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Magnetic Ordering in Boron-Rich Borides TbB_{66} and GdB_{66}

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Magnetic ordering in TbB_{66} and GdB_{66} was investigated at very low temperatures. Measurements of ac susceptibility have shown rather clear features of magnetic ordering below 1 K, at 0.34 K for TbB_{66} and at 0.20 K for GdB_{66}. However, no clear evidence of long range magnetic order was found by neutron scattering experiments. Reasons leading to these observations are discussed.

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1. Introduction

Magnetism of boron-rich borides which contain B_{12} icosahedra is attracting increasing interest as these compounds are magnetically dilute f-electron insulators which show larger magnetic interactions than expected [1]. The lattice constant of RB_{66} borides, where R represents a rare-earth atom, is about $a \approx 23.5$ Å and from the electronic point of view they are insulators/semiconductors (e.g. the room electrical resistivity of GdB_{66} is about 5×10^2 Ω cm [1]). When starting to study their magnetic properties it is useful to compare their properties with RB_{12} and RB_{50} borides, which also contain B_{12} icosahedra. Dodecaborides RB_{12} crystallize in fcc structure ($a \approx 7.5$ Å) and exhibit a variety of physical properties which result mainly from strong electron correlations of the $4f$ shell of rare earth ions. Many of them are metals which order antiferromagnetically below 20 K and the Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect interaction between magnetic ions mediated by conduction electrons is relevant for the observed magnetic order [2–4]. The RB_{50} compounds, on the other hand, are insulators and crystallize in an orthorhombic structure with lattice constants $a \approx 16.6$ Å, $b \approx 17.6$ Å, and $c \approx 9.5$ Å. Despite these compounds representing a rather dilute magnetic system, the magnetic interaction was observed to be surprisingly strong. For TbB_{50} the transition temperature was determined to be $T_N \approx 17$ K [5]. Further investigations of this type of compound indirectly pointed to B_{12} icosahedra to be the mediator of strong magnetic interactions [6], particularly strong covalent bonds within the B_{12} icosahedra are expected to play an important role.

Thus, TbB_{66} and GdB_{66} seem to be suitable candidates for the study of complex interaction mechanisms or of covalent effects in very diluted magnetic systems, because other exchange mechanisms like the RKKY exchange (as there are no conduction electrons available) or the Heisenberg exchange (as the distances between rare earth ions are too large) can well be ruled out in these compounds.

2. Experimental

The investigated TbB_{66} and GdB_{66} samples were prepared by inductive melting. To cool the investigated samples below 1 K, dilution refrigerators in Kosice and Berlin were used. For ac susceptibility measurements the standard method with a primary coil and two contrary wound secondary coils was used. Powder neutron diffraction experiments on TbB_{66} in zero magnetic field were performed at the V1 diffractometer at BENSC, Helmholtz Zentrum Berlin.

3. Results and discussion

3.1 ac susceptibility measurements

The temperature dependence of ac susceptibility of TbB_{66} is shown in Fig. 1. It exhibits a pronounced maximum at 0.34 K pointing to magnetic ordering at this temperature. A similar expressive maximum in ac susceptibility was observed also for GdB_{66}, in this case at 0.20 K (Fig. 2).

3.2 Neutron scattering experiments

To verify the indications for magnetic ordering and to determine the formed magnetic structure, neutron scattering experiments on a powder TbB_{66} sample with ^{11}B boron enrichment were performed at 100 mK. However, no direct/clear evidence of long range magnetic order was found.

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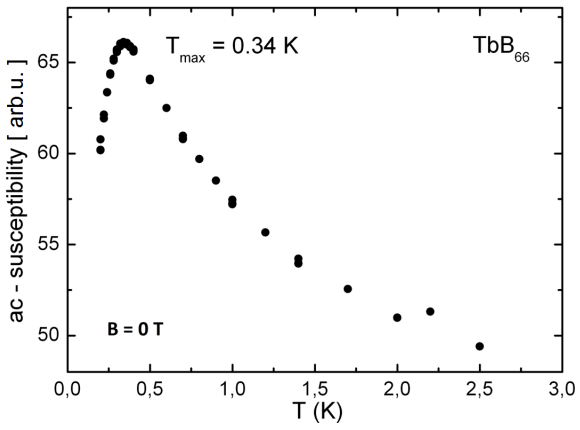

Fig. 1. Temperature dependence of ac susceptibility of TbB_{66} down to 0.2 K.

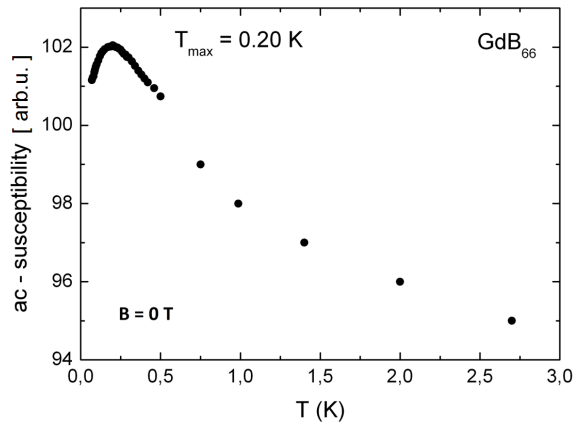

Fig. 2. Temperature dependence of ac susceptibility of GdB_{66} down to 0.1 K.

3.3 Discussion

The received results point to the fact that the magnetic order in RB_{66} compounds is probably of short range nature which is may be caused by the inhomogeneous distribution of rare earth ions in the boron lattice. In this case magnetic short range order appears only in parts of the sample with a higher concentration of rare-earth ions. However, spin glass ordering cannot be ruled out. Further investigations, above all ac susceptibility at different frequencies and heat capacity measurements at very low temperatures are under way to learn more about the process and nature of magnetic ordering in these compounds.

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