The complex magnetic transitions and superconductivity of HoNi$_2$B$_2$C were studied via the dependence of the heat capacity on temperature and in-plane field angle. We provide an extended, comprehensive magnetic phase diagram for $B \parallel \{100\}$ and $B \parallel \{110\}$ based on the thermodynamic measurements. Three magnetic transitions and the superconducting transition were clearly observed. The 5.2 K transition ($T_N$) shows a hysteresis with temperature, indicating the first order nature of the transition at $B = 0$ T. The 6 K transition ($T_M$), namely the onset of the long-range ordering, displays a dramatic in-plane anisotropy: $T_M$ increases with increasing magnetic field for $B \parallel \{100\}$ while it decreases with increasing field for $B \parallel \{110\}$. The anomalous anisotropy in $T_M$ indicates that the transition is related to the a-axis spiral structure. The 5.5 K transition ($T^*$) shows similar behavior to the 5.2 K transition, i.e., a small in-plane anisotropy and scaling with Ising model. This last transition is ascribed to the change from a' dominant phase to c' dominant phase.

I. INTRODUCTION

An understanding of the interplay between magnetism and superconductivity has been an area of intensive research because of their seemingly antagonistic tendencies. The limited availability of suitable examples and the low magnetic transition temperatures of those that exist have made the studies very difficult experimentally. The recently found RNi$_2$B$_2$C family where R is rare-earth element offered a new venue because the magnetic transitions ($T_M$) occur in an easily accessible temperature range with a variation of $T_M/T_C$ ranging from 1.75 for Dy to 0.14 for Tm. Further, high-quality single crystals have become available. Among the magnetic members, HoNi$_2$B$_2$C is particularly interesting because its complex magnetic phases are observed to coexist with superconductivity. Neutron scattering revealed three types of magnetic order. A commensurate antiferromagnetic structure is formed with $q = c^*$, i.e., $(0 0 1)$ below 6 K in which the spins point ferromagnetically within the $a - b$ plane and antiferromagnetically along the c-axis. Two incommensurate structures coexist over a finite temperature range, i.e., 5 K ≤ $T$ ≤ 6 K: the $c^*$ structure with $q = (0 0 0.915)$ where the ferromagnetically aligned spins are rotated by 165° and the $a^*$ structure with $q = (0.585 0 0)$ for which the detailed structure is still unknown.

HoNi$_2$B$_2$C exhibits a near-reentrant superconductivity, i.e., reentrant resistive behavior in a small magnetic field, due to competition between superconductivity and exchange-coupled antiferromagnetic order. In a theoretical analysis of the interplay, the onset of the c-axis incommensurate state has been shown to suppress superconductivity, leading to the near-reentrant behavior. Kreyssig et al., however, found that in Y-doped quaternary compound, Ho$_{1-x}$Y$_x$Ni$_2$B$_2$C, only the incommensurate a-axis feature remains in the same temperature range as the near-reentrant behavior while the c-axis spiral structure exists over a much wider temperature range, indicating that it is the a'-structure connected with Fermi surface nesting that enhances the pair-breaking effect. For a further understanding of the interplay, it is necessary to study the nature of the magnetic transitions and to establish the $B - T$ phase diagram of HoNi$_2$B$_2$C.

The magnetic phase diagram of HoNi$_2$B$_2$C has been studied extensively in the context of the interplay between magnetism and superconductivity, mostly below $B = 1$ T. Zero-field specific heat measurements and other surface sensitive measurements indicate that there are three magnetic transitions ($T_N$, $T^*$, $T_M$), while other specific heat data show only two transitions ($T_N$, $T_M$), where $T_N = 5.2$ K is the Neel temperature, $T_M = 6$ K is the onset of a long-range magnetic ordering, and $T^* = 5.5$ K is ascribed to some change of the oscillatory magnetic state. It has been suggested that the low temperature physical properties of polycrystalline HoNi$_2$B$_2$C depend on thermal treatment as well as chemical composition while the properties of single crystals are relatively less affected. Even though majority of measurements indicate the presence of $T^*$ at 5.5 K, thermodynamic measurements on single crystals that show the magnetic transition to be a truly bulk property are rare.

In this paper, we report specific heat measurements of single crystal HoNi$_2$B$_2$C as a function of temperature and magnetic field to provide an extended (up to 6 T), comprehensive magnetic phase diagram for $B \parallel \{100\}$ and $B \parallel \{110\}$. Three magnetic transitions are clearly re-
transition at 6 K is marked as $T_M$, the second at 5.5 K, as $T^*$, and the third at 5.2 K as $T_N$. There is also a specific heat discontinuity around 8 K related to superconducting transition but it is too small to be seen on the same scale.

The bottom panel shows the specific heat discontinuity related to the superconducting transition in the vicinity of $T_c$. The circles describe the specific heat difference, $\Delta C = C(B,T) - C(0,T)$, in the vicinity of $T_c$ for $B = 0, 50, 100$ mT solved, confirming that $T^*$ is a bulk magnetic transition. The 5.2 K transition ($T_N$) shows hysteresis with temperature, indicating a first order antiferromagnetic transition. The 6 K transition ($T_M$), namely the onset of long-range order, displays a dramatic in-plane anisotropy: $T_M$ increases with increasing magnetic field for $B \parallel [100]$ while it decreases with increasing field for $B \parallel [110]$. The anomalous anisotropy in $T_M$ indicates that the transition is related to the $a$-axis spiral structure. The 5.5 K transition ($T^*$) shows a small in-plane anisotropy and was ascribed to the transition from $\alpha$-dominant phase to $c^*$-dominant phase. The specific heat jump related to the superconducting transition was observed at 8 K.

II. SUPERCONDUCTIVITY

We have used two single crystals of HoNi$_2$B$_2$C, labeled sample A and sample B for the heat capacity measurements. Both samples were from a same batch. A high temperature flux method was used to grow the sample using Ni$_2$B as a solvent. The details are described elsewhere.

The heat capacity measured by ac calorimetry was converted to absolute values using literature data. The upper panel of Fig. 1 shows the specific heat of HoNi$_2$B$_2$C vs. temperature at zero field. The three sharp peaks mark the magnetic transitions noted above. The first

FIG. 1: Top panel: the specific heat of sample A in zero field. Bottom panel: the specific heat discontinuity, $\Delta C(B,T) = C(B,T) - C(0.2T,T)$, in the vicinity of $T_c$ for $B = 0, 50, 100$ mT
as that of HoNi$_2$B$_2$C (= 0.76).

III. MAGNETIC PHASE TRANSITIONS

A. Magnetic transition at 5.2 K ($T_N$)

The top panel of Fig. 2 shows a semi-log plot of the specific heat of HoNi$_2$B$_2$C (sample A) as a function of temperature at several magnetic fields $B \parallel [100]$. The bottom panel of Fig. 2 describes the specific heat of sample B for $B \parallel [110]$. The critical temperatures $T_N$ from C vs. $T$ in constant fields (squares) and the critical fields $B_N$ from the isothermal $C$ vs. $B$ at constant temperatures (circles; see Figs. 4 and 5) were plotted for both field directions. There is negligible in-plane $B_N$ anisotropy between [100] and [110] directions. The dashed line is the least-squares fit of $B_N = A(1 - T/T_N)^{0.5}$ in the vicinity of $T_N$, showing Ising-like behavior. Experimental data were explained well with $A = 1.41 \pm 0.01$ T.

The bottom panel of Fig. 3 shows the hysteresis of the specific heat of sample B as a function of temperature. The circles describe the data with increasing temperature and the crosses, with decreasing temperature. Inset: the temperature offset of the sample as a function of temperature.
one when it goes through a phase transition with latent heat.  

B. The 5.5 K magnetic transition ($T^*$)

The 5.5 K magnetic transition ($T^*$) is as sharp as the AF transition, for sample A at least, indicating it could also be a first order phase transition (see Fig. 2). The magnetic field dependence of the specific heat is also similar to the $T_N$ counterpart, i.e. the critical temperature decreases with increasing magnetic field. Initially, the peak intensity at $T^*$ becomes stronger with increasing field while that of the AF transition decreases monotonically, transferring some of its entropy to the $T^*$ transition. Fig. 6 shows the $B^* - T$ phase diagram both for $B \parallel [100]$ (open) and $B \parallel [110]$ (solid), where $B^*$ is the critical field corresponding to the $T^*$ transition. The in-plane $B^*$ anisotropy between [100] and [110] directions is small, similar to the $B_N$ transition. The $B^*$ temperature dependence near $T^*$ was explored in terms of the antiferromagnetic theory by Fisher as was done in the $T_N$ transition. The suppression of the critical temperature was explained reasonably well by $B^* = A(1 - T/T^*)^{0.5}$ with $A = 1.39 \pm 0.01$ T (dashed line), a value similar to that of the $T_N$ analysis.

What could be responsible for the 5.5 K transition? Neutron scattering showed that all three magnetic structures, i.e. $c$-axis commensurate AF magnetic structure, $c$-axis AF helical structure, and $a$-axis incommensurate structure, coexist between 5 K and 6 K. In the previous section, we argued that the 5.2 K transition is due to the magnetic transition to the $c$-axis commensurate AF structure ($T_N$) and is a first order phase transition. All the similarities between the 5.5 K transition and the 5.2 K transition point toward associating $T^*$ with the $c$-axis incommensurate AF structure. Then, the next question is “Is it a first order phase transition?” The specific heat at zero field did not show any noticeable hysteresis with temperature at $T^*$ (see Fig. 3). In the dc temperature offset (see the inset of Fig. 3), however, there may be a small hysteresis at $T^*$, but the feature is well within the scattered data. Coexistence of the phases also suggests that it is 1st order. At this point, it is not clear if the 5.5 K magnetic transition is a first order phase transition or not.
The 6 K magnetic transition ($T_M$)

The 6 K magnetic transition ($T_M$) is known to be the onset of a long-range magnetic order. The precise nature of the $T_M$ transition is not well characterized, although recent work argues that it represents the onset of the $a^*$ modulated phase. In this section, we present the specific heat as a function of temperature both for $B \parallel [100]$ and $B \parallel [110]$ directions and confirm that the 6 K transition is due to the onset of the $a$-axis incommensurate magnetic structure.

Fig. 2 shows a dramatic difference in the critical temperature $T_M$ with magnetic field directions. For $B \parallel [100]$, the critical temperature scarcely changes with increasing magnetic field below 0.5 T in agreement with Detlefs et al. In higher fields, it decreases rapidly as is expected for an antiferromagnetic transition and the transition shape becomes broadened, making it hard to interpret. For $B \parallel [100]$, the critical temperature increases with increasing magnetic field while keeping its transition shape. The anomalous in-plane anisotropy of $T_M$ strongly suggests that the origin of this transition is very different from the other two magnetic transitions ($T_N$, $T^*$) and is, therefore, due to the onset of the $a$-axis incommensurate magnetic structure. Detlefs et al. recently hypothesized that the $a^*$ magnetic structure is related to the Fermi surface (FS) nesting along [100] direction. Since a distortion along [110] direction is more disruptive to the nesting feature than a distortion along [100] direction, it results in the absence of the $a^*$ phase for HoNi$_2$B$_2$C and DyNi$_2$B$_2$C at low temperature and low magnetic field where local magnetic moments are aligned along [110] directions. If the $a^*$ phase is the ground state for 5.5 K $\leq T \leq$ 6.0 K in HoNi$_2$B$_2$C, a magnetic field applied along [110] will disrupt the magnetic phase while the field along [100] will not. The anomalous magnetic field dependence of specific heat is consistent with this scenario. The enhancement of the $a^*$ phase for $B \parallel [100]$, however, is beyond this explanation.

The top panel of Fig. 7 shows the specific heat data around $T_M$ in several constant magnetic fields from 0 T to 1.5 T for $B \parallel [100]$. For clarity, selective data are shown in the bottom panel, where $x$-axis is the reduced temperature $1 - T/T_M$ and $y$-axis is the normalized specific heat $C/C_{T_M}$ for $C_{T_M}$ is the specific heat at $T_M$. For clarity, selective data were shown.

FIG. 7: The top panel blows up the specific heat of sample A vs. temperature near $T_M$ in several magnetic fields when $B \parallel [100]$: from left to right, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.5 T. The bottom panel shows a scaling behavior of $T^*$ transition where $x$-axis is the reduced temperature $1 - T/T_M$ and $y$-axis is the normalized specific heat $C/C_{T_M}$ for $C_{T_M}$ is the specific heat at $T_M$. For clarity, selective data were shown.
of temperature range, i.e. $3 \times 10^{-3} < \tau < 3 \times 10^{-1}$. When temperature is close enough to $T_M (= 5.98)$ K, however, the specific heat data can be explained better with the logarithmic function than the power-law dependence. The logarithmic singularity may be interpreted as a manifestation of two-dimensional spin structure in HoNi$_2$B$_2$C where the Ho$^{3+}$ local moment is confined to the Ho-C basal plane for temperatures below 100 K.\textsuperscript{26} We note that the value of the coefficient of the logarithmic term, $C = 3$ J/mol·K, is of the order of magnitude found in the exact theory of 2D Ising antiferromagnets.\textsuperscript{30} The lower temperature side of $T_M$ is severely contaminated by the adjacent magnetic transition $T^*$, rendering analysis difficult.

D. Magnetic phase diagram

It has long been known that there is an extreme magnetic anisotropy associated with the crystalline electric field (CEF) splitting of the Hund’s rule ground state for the magnetic members of the borocarbide family RNi$_2$B$_2$C (R=Er, Tb, Ho, Dy).\textsuperscript{22} The R$^{3+}$ local moment is confined to the R-C basal plane for temperatures below roughly 100 K, i.e. temperatures well above the magnetic ordering temperatures. In addition, a strong in-plane anisotropy has been observed, leading to the local moments essentially being confined to either the [100] (R = Er, Tb) or [110] (R = Ho, Dy).\textsuperscript{22} Therefore, it became necessary to study physical properties as a function of magnetic field direction.

Fig. 9 summarizes the magnetic phase diagram of HoNi$_2$B$_2$C both for $B$ || [110] (upper panel) and for $B$ || [100] (lower panel) directions based on specific heat as a function of temperature and magnetic field. The 5.2 K transition, $T_N$, is due to the commensurate c–axis AF structure and found to be a first order phase transition. The critical field $B_N$ (triangles) shows little in-plane anisotropy. The 5.5 K transition, $T^*$, is due to the c-axis oscillatory magnetic structure. The in-plane anisotropy of $B_M$ (circles) is small and the nature of the transition is still controversial. The 6 K transition, $T_M$, is ascribed to the onset of the a-axis oscillatory magnetic structure. The in-plane anisotropy of $B_M$ (squares) is very surprising: the critical temperature decreases with increasing magnetic field for $B$ || [110] while it increases, for $B$ || [100]. The critical temperature $T_M$ initially increases with magnetic field at a rate of 0.48 K/T, then it starts to bend around at about 1.5 T and decreases (see the inset of the bottom panel of Fig. 9). The decrease of the critical temperature is expected for antiferromagnet.\textsuperscript{23,24} The increase, however, is difficult to understand.

The magnetic phase diagram built from specific heat data is consistent with the viewpoint of three distinct magnetic transitions between $5K \leq T \leq 6K$ at zero field.\textsuperscript{21} At low fields, the critical temperature $T_M$ scarcely changes with magnetic field for $B$ || [110] in
agreement with Du Mar et al.\textsuperscript{2} while the increase of $T_M$ for $B \parallel [100]$ is more evident, probably due to high-quality single crystal used in this study. In the high-field regime, our data show a dramatic difference in $T_M$ between the two field directions, i.e., $B \parallel [100]$ and $B \parallel [110]$ as is evident in Fig. 9 while previous studies drew a conclusion of similar behavior between the two directions. For $B \parallel [110]$, Ref 9 shows only two transitions below 5 K, i.e., $T_N$ and $T^∗$. In contrast, our data clearly show three transitions at the same temperature range. A close examination reveals that the $T^∗$ marked transition below 5 K in Ref 9 corresponds to $T_M$ in our phase diagram, indicating that the real $T^*$ transition is missing in that phase diagram.

There has been a speculation of a fourth magnetic phase based on resistance measurements where there occurs a slope change at 3.8 K for $B \parallel [100]$.\textsuperscript{13} In our bulk measurement, the additional feature was not observed, suggesting that the 3.8 K feature is an extrinsic property sensitive to the surface state. Instead, we found an additional phase line for $B \parallel [110]$, most readily seen as a shoulder in the 4.8 K and 5.0 K data in Fig. 5. It separates the $B_M$ and the $B^*$ lines and is merged to $B_M$ at 4 K (crosses in the upper panel of Fig. 9). The origin of this feature has yet to be elucidated.

**IV. MAGNETIC FIELD-ANGLE HEAT CAPACITY**

In non-magnetic borocarbides, the low temperature heat capacity directly measures the electronic density of states.\textsuperscript{22} In magnetic systems, however, the magnetic specific heat dominates. For example, the electronic and lattice components of the specific heat account for less than 5% of the total below 8 K in HoNi$_2$B$_2$C. Consequently, in HoNi$_2$B$_2$C, the field-angle dependent heat capacity used to investigate the superconducting gap nature in the non-magnetic systems, here mainly explores the magnetic structure. In this section, we present the in-plane field-angle heat capacity of the Ho-based borocarbide and discuss the data based on the phase diagram built in the previous sections.

Fig. 10 shows the low-field heat capacity vs field angle measured with respect to the a-axis of HoNi$_2$B$_2$C at several different temperatures. The periodicity of the peaks is 90 degrees at all temperatures, indicating that the peak in the specific heat is simply due to the in-plane anisotropy between [100] and [110] directions. Canfield et al. observed magnetization modulation as a function of magnetic field angle at 2 K.\textsuperscript{16} The oscillation feature was then interpreted in terms of metamagnetic states, i.e., ($\uparrow\downarrow$) for $B \leq B_{c1}$, ($\uparrow\uparrow\downarrow$) for $B_{c1} \leq B \leq B_{c2}$, ($\uparrow\uparrow\uparrow$) for $B_{c2} \leq B \leq B_{c3}$ and ($\uparrow\uparrow\uparrow\uparrow$) for $B \geq B_{c3}$. Here the arrow $\uparrow$ is a moment along the [110] axis, $\downarrow$ is a moment along the [110], and $\rightarrow$ is a moment along the [110] axis. In our magnetic phase diagram (Fig. 9), $B_{c1}$ corresponds to $B_N$, $B_{c2}$ to $B^*$, and $B_{c3}$ to $B_M$ respectively.

At 2.5 and 6.1 K, the low-field heat capacity has maxima for the field along $<100>$ directions. The area under the field-angle heat capacity is proportional to magnetic entropy change, indicating that there is more magnetic disorder for the field along $<100>$ than along $<110>$. The field-angle entropy modulation can be explained by the fact that the net moment of Ho$^{3+}$ ions is directed along $<110>$ directions in the commensurate antiferromagnetic phase.\textsuperscript{35} At 4.8, 5.2, and 5.6 K where helical magnetic phases appear, the peak positions shifted by 45° to $<110>$ directions, indicating that the magnetic moments are preferably aligned along $<100>$ directions. The peak intensities are also asymmetric with field angle, which may be related to the oscillatory magnetic structures observed\textsuperscript{13} in this temperature range.

Fig. 11 shows the field-angle heat capacity at 6.1 K in 0.5, 1, 2, and 4 T. According to the magnetic phase diagram of HoNi$_2$B$_2$C (see Fig. 9), the heat capacity should exhibit monotonic 4-fold angular oscillation at high fields because there is no transition other than the $T_M$ transition nearby this temperature. The peaks along [100] at 0.5 T, however, were split into two peaks at 1 T and the minima along [110] were totally flattened out. At 2 T, the distance between the two split peaks becomes narrower and the flat minima become broad maxima. In 4 T, the split peaks merge and show a delta function like peak and the broad maxima at [110] in 2 T returns to a broad minima as was in 0.5 T. Two satellite peaks appear

![Fig. 10: Heat capacity of sample B as a function of field angle at several temperatures. The magnetic field is rotated within the basal plane and is measured with respect to a-axis. The detailed condition is marked in each responsible panel.](image-url)
FIG. 11: Heat capacity of sample B as a function of field angle at 6.1 K in 0.5, 1, 2, 4 T. The circles describe the data with rotating magnetic field clockwise and the crosses, with rotating field counter clockwise.

at ±12° of [100] peaks.

The seemingly anomalous peak splittings along [100] may be explained as following. In 1 T, $T_M$ moves from 3.7 K for $B \parallel [110]$ to 6.5 K for $B \parallel [100]$ or $T_M = 3.7 + 0.031\alpha$ where $\alpha$ is the in-plane field angle measured against the a-axis. To move the $T_M$ through 6.1 K means a field angle of 0.4 K/0.031 K-deg$^{-1} \approx 12.5°$, which is consistent with the peak positions at $10\pm1.5°$. At 2 T, the $T_M$ moves from 0 K for $B \parallel [110]$ to 6.57 K for $B \parallel [100]$, predicting 6.3° of peak splitting. Experimentally, the observed splitting was 6 ± 1.5°. The larger the field, the faster the peak has to move in angle. Even though the anomalous peak splitting could be accounted for in terms of a simple linear relation between $T_M$ and field angle $\alpha$, the interchange between minima and maxima at [110] and the satellite peaks in 4 T have yet to be understood, suggesting that the magnetic phase in high fields is not as simple as was originally envisaged.

In Fig. 11, the circles describe the data when the in-plane magnetic field was rotated clockwise and the crosses, when the field was rotated counterclockwise. In this measurement, the angle spacing between two data points is 3°. At 0.5 T, the two data sets do not show hysteresis in phase, but show hysteresis in amplitude with field direction. Above 1 T, there is a systematic shift in phase by $\sim 3°$ with field direction, indicating that the related transition may be a first order magnetic transition. The experimental error bar in determining the field angle is ±1°, which is less than that of the angle shift in peak positions. The hysteresis with field-angle direction may be another manifestation of the first order nature of the metamagnetic transition from (↑↑→) to (↑↑↑) phases, which were observed in neutron-diffraction study by Campbell et al.\(^\text{12}\).

V. SUMMARY

We have studied the magnetic superconductor HoNi$_2$B$_2$C via specific heat as a function of temperature, magnetic field, and magnetic field angle. The small value of the specific heat discontinuity at $T_c$ indicates that Ho$^{3+}$ ions act as pair breakers as was predicted in Abrikosov-Gor’kov theory. The temperature hysteresis of the zero-field specific heat at $T_N$ provides direct evidence that the magnetic AF transition is of first order, accompanying a change from tetragonal to orthorhombic structure. The similarity of $T^\ast$ transition to the $T_N$ transition indicates that it is due to the $c^\ast$ spiral structure. The anomalous in-plane anisotropy of $T_M$ transition was explained in terms of the 0.6 $a^\ast$ Fermi surface nesting feature.

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