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METAMAGNETIC STATES IN SINGLE CRYSTAL $HoNi_2B_2C$ AT $T \approx 1.9$ K: TORQUE MAGNETOMETRY STUDY

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Metamagnetic transitions in single-crystal rare-earth nickel borocarbide $HoNi_2B_2C$ have been studied at $T \approx 1.9$ K with a Quantum Design torque magnetometer. With increasing field, transitions to antiferromagnetic, ferrimagnetic, non-collinear and saturated paramagnetic states take place. The critical fields of the transitions depend crucially on the angle θ between applied field and the easy axis [110]. Measurements of torque along the c-axis have been made while changing the angular direction of the magnetic field (parallel to basal tetragonal *ab*-planes) and with changing field at fixed angle over a wide angular range. Two new phase boundaries in the region of the non-collinear phase have been observed, and the direction of the magnetization in this phase has been precisely determined. At low field the antiferromagnetic phase is observed to be multidomain. In the angular range around the hard axis ($-6° \le \phi \le 6°$, where ϕ is the angle between the field and hard axis [100]) the magnetic behavior is found to be "frustrated" with a mixture of phases with different directions of the magnetization.

The rare-earth nickel borocarbides of the type RNi_2B_2C, where R is a rare-earth element have unique superconducting and/or magnetic properties. In this report, a torque magnetometry study of metamagnetic transitions at low temperature $T \approx 1.9$ K in single-crystal borocarbides with R = Ho is presented. Magnetic states in this magnetic superconductor are determined by magnetic moments of Ho ions that lay in the *ab* planes, aligning along easy axis [110], resulting in a high magnetic anisotropy for this compound. In zero field $HoNi_2B_2C$ is a superconductor below $T_c \approx 8.7$ K and antiferromagnetic (AFM) below the Néel temperature, $T_N \approx 5.5$ K. Below 4 K, with increasing magnetic field H perpendicular to the tetragonal c axis, the sequence of transitions from antiferromagnetic (↑↓) to ferrimagnetic (↑↑↓), non-collinear (↑↑→), and ferromagnetic-like (↑↑) states takes place at critical fields H_{m1}, H_{m2}, H_{m3}, respectively [1]. For AFM phase symbol (↑↓) means that the Ho moments are parallel to one of the easy directions (say, [110]) in one half of the *ab* planes; whereas, they are parallel to the opposite direction (that is, to $[\bar{1}\bar{1}0]$) in the other half of the *ab*-planes. For ferrimagnetic phase (↑↑↓), spins in two thirds of the *ab*-planes are parallel to one of the easy axis; whereas, those in the remaining one third are antiparallel to that axis. In a similar way the symbols for other phases are determined. The AFM phase is known to be orthorhombic distorted from tetragonal symmetry [2]. Neutron diffraction studies show that these magnetic states are c-axis modulated except for the non-collinear phase (↑↑→), which is $\underline{a}$-axis modulated [3, 4]. The critical fields of metamagnetic transitions depend strongly on the angle θ between H and the nearest easy axis [110] or on the angle ϕ between H and the nearest hard axis [100]. The known theoretical models [5, 6] can provide an explanation for behavior of all phases except the non-collinear one. Generally, a number of important issues in this matter remain open.

A PPMS Model 550 Torque Magnetometer (Quantum Design) was used to study the angular dependences of metamagnetic transitions. It measures the torque $\vec{\tau} = \vec{M} \times \vec{H}$, so that $\tau = MH\sin(\beta)$, where β is the angle between the external magnetic field and the magnetization. A small, about 0.15 mg, single-crystal rectangular plate of $HoNi_2B_2C$ was used for this study (the sample was grown by P. C. Canfield, Ames Laboratory, Iowa). The torque measurements were made under changes of magnetic field for different constant angles, or changes of angular direction of the magnetic field for different constant magnetic fields. Examples of the magnetic-field and angular dependences of the torque are shown in Figs. 1 and 2, where transitions manifest themselves as sharp changes of the torque at critical fields (or corresponding angles). An enlarged portion of the angular phase diagram obtained for first two transitions is shown in Fig. 3. The total phase diagram is represented in Fig. 4.

Angular diagram (Figs. 3 and 4) appears to be periodic (with period of 90°). This implies that low-temperature orthorhombic distorsions of the tetragonal lattice of $HoNi_2B_2C$ do not cause an appreciable disturbance of angular symmetry of the metamagnetic transitions. Due to limitation in text space, we will mention only the most important and new features of the metamagnetic states, found in this study.

A. The magnetization of the antiferromagnetic (↑↓) phase must be equal to zero, and the same should be expected for the torque (at $H < H_{m1}$). The experimental ev-

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idence is inconsistent with this (Fig. 1). The magnitude $\tau(H)/H$ is nonzero below H_{m1}. Moreover, the $\tau(H)/H$ curves also show appreciable field dependence with hysteretic behavior below 0.1 T. The nonzero absolute value of $\tau(H)/H = M\sin(\beta)$ implies that $M \neq 0$ as well. This is possible for the AFM state if a multidomain AFM structure exists. This may result from the availability of four (or at least two) equivalent easy ⟨110⟩ directions in $HoNi_2B_2C$. Then, on cooling below the Néel temperature, domains can easily appear. The low-temperature orthorhombic distortions in borocarbides [2] could facilitate this process since a shortening of the crystal lattice take place along the two equivalent easy directions. When multidomain (or at least two-domain) AFM structures exist, the magnetic moments of the domains may not be completely compensated, and the torque can be nonzero. It should be noted that the torque hysteresis in the AFM phase takes place in the superconducting state. In this case, the nonzero torque and the hysteresis in the low-field range (Fig. 1) may also be related to trapped flux generated on passing through the critical field.

B. According to Ref. 1, for small deviations of the magnetic field from an easy axis ⟨110⟩ ($-6° \le \theta \le 6°$), the (↑↓) – (↑↑↓) – (↑↑) sequence of metamagnetic transitions takes place. Here, the transition to the non-collinear (↑↑→) phase is omitted. This is in disagreement with theory [5], which supposes that this sequence of transitions is possible at $\theta = 0$ only. An analysis of the magnetic-field dependences of the torque for different angles, including angles close to $\theta = 0$, leads to the conclusion that the angular range for this sequence of transitions is far less ($-1° \le \theta \le 1°$) than that indicated in Ref. 1. Thus, torque results (see also Fig. 4) can be considered to support the assertion in [5] that the sequence of transitions (↑↓) – (↑↑↓) – (↑↑) occurs only for $\theta = 0$.

C. It can be seen in Fig. 1 that for some angle the transitions at H_{m2}, (↑↑↓) – (↑↑), and at H_{m3}, (↑↑→) – (↑↑), show a change of sign of the torque, which clearly indicate a change of direction of the net magnetization from one side of the applied field to the other. According to the model [5], in the phase (↑↑→) the magnetization is tilted by an angle $\Phi = \arctan(1/2) \approx 26.6°$ to the easy ⟨110⟩ axis closest to the magnetic field. This implies that, for example, the angle β in torque $\tau = MH\sin(\beta)$ is expected to change 26.6° at (↑↑↓)–(↑↑→) transition. The angles β and θ are identical in the ferrimagnetic phase. The relation $\beta = \theta - \Phi$ should hold after the transition. A change in sign of the torque at the transition is expected for field directions, for which $|\theta| < \Phi$ and is found (Fig. 1). This magnetization rotation at the transition also causes corresponding features in angular dependences of torque (not shown). The torque measurements permit a precise determination of Φ. In particular, for fields 1.1 and 1.2 T this angle is about 23°, which is close to but smaller than theoretical value of Φ. For fields $H = 1.4$ T and 1.6 T this angle is about 22° and 20°, respectively, indicating that Φ depends on the applied field. Since existing theories [5, 6] are not able to describe the non-collinear (↑↑→)

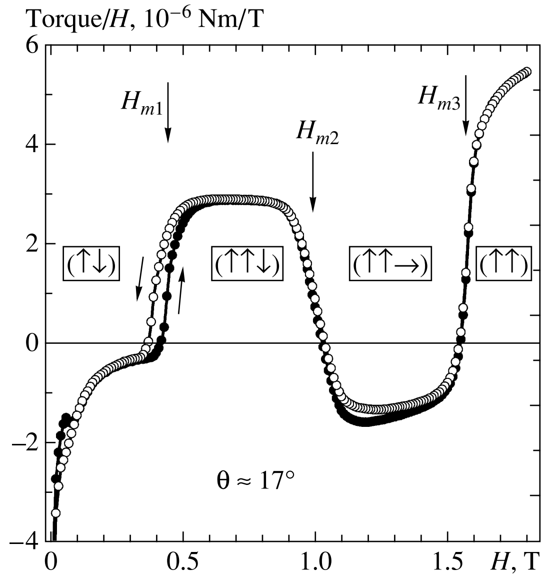

Fig. 1. Field dependence $\tau(H)/H$, recorded with increasing and decreasing magnetic field H at $\theta \approx 17°$. Positions of the critical fields H_{m1}, H_{m2}, H_{m3} are shown by arrows. The symbols (↑↓), (↑↑↓), (↑↑→), and (↑↑) show areas of different magnetic phases.

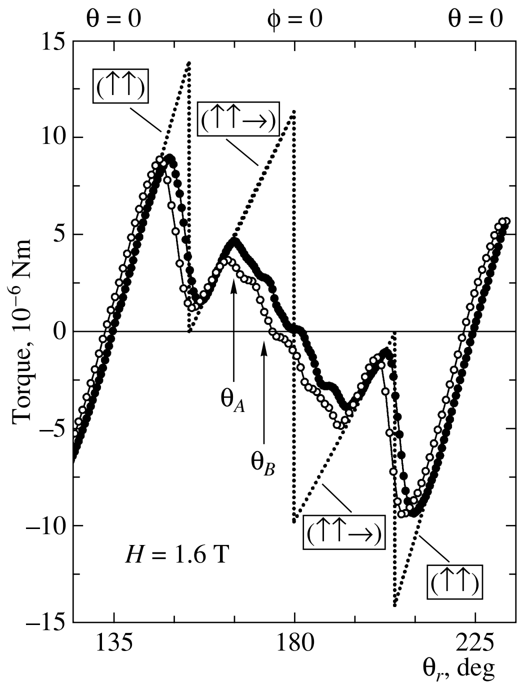

Fig. 2. Angular dependence of torque at $H = 1.6$ T. θ_r is the rotator angle. $\theta = 0$ and $\phi = 0$ correspond to easy [110] and hard [100] axes, respectively. Filled and empty circles represent increasing and decreasing angle, respectively. Dashed lines show the expected angular dependence of the torque in the angular areas of the ferromagnetic-like (↑↓) and non-collinear (↑↑→) phases with $\beta = \theta$ for the (↑↓) phase and $\beta = \theta - \Phi$ for the (↑↑→) phase over the predicted range of existence of these phases (β is the angle between the field and the magnetization, Φ is angle between magnetization and an easy axis in the (↑↑→) phase). θ_A and θ_A indicate positions of drastic changes in the torque corresponding to two new phase boundaries represented by the lines H_A and H_B in Fig. 4. Increased angular hysteresis is evident around the hard axis.

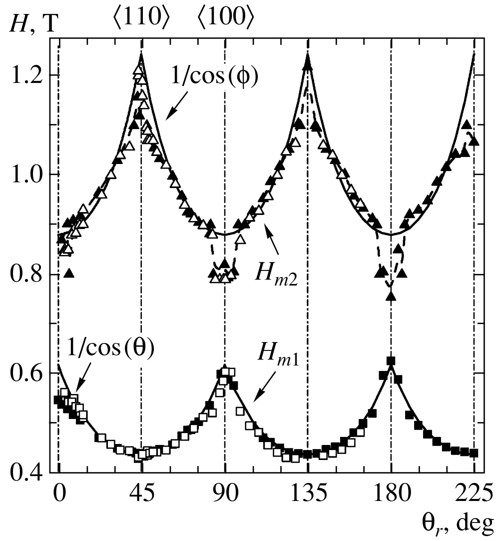

Fig. 3. Portion of phase diagram presenting the first two transitions. Solid curve H_{m1} represents $H_{m1}(\theta) = H_{m1}(0)/\cos(\theta)$ with $H_{m1}(0) = 0.437$ T for the [(↑↓)–(↑↑↓)] transition (θ is the angle between the field and the nearest easy axis). Curve H_{m2} represents $H_{m2}(\theta) = H_{m2}(0)/\cos(\phi)$ with $H_{m2}(0) = 0.88$ T for the [(↑↑↓)–(↑↑→)] transition (ϕ is the angle between field and nearest hard axis).

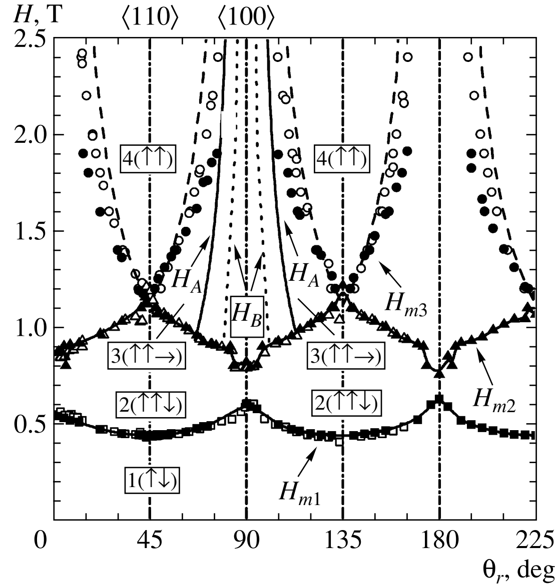

Fig. 4. The angular phase diagram of metamagnetic states in $HoNi_2B_2C$ at $T = 1.9$ K obtained in this study from torque measurements. Only increasing field data are shown. θ_r is angle on the sample rotator. $\theta_r = 45°$, 135° and 225° correspond to different ⟨110⟩ easy axes to one degree accuracy. Symbols for different magnetic states and corresponding critical fields are the same as in Fig. 1. Easy ⟨110⟩ and hard ⟨100⟩ axes are marked in one quadrant. Dashed lines in the upper part represent the dependence $H_{m3}(\phi) = H_{m3}(0)/\sin(\phi)$ according to models of Refs. [4, 5]. Filled and empty points are obtained with the low- and high-moment torque chips, respectively. Solid and dotted lines (H_A and H_B) shown in only one quadrant correspond to new phase boundaries revealed here. Filled and empty squares, triangles and circles represent results of measurements with low- and high-moment torque chips, respectively).

phase properly, this new data can be helpful for understanding its nature.

D. The angular behavior of H_{m2} near the hard axis ⟨100⟩ ($-6° \le \phi \le 6°$) does not follow the theoretical relation $H_{m2}(\theta) = H_{m2}(0)/\cos(\phi)$ [5, 6] so that the H_{m2} values in this region are smaller than predicted (Fig. 3). Also, in this angular range the first (↑↓) – (↑↑↓) and second (↑↑↓) – (↑↑→) metamagnetic transitions almost merge together. This "frustrated" behavior of the magnetic system shows itself also in the angular torque dependences near the hard axis in the field region of the non-collinear phase (Fig. 2). It is seen that the torque goes to zero as the direction of the magnetic field approaches $\phi = 0$, which is in sharp contrast to the picture expected from models [5, 6] (dashed lines in Fig. 2). Two critical angles, θ_A and θ_B, indicate positions of two new phase boundaries, revealed in this study. Both critical fields, H_A and H_B, corresponding to these angles, behave as $1/\sin(\phi)$ (the same as critical field H_{m3}) (Fig. 4). We have called the phase between angles θ_A and θ_B the intermediate state. The angle θ_B determines a transition to another new phase which we have called border state. This "frustrated" phase is possibly a mixed (two-domain) state. At $\phi = 0$ directions of the domain magnetization are symmetric relative to a hard axis, so that total magnetization is along the hard axis giving zero torque. A similar "frustrated" behavior near the hard axis was found in the region of existence of the ferrimagnetic (↑↑↓) phase. In this case it may be associated with the so-called C6 (↑↓↑→←→) phase, predicted in [6], for which the net magnetization is along the hard axis that can also cause torque to be zero at $\phi = 0$.

In conclusion, these torque measurements clarify some important features of these metamagnetic states and indicate new phase states in $HoNi_2B_2C$. Work is supported by the Robert A. Welch Foundation (Grant A-0514) and NSF (DMR-0315476, DMR-0422949).

PREFERENCES

1. *Canfield P.C., Bud'ko S.L., Cho B.K. et al.* // Phys. Rev. B. 1997. V. 55. P. 970.
2. *Kreyssig A., Loewenhaupt M., Freudenberger J. et al.* // J. Appl. Phys. 1999. V. 85. P. 6058.
3. *Campbell A.J., McK Paul D., McIntyre G.J.* // Phys. Rev. B. 2000. V. 61. P. 5872.
4. *Detlefs C., Bourdarot F., Burlet P. et al.* // Ibid. P. R14916.
5. *Kalatsky V.A., Pokrovsky V.L.* // Phys. Rev. B. 1998. V. 57. P. 5485.
6. *Amici A., Thalmeier P.* // Ibid. P. 10684.