Magnetic anisotropy of the spin-antiferromagnet GdNi$_2$B$_2$C probed by high-frequency ESR

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Abstract. A Gd$^{3+}$ electron spin resonance study in a frequency range 10 $- $350 GHz on a single crystal of the spin-only antiferromagnet GdNi$_2$B$_2$C is reported. The Korringa relaxation broadening of the Gd–ESR signal is surprisingly anisotropic implying anisotropic interactions between the localized Gd$^{3+}$ spins and the conduction electrons. In the antiferromagnetic state at $T < T_N = 19.4$ K we observe an out-of-plane anisotropy gap of $\sim 76$ GHz. We also observe an in-plane gap with the same order of magnitude. Numerical analysis of the spin excitation modes in the ordered state assuming classical dipole-dipole interactions agrees with the experiment qualitatively well. However, quantitatively it yields appreciably higher gap values.

1. Introduction

The layered metallic compounds $RNi_2B_2C$ ($R =$ rare earth ions) attracted much attention in the last years for their rich phase diagrams. Superconductivity and antiferromagnetic (AFM) order with commensurate or incommensurate spin structures can be found in this material family. GdNi$_2$B$_2$C is a spin-only magnet owing to the absence of the orbital momentum in the ground state of the Gd$^{3+}$ ($4f^7$) ion, $J = S = 7/2, L = 0$. Therefore, magnetic anisotropy is determined by the dipole-dipole interactions and second-order anisotropic spin-orbit corrections to the magnetic exchange between Gd ions. GdNi$_2$B$_2$C crystallizes in the tetragonal structure with Gd-C layers sandwiched between the Ni-B layers along the tetragonal c-axis. This compound AFM orders at $T_N = 19.4$K with the in-plane (ab) orientation of the ordered moments characterized by a slightly incommensurate ordering wave vector (0.55, 0, 0) [1]. Its phase diagram comprises different magnetic field induced phases in the AFM ordered state [2]. Remarkably, a strong magnetoelastic coupling has been found at $T < T_N$ which yields field induced symmetry-breaking distortions [3, 4]. To obtain further insights into electronic properties and magnetic interactions in GdNi$_2$B$_2$C we have performed measurements of electron spin resonance (ESR) on a high-quality single crystal in the paramagnetic and AFM phases.

2. Experimental

High-frequency (up to $\nu = 350$ GHz) high-field (up to $H = 15$ T) ESR measurements were carried out in the reflection geometry in the temperature range $2 - 230$ K with a home-made spectrometer described in Ref. [5]. Low-frequency ($\nu = 9.5$ GHz) ESR data were obtained with
a standard X-band Bruker spectrometer in the temperature range 16 – 180 K. For υ- and T-dependent measurements the magnetic field \( H \) was set subsequently along and perpendicular to the tetragonal \( c \)-axis of the single crystal, \( H \parallel c \) and \( H \perp c \), respectively.

### 3. Results and Discussion

#### 3.1. Paramagnetic state \( T > T_N \)

In the paramagnetic state the resonating Gd spins yield an ESR absorption signal of the Lorentzian shape at the resonance field \( H_{res} \) corresponding to a \( g \)-factor very close to a spin-only value of 2. Whereas \( H_{res} \) is almost independent of temperature (Fig. 1a, inset), the linewidth \( \Delta \) exhibits a pronounced \( T \)-dependence (Fig. 1a). Above \( \sim 60 \) K it follows a linear \( T \) dependence

\[
\Delta H = a + bT
\]

typical for ESR of localized spins in a metallic matrix. Here \( a \) denotes the part of the linewidth which is due to temperature independent broadening mechanisms. The second term describes the line broadening due the exchange interaction of Gd ions with conduction electrons (CE) which yields a so-called Korringa relaxation of localized spins characterized by a parameter \( b \sim J_{Gd-CE} N(E_F) \) (see, e.g., [6]). Here \( J_{Gd-CE} \) is the exchange coupling constant between the spin of Gd and CE and \( N(E_F) \) is the electronic density of states at the Fermi surface.

The linewidth practically does not depend on the frequency and correspondingly on the field of the measurement (cf. 9.5 GHz and 93 GHz data on Fig. 1a), suggesting that inhomogeneous broadening mechanisms are insignificant. However, \( \Delta H \) appreciably depends on the magnetic field direction. From the high temperature linear fit one obtains \( b^\parallel = 4.85 \times 10^{-4} \) T/K and \( b^\perp = 2.85 \times 10^{-4} \) T/K for \( H \parallel c \) and \( H \perp c \), respectively. Classically the Korringa relaxation is assumed to be isotropic since it implies an isotropic exchange interaction of localized spins with s-like CE [6]. The anisotropic \( b \) can be observed in the systems where local moments interacting...
with CE are substantially anisotropic due to the contribution of the orbital momentum [7]. However, in the case of Gd the orbital moment is absent. The observed anisotropy of the Korringa coefficient $b$ thus give evidence that in GdNi$_2$B$_2$C the CE at the Fermi surface are essentially non-s like. This finding strongly support the band structure calculations in Ref. [8] where a significant contribution of the Ni(3$d$)-derived states at the Fermi level has been found.

Below $T \sim 60$ K the $\Delta H(T)$-dependence deviates from the Korringa behavior. The linewidth increases by a factor of $\sim 3$ while approaching the ordering temperature $T_N$ $\approx$ 19 K. Such critical broadening indicates an enhancement of the short range AFM correlations preceding the magnetic phase transition to the AFM long-range ordered state. At $T_N$ the bulk ESR signal at the low X-band frequency ($\nu$ = 9.5 GHz) vanishes owing to the opening of the AFM gap whereas the high-frequency ESR response can be observed also at $T < T_N$.

3.2. AFM state $T < T_N$

At temperatures below the AFM phase transition at $T_N = 19.4$ K [1] the position of the bulk ESR signal measured at a constant (high) frequency $\nu$ shifts to low resonance fields [9] indicating an opening of an AFM energy gap $\nu_{af}$ for the resonance excitations. A representative $H_{res}(T)$-dependence for $\nu$ = 93 GHz is shown in Fig. 1b. Within the experimental error there is no significant difference of $H_{res}$ between two orientations of the external field, $H \parallel c$ and $H \perp c$. The linewidth for both orientations continues to grow even below $T_N$ (Fig. 1c). However, within the AFM phase the $\Delta H(T)$-dependence exhibits a sharp maximum at $T_R$ $\sim$ 15 K and $T_R^{\perp}$ $\sim$ 11 K for $H \perp c$ and $H \parallel c$, respectively, followed with a decrease of the linewidth at lower temperatures. Comparison with the proposed phase diagram for GdNi$_2$B$_2$C (see Ref. [2]) enables to identify these kinks with the reorientation of the magnetic structure at $T_R < T_N$ from an incommensurate easy-plane (in the basal (ab)-plane) to that with the ordered component of the magnetic moment along the c-axis [1]. The occurrence of the kinks at $T_R$ indicates thus a significant enhancement of spin fluctuations by approaching the reorientation transition which results in a strong broadening of the ESR signal.

The gap for magnetic excitations in the AFM state $\nu_{af}$ occurs due to anisotropy of magnetic

![Figure 2](image-url)
interactions and can be described by the product of the exchange $H_{ex}$ and anisotropy $H_{aniso}$ fields: $\nu^2 = (g\mu_B/h)^2 H_{ex} H_{aniso}$. Here $g$ is the $g$-factor ($\approx 2$ in the case of Gd$^{3+}$), $\mu_B$ is the Bohr magneton and $h$ is the Planck constant. In order to determine the magnitude of $\nu_{af}$ the ESR signals have been measured at a low temperature of 6 K at several excitation frequencies $\nu$. The results are summarized in Fig. 2a. For both magnetic field orientations the $\nu$ vs. $H_{res}$ dependence can be well fitted with a standard relation that neglects a small incommensurability of the spin structure: $\nu^2 = \nu_{af}^2 + (gH_{res}\mu_B/h)^2$. The fit requires a field-dependent gap which has been taken in the form $\nu_{af} = \nu_0(1 - H^2/H_{sat}^2)$ with the saturation field value $H_{sat} = 11.9$ T [2].

Fitting the data for both directions yields the gap $\nu_{af} = 76$ GHz (0.31 meV). One should note that for $H \perp c$ an additional weak excitation with a larger gap of 93 GHz can be resolved in the spectra (Fig. 2a). It arises within the spin-reoriented phase [2] and is still visible at $H$ close to $H_{sat}$. The origin of this excitation requires further clarification.

It is interesting to compare the spin excitations in the AFM state of GdNi$_2$B$_2$C observed by high frequency ESR with a theoretical model [4] assuming that magnetic anisotropy in this compound is given by classical dipole-dipole interactions. The numerical analysis performed using the MCPHASE program package [10] is presented in Fig. 2b,c. Qualitatively the field dependence of the strongest excitation observed in ESR for both field directions is similar to the theoretical calculation. At $H > 8$ T the results even satisfactorily agree quantitatively. At smaller fields the discrepancy increases so that the value of the AFM gap $\nu_{af}(H = 0)$ is overestimated by a factor of 2. This discrepancy can be reduced by changing the value of the isotropic exchange in the model which however affects the positions of the phase boundaries in the magnetic phase diagram [4]. In addition, further sources of magnetic anisotropy may play a role in GdNi$_2$B$_2$C. One can conjecture that, e.g., anisotropic interactions of Gd spins with conduction electrons revealed by ESR in the paramagnetic state may counteract in the AFM state the magnetic anisotropy due to the dipole-dipole interaction.

4. Conclusions

Multi-frequency ESR studies of a single crystal of GdNi$_2$B$_2$C reveal an anisotropic exchange interaction between the localized Gd spins and conduction electrons in the paramagnetic phase. At high excitation frequencies the ESR modes have been also observed in the AFM state. The ESR response is found to be sensitive to the reorientation of the magnetic structure and reveals a strong enhancement of magnetic fluctuations associated with this reorientation. The field dependence of the resonance excitations in the AFM state is similar for both in-plane and out-of-plane orientation of the magnetic field. The measured AFM anisotropy gap differs from that expected for an anisotropy gap given by classical dipole-dipole interactions.

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5. References

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This signal can be attributed to a small amount of an impurity paramagnetic phase in the sample.
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