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LETTER TO THE EDITOR

Magnetic structure of $GdNi_2B_2C$ as seen from the transferred magnetic hyperfine field at the Ni site

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Abstract. ^{57}Fe Mössbauer spectroscopy has been used to investigate the magnetic structure of $GdNi_2B_2C$ ($T_N \sim 20$ K) doped with 1 at.% ^{57}Fe. Our experiments on polycrystalline $GdNi_2B_2C$ reveal the known spin-rearrangement magnetic phase transition at $T_R \sim 14$ K, which until now has not been observed in polycrystalline samples. A spiral-like spin structure with a variable angle between the moment direction and the *b*- or *c*-axis seems to be the correct model for the spin structure of $GdNi_2B_2C$.

The interplay between superconductivity and magnetism is one of the most attractive features of the rare-earth (RE) nickel borocarbides ($RENi_2B_2C$) [1–6]. The magnetic structure of these compounds usually is not that of a simple collinear antiferromagnet but is rather complex. The knowledge of the detailed magnetic structure in all $RENi_2B_2C$ systems, which seems to be responsible for the suppression of superconductivity [7–11], is important if one is to understand the interplay between superconductivity and magnetism in these systems. $GdNi_2B_2C$ showing magnetic order below $T_N \sim 20$ K [11] is one of the few non-superconducting $RENi_2B_2C$ compounds. Due to the strong neutron absorption of natural Gd, until now there have been no neutron diffraction studies of $GdNi_2B_2C$ made in order to clarify its magnetic structure. However, information on the magnetic structure was recently obtained by resonant and non-resonant x-ray magnetic scattering experiments [13]. From these studies new information was obtained about a second magnetic phase transition below $T_N \sim$ 19.4 K reported previously [12]: the incommensurate antiferromagnetic state, which develops at T_N, has its magnetic wave vector in the basal (a, b)-plane of the tetragonal structure; below $T_R \sim 13.6$ K, however, an ordered component of the moment develops along the *c*-axis.

Having shown that ^{57}Fe Mössbauer effect (ME) spectroscopy can be a powerful tool in the study of the magnetic moment structure in $RENi_2B_2C$ via the Fe(Ni) site [9], we decided to study $GdNi_2B_2C$ using this method. It should be mentioned at this point that this method can be applied quite generally to Fe in systems having the $ThCr_2Si_2$ structure (e.g. $CeRu_2Ge_2$ [14], $RFe_2(Ge, Si)_2$ [15]), wherein the Fe magnetic moment always seems to be negligibly small. We should also mention that there has been a ^{155}Gd ME study of $GdNi_2B_2C$ [16]. In this work, however, no information on the magnetic moment structure beyond the well-known fact that long-range magnetic order sets in below ∼21 K is given.

We now consider the experimental results. Polycrystalline samples of the compound $Gd(^{57}Ni_{0.99}Fe_{0.01})_2B_2C$ were prepared by melting stoichiometric amounts of high-purity

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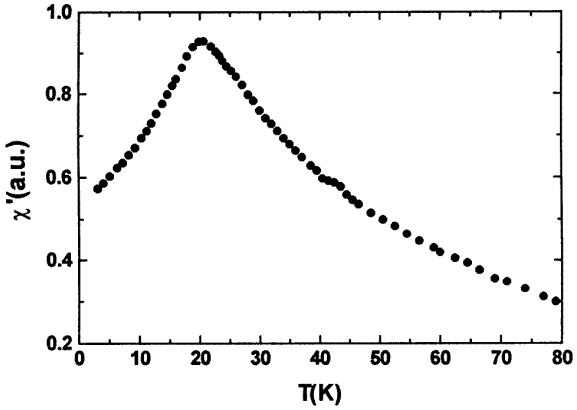

Figure 1. AC susceptibility data for polycrystalline $GdNi_2B_2C$. The transition at $T_N \sim 20$ K is clearly seen, but the second transition at $T_R \sim 14$ K cannot be detected.

elements in an arc furnace at ∼300 mbar of Ar pressure, and subsequently annealing at ∼1000 °C for two days. X-ray diffraction shows that the $ThCr_2Si_2$-like structure is formed as a majority phase with a low level (<10%) of impurity phases, typical for polycrystalline samples of borocarbides. The lattice parameters obtained are consistent with those of the pure $GdNi_2B_2C$ [3].

The samples were first magnetically characterized by AC susceptibility measurements and the result is shown in figure 1. Despite the fact that one can clearly see the magnetic phase transition occurring at $T_N \sim 20 \pm 1$ K, the spin-reorientation transition at $T_R = 13.5$ K, as is well known, cannot be observed via AC susceptibility measurements on powder samples due to the polycrystalline averaging of the magnetic signal; it can only be observed in this way for single crystals [12].

^{57}Fe ME spectra were taken with the $GdNi_2B_2C$ sample in a variable-temperature helium cryostat and the ^{57}Co:Rh source at room temperature, moving in a sinusoidal mode outside of the cryostat. The same sample has been used for AC susceptibility as well as for ^{57}Fe ME measurements. Figure 2 shows the ^{57}Fe ME spectra taken in the temperature region 22 K $\geqslant T \geqslant$ 4.2 K. The spectrum at $T = 22$ K, i.e. above the magnetic ordering temperature T_N, can be fitted with a pure quadrupole doublet (isomer shift IS $= -0.02$ mm s^{-1}, quadrupole splitting $\Delta E_Q = 0.27$ mm s^{-1}, linewidth $\Gamma = 0.25$ mm s^{-1}) and in addition a very weak impurity component (∼5% intensity) probably corresponding to the impurity phase also observed in the x-ray analysis (see above). Lowering the temperature below T_N does not change the ME spectrum until, at around $T \sim 15$ K, a *symmetric* broadening of the spectrum occurs which increases further with decreasing temperature (see figure 2). It seems reasonable to interpret this broadening of the ME spectra at low temperatures as caused by a magnetic hf field, B_{hf}, at the ^{57}Fe nucleus.

In the present case the effect of B_{hf} is, however, quite different from that observed for other $RENi_2B_2C$ compounds: B_{hf} for both $TbNi_2B_2$ and $HoNi_2B_2C$ causes an *asymmetry* in the ME spectra [9], typical for the cases where the magnetic hf and the electrical quadrupole interaction are of the same order of magnitude. In order to explain the symmetric $GdNi_2B_2C$ spectra with a combined magnetic hf and electrical quadrupole interaction, the angle θ between B_{hf} and V_{zz} (the main component of the electric field gradient tensor) is either just the so-called 'magic angle' ($\theta \sim 55°$) or is not well defined, i.e. all angles in the

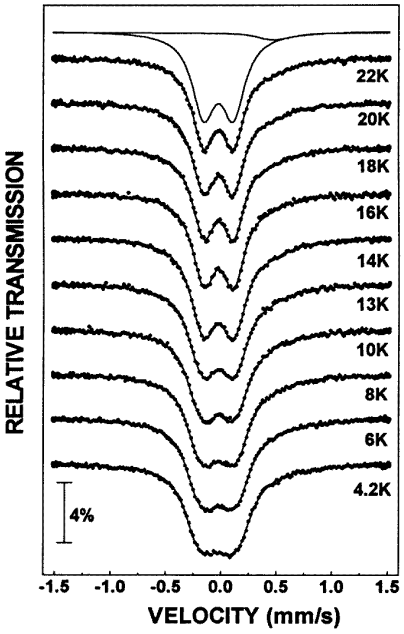

Figure 2. ^{57}Fe Mössbauer spectra of $GdNi_2B_2C$ for the temperature region 22 K $\geqslant T \geqslant$ 4.2 K.

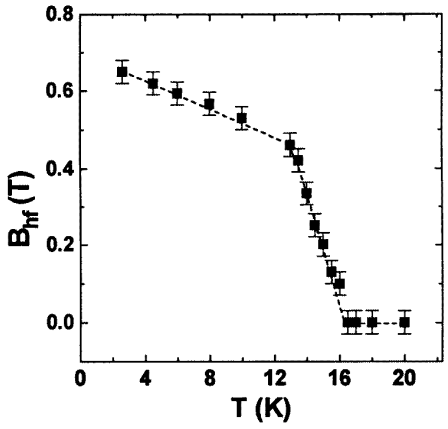

Figure 3. The temperature dependence of the magnetic hf field B_{hf} at the ^{57}Fe nucleus in $GdNi_2B_2C$ obtained using the full Hamiltonian to fit the spectra shown in figure 2. The dependences are the same for the two different models proposed in the text.

range $0 \leqslant \theta \leqslant 90^\circ$ occur with equal probability. It is rather obvious that one cannot distinguish between these two possibilities, especially if B_{hf} is quite small as is the case for the $GdNi_2B_2C$ spectra. Nevertheless we have used two different models for the fits of the ME spectra using the full Hamiltonian (magnetic hf and electrical quadrupole interaction). In both models the values obtained for the 22 K spectrum, i.e. for $T > T_N$, for the electrical quadrupole interaction parameter ΔE_Q and the linewidth Γ have been kept fixed for the analysis for all temperatures below T_N. In model A, the angle θ was a free fitting parameter, while in model B, θ was assumed to take all values in the range $0 \leqslant \theta \leqslant 90^\circ$.

We now discuss the results. Figure 3 shows the results obtained from the two above-described fitting procedures, namely, the magnetic hf field B_{hf} at the ^{57}Fe nucleus as a function of temperature. As expected, the values of $B_{hf}(T)$ are, within the experimental errors, the *same* for both fitting procedures, i.e. we cannot distinguish between the two fitting models. B_{hf} is essentially zero (<0.05 T) not only for $T > T_N \sim 20$ K (as determined by AC susceptibility measurements on the same sample; see above), but also for temperatures which are definitely lower than T_N.

Only for $T \sim 17$ K is a sharp rise of B_{hf} seen, until a value of $B_{hf} \sim 0.5$ T is observed at $T_R \sim 14$ K. Below T_R the increase of B_{hf} with decreasing T is much weaker. From this result one can reach the following straightforward conclusions: the magnetic moment structure in the temperature region $T_N \leqslant T \leqslant 17$ K is that of a collinear antiferromagnet (no transferred hf field at the Fe(Ni) site). Below $T \sim 17$ K this collinear antiferromagnetic structure is continuously changing until at $T_R \sim 14$ K another stable spin structure has been formed.

What can we say about this new spin structure from Mössbauer spectroscopy alone? Since we cannot distinguish between fitting model A and model B, there are two possibilities for the new spin structure below T_R: (A) a fixed angle $\theta \sim 55^\circ$ or (B) a random angle between the moment direction and the *c*-axis (the V_{zz}-direction). Comparing this result with that from magnetic x-ray scattering experiments we suggest that model (b) proposed in reference [13] is the correct model, i.e. that there is a spiral-like structure with a variable angle between the moment direction and the *b*- or *c*-axis. We think that the alternative model (a) proposed in reference [13], namely, a transverse spin modulation, is very unlikely to be appropriate, since we see no reason for the angle between the moment direction and the *c*-axis to be restricted to a value of around 55°.

In conclusion, ^{57}Fe ME experiments on polycrystalline $GdNi_2B_2C$ reveal the known magnetic phase transition (reorientation transition) at $T_R \sim 14$ K. Using macroscopic measuring techniques, e.g. AC susceptibility, this transition cannot be seen in polycrystalline samples but only in single crystals. Our experiments thus show that by using a local measuring technique, e.g. Mössbauer spectroscopy, one can get information usually accessible only via measurements on single crystals. Furthermore, our ME results suggest that there is not a well-defined angle between B_{hf} and V_{zz}. From this we may conclude that the spiral-like spin-structure model proposed in reference [13] for the spin structure below T_R is more favourable than the alternative transverse-wave model, which cannot be distinguished from the former model by magnetic x-ray scattering experiments.

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