72 | 6.6 | 6.5 | 9.0 | [010] | 0.361 |
| Dy | 6.0 | -13 | 10.73 | 10.65 | 8.4 | 7.6 | 10.0 | [010] | 0.338 |
| Ho | 6.1 | -11.6 | 10.58 | 10.61 | 8.5 | 8.2 | 10.0 | [001] | 0.358 |
| Er | 4.3 | -14 | 9.75 | 9.58 | 5.6 | 8.8 | 9.0 | [001] | 0.333 |

(M) magnetometric and (N) neutron diffraction data, * see the text.

m.m.d. — direction of magnetic moment.

The neutron diffraction pattern of $PrCuGe$ at 1.5 K shows the presence of additional peaks resulting from the onset of magnetic order. The magnetic reflections could be indexed assuming a magnetic structure with the propagation vector $k = [\frac{1}{3}, 0, 0]$. Similar reliability factors, R_m are given by two different models of the magnetic structure: either a sine modulated structure with the magnetic moment equal to $1.45(9)\mu_B$ which forms the angle of $46(11)^\circ$ with the c -axis, or helicoidal structure with magnetic moment equal to $1.02(7)\mu_B$, located in the basal plane and making the angle of $29(3)^\circ$ with the a -axis.

At $T = 1.5$ K the magnetic peaks of $NdCuGe$ neutron diffraction pattern could be indexed with the wave vector $k = [\frac{1}{2}, 0, 0]$. The magnetic moment of $1.24(5)\mu_B$ is parallel to the a -axis.

Neutron diffraction patterns of $TbCuGe$, $DyCuGe$ and $HoCuGe$ recorded at low temperatures (2.0, 1.5 and 1.6 K, respectively) and $ErCuGe$ at 3.1 K contain diffraction peaks of magnetic origin which are indexed with the wave vector $k = (\frac{1}{3}, 0, 0)$. The minimum of R_m corresponds to the sine modulated structure. The magnetic moment distribution is described by $gJ \cos(2kr)$, where gJ is the maximum ordered moment per R^{3+} ion, and r is the real space distance. The magnetic moments are parallel to the b -axis [010] for $R = Tb$ and Dy and parallel to c -axis [001] for $R = Ho$ and Er . A different magnetic pattern is observed for $ErCuGe$ at 2.0 K. The magnetic peaks are characterized by two wave vectors: $k_1 = [\frac{1}{3}, 0, 0]$ and $k_2 = [\frac{1}{6}, 0, 0]$. The best agreement with the experimental magnetic intensities gave a sine modulated structure with the magnetic moments parallel to the c -axis.

Figure 1 shows the temperature dependence of the intensities of magnetic peaks for $RCuGe$ ($R = Pr, Nd, Tb, Dy, Ho$, and Er) compounds. The Néel points deduced from neutron diffractograms agree fairly well with those derived from the magnetic measurements. In the case of $ErCuGe$ the change of the magnetic structure was detected at $T_i = 3$ K. The magnetic parameters of all title compounds obtained from neutron diffraction measurements are summarized in Table.

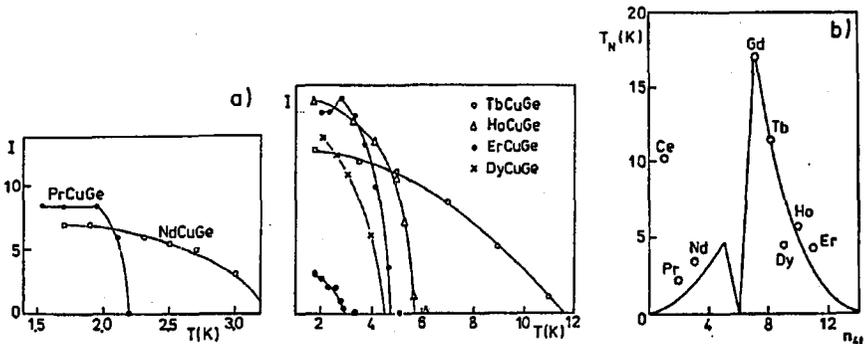


Fig. 1. (a) Temperature dependence of intensities of the magnetic peaks, (b) ordering temperatures $RCuGe$ compounds plotted against the number of $4f$ electrons. The solid line represents the de Gennes function.

3. Discussion

Magnetic order observed in lanthanide intermetallic compounds results from two competing factors: exchange interactions and magnetocrystalline anisotropy. The latter determines the preferred orientation of the magnetic moment which arises from highly localized $4f$ electrons.

The long interatomic distances, *ca.* 3.6 Å along the hexagonal axis and *ca.* 4.3 Å in the basal plane, the sine modulated character of the magnetic ordering and proportionality of the Néel temperatures to the de Gennes function $(g_J - 1)^2 J(J + 1)$ (see Fig. 1) indicate that the RKKY mechanism could be accepted as a major factor responsible for the setting up of the long range magnetic order.

In the RCuGe compounds with heavy lanthanide atoms the change of the direction of the magnetic moment is observed. For the compounds with R = Tb and Dy the magnetic moments are perpendicular to the *c*-axis, while for R = Ho and Er they are parallel to the *c*-axis. The anisotropy of the direction of the magnetic moment results from the crystal electric field (CEF). For the lanthanide atoms in the crystal lattice with hexagonal point symmetry the CEF Hamiltonian is given by

$$H_{\text{CEF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^6 O_6^6,$$

where O_n^m are Stevens operators and B_n^m are CEF parameters defined by Hutchings [10]. In this Hamiltonian, B_2^0 is predominant and the orientation of the magnetic moment in respect to the hexagonal axis can be related to the sign of B_2^0 : the negative sign demonstrates that the magnetic moments are parallel to the hexagonal axis, the positive one — that they are normal to it or make an angle with it. The results indicate that with the rise of the number of $4f$ electrons the change of the sign of this parameter is observed. For the compounds with a hexagonal crystal structure the sign of B_2^0 depends also on the *c/a* ratio [11]. From Table it can be concluded that the *c/a* ratios for heavy lanthanide compounds distinctly change from 1.6968 in DyCuGe to 1.6723 in HoCuGe. This is accompanied by the change of the moment direction from normal to the *c*-axis (DyCuGe) to parallel to it (HoCuGe).

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