Magnon Specific Heat of Single Crystal Borocarbides $R\text{Ni}_2\text{B}_2\text{C}$ ($R=\text{Tm, Er, Ho, Dy, Tb, Gd}$).

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Abstract
Zero-field specific heat of the single crystals $\text{RNi}_2\text{B}_2\text{C}$ ($R=\text{Er, Ho, Dy, Tb, Gd}$) was measured within the temperature range $0.1 \text{ K} < T < 25 \text{ K}$. Linearized spin wave analysis was successfully applied to account for and to rationalize the thermal evolution of the low-temperature magnetic specific heats of all the studied compounds (as well as the one reported for TmNi$_2$B$_2$C) in terms of only two parameters, namely an energy gap $\Delta$ and a characteristic temperature $\theta$. The evolution of $\theta$ and $\Delta$ across the studied compounds correlates very well with the known magnetic properties. $\theta$, as a measure of the effective RKKY exchange couplings, scales reasonably well with the de Gennes factor. $\Delta$, on the other hand, reflects predominately the anisotropic properties: $\sim 2 \text{ K}$ for GdNi$_2$B$_2$C, $\sim 6 \text{ K}$ for ErNi$_2$B$_2$C, $\sim 7 \text{ K}$ for TbNi$_2$B$_2$C, and $\sim 8 \text{ K}$ for each of HoNi$_2$B$_2$C and DyNi$_2$B$_2$C. The equality in $\Delta$ of HoNi$_2$B$_2$C and DyNi$_2$B$_2$C, coupled with the similarity in their magnetic configurations, indicates that a variation of $\theta$ in the solid solution Ho$_{2-y}$Dy$_y$Ni$_2$B$_2$C ($x < 0.8$ and $T_c < T_N$) would not lead to any softening of $\Delta$. This supports the hypothesis of Cho et al (PRL 77, 163 (1996)) concerning the influence of the collective magnetic excitations on the superconducting state. This work underlines the importance of spin-wave excitations for a valid description of low-temperature thermodynamics of borocarbides.

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I. INTRODUCTION

Wide varieties of magnetic ground structures are manifested in $\text{RNi}_2\text{B}_2\text{C}$ series (see e.g. Refs. and Tab.I): members with $R=\text{Ho, Dy, Nd, Pr}$ adopt commensurate antiferromagnetic (AF) arrangements while those with $R=\text{Tm, Er, Tb, Gd}$ assume squared-up spin-density wave (SDW) states. These ground structures are stabilized by a fine balance of exchange, crystalline electric field (CEF), and magnetoelastic forces. Under magnetic field or temperature variation, most of these structures undergo a cascade of phase transformations, yielding rich varieties of field-temperature ($H-T$) phase diagrams (see e.g. Refs.). Interestingly, most of the zero-field magnetic states of $R=\text{Tm, Er, Ho, Dy}$ coexist with superconductivity, presenting model compounds wherein the interplay between superconductivity and magnetism can be investigated. Such investigations revealed that the superconductivity, though much influenced by, has a very weak influence on the prevailing magnetic order: the energy gain due to the onset of magnetic order dominates by two order of magnitudes over that due to the onset of superconductivity.

The zero-field part of the $H-T$ phase diagram of HoNi$_2$B$_2$C is particularly interesting: superconductivity sets-in at $T_c \simeq 8 \text{ K}$. Just below this point, an incommensurate spiral state, $\vec{k}_c \simeq 0.92 \hat{a}$, develops (see e.g. Refs.). Furthermore, at $\sim 6.3 \text{ K}$, an additional modulated state with $\vec{k}_\phi \simeq 0.55 \hat{a}$ emerges and around 5 K a deep minimum in $H_c2$ develops. At $T_N=5 \text{ K}$, an orthorhombic lattice distortion sets-in and, concomitantly, both the spiral and the a-axis modulated states are replaced by a commensurate AF structure which coexists with superconductivity down to the lowest measurable temperatures. Generalized susceptibility calculation related these modulated state to maxima in the exchange coupling transform, $J(k)$.

Remarkably, the magnetic ground structures of the heavy and magnetic $\text{RNi}_2\text{B}_2\text{C}$ compounds (see Tab.I) are particularly simple: in spite of the manifestation of an orthorhombic distortion and a liquid-helium-temperature magnetic modulation, their ground structures are either an equal-amplitude, AF-type squared-up state (as in $R=\text{Tm, Er, Tb, Gd}$) or an equal-amplitude, collinear, commensurate AF state (as in $R=\text{Ho, Dy}$). Then, it is of interest to investigate whether the low-temperature thermodynamics of these $\text{RNi}_2\text{B}_2\text{C}$ can be described in terms of small-amplitude spin-wave excitations and, in addition, to elucidate the character and dimensionality of these excitations. These magnetic excitations can be probed by various techniques, among which is the magnetic specific heat. We carried out extensive zero-field specific heat measurements on five single crystals $\text{RNi}_2\text{B}_2\text{C}$ ($R=\text{Er, Ho, Dy, Tb, Gd}$) covering at least the range $0.1 \text{ K} < T < T_N$. These
specific heats, together with that of TmNi$_2$B$_2$C (Ref.12), reveal a diversified and wide varieties of thermal evolutions. None the less, based on a phenomenological model, all of the specific heat curves can be systematized in terms of only two parameters, namely an effective exchange coupling and a magnetic anisotropy interaction.

The format of this paper is as follows: In Sec.II, we derive an approximate, but of wide applicability, expression for the magnon specific heat. Experimental techniques and procedures are described in Sec.III. Results and their analysis are described in Sec.IV and discussed in Sec.V.

II. APPROXIMATE EXPRESSIONS FOR MAGNON SPECIFIC HEAT OF RNi$_2$B$_2$C.

The magnetic structures of RNi$_2$B$_2$C (R=Tm-Gd) can be visualized as magnetic layers that are stacked along the c-axis. The most dominant interactions are the RKKY and anisotropic couplings. The former can be approximated by effective isotropic couplings while the latter (a combination of dominant CEF and weaker dipolar and anisotropic exchange forces) by an easy-axis anisotropy field $\vec{H}_a$. This $\vec{H}_a$ representation is convenient for spin-wave calculation and is valid for the low-temperature orthorhombic-distorted equal-amplitude collinear AF or squared-up phases of borocarbides (see Sec.V). Considering the above mentioned magnetic arrangement and the crystal symmetry, the magnetic couplings can be conveniently divided into two classes: $J^{A}_{ij}$ that couples moments $i$ and $j$ within the same layer (denoted as A or B) and $J^{AB}_{ij}$ that couples moments from different layers. Then at zero external field, the following Hamiltonian is expected to capture most of their low-temperature properties:

$$\mathcal{H} = - \sum_{<ij>,L \in A,B} J^{L}_{ij} \vec{S}_i^L \cdot \vec{S}_j^L + \sum_{<ij \in A,B} J^{AB}_{ij} \vec{S}_i^A \cdot \vec{S}_j^B - g\mu_B \vec{H}_a \cdot \sum_{j \in A} \vec{S}_j^A + g\mu_B \vec{H}_a \cdot \sum_{j \in B} \vec{S}_j^B \tag{1}$$

The first and second sum represent, respectively, interactions within the same layer and among different layers. The last two terms represent the anisotropic interactions. By standard spin-wave analysis, we obtained the following dispersion relation:

$$\hbar \omega_k = \sqrt{[SJ_\perp(0) - SJ_\perp(k) + SJ_\parallel(0) + g\mu_B H_a]^2 - [SJ_\parallel(k)]^2} \tag{2}$$

where $J(k) = \sum J_{ij} \exp(ik(r_i - r_j))$. $J_\parallel$ ($J_\perp$) represents the Fourier-transform parallel (perpendicular) to the c-axis. The energy gap, $\Delta = \hbar \omega_{k=0}$, is:

$$\Delta = \sqrt{(g\mu_B H_a)^2 + 2J_\parallel(0)g\mu_B SH_a} \tag{3}$$

which defines an AF resonance frequency similar to the uniform mode of ordinary AFs.\textsuperscript{$13,14,15,16$} Evidently, (i) $\Delta$ is zero whenever there is no anisotropy and (ii) $\Delta$ does not depend on the type nor the strength of the intralayer coupling.

For evaluating the magnon specific heat (or other thermodynamical quantities), an explicit expression of $J(k)$ is required. In the absence of such an expression and for low-temperature range, it is a common practice to assume a long-wave limit. Here, we restricted the expansion of $J(k)$ to the nearest neighbors only, leading to:

$$\hbar \omega_k \approx \sqrt{\Delta^2 + C_x k_x^2 + C_y k_y^2 + C_z k_z^2} \tag{4}$$

where $C_x$ ($\approx C_y$ for weak orthorhombic distortion) and $C_z$ are functions of the exchange couplings and geometrical factors ($a$ and $c$ are unit-cell parameters):

$$C_x = 16(J_\perp + J_\parallel)S^2 a^2 + 2J_\perp S(g\mu_B H_a) a^2 \tag{5}$$

$$C_z = 16 J_\parallel^2 S^2 c^2$$
Then, the zero-field magnon specific heat is (rewritten so as to conform with the notation of Ref. 15):

\[
C_M(T) = 3 \frac{3}{4} R (\Delta/\theta)^3 \left( \Delta/2T \pi^2 \right) \sum_{m=1}^{\infty} [\text{BesselK}(2, m\Delta/T) + \text{BesselK}(4, m\Delta/T)] 
\] (6)

where \(\text{BesselK}\) represents the modified Bessel function and

\[
\theta = z |J_{\text{eff}}| S = 3^{\frac{3}{4}} 2(C_x C_y C_z)^{\frac{1}{2}} / (a^2 c) \] (7)

is a characteristic temperature, based on which \(|J_{\text{eff}}|\) can be defined as being an effective exchange interaction that couples the magnetic moment to its \(z\) nearest neighbors. Notice that both Eq.6 and Eq.7 account as well for the weakly orthorhombic-distorted state.

For \(T < \Delta\), Eq.6 reduces to the exponential form:

\[
C_M(T) \approx 2 \frac{3}{4} R \Delta^2 \frac{\pi^2}{T^2} \exp(-\Delta/T) \] (8)

while for isotropic compounds or \(T \gg \Delta\), it reduces to the high-temperature limit \(14,15\):

\[
C_M(T) \approx 3 \frac{3}{4} \pi^2 R (T/\theta)^3 \] (9)

in this case, Eq.7 highlight the useful definition of \(|J_{\text{eff}}|\).

A long-wave dispersion relation for an isotropic quasi-2d case can be derived from Eq.4 if we set \(C_z \ll C_x (|J|| \ll J_\perp)\) and \(H_a = 0\):

\[
\hbar \omega_k \approx 8J_{\parallel}S + J_{\perp}Sa^2(k_x^2 + k_y^2) \] (10)

Then, to lower order in \(8J_{\parallel}S/T\), one obtains \(C_M(T) = \pi RT/12SJ_\perp\) which reproduces the leading linear-in-\(T\) term in the expression reported by Movshovich et al. \(12\) who (starting from a quadratic dispersion relation and including correction for the 2d and the magnon-magnon interaction) obtained for the range \(|J|| \ll T < T_N\):

\[
C_M(T) = (\pi R/12)(T/SJ_\perp - 6J_{\parallel}/\pi^2J_\perp + 4J_{\parallel}/3\pi^2J_\perp T) \] (11)

It is worth remarking that Eqs.11-13 are of a more wide applicability than our above analysis might have suggested. Furthermore, depending on only \(\theta\) and \(\Delta\), a variety of expressions for the magnetic specific heat can be derived; each admitting different limits for its thermal evolution depending on the relations among \(T\) and \(\Delta\) (compare Eq.6 with the limit Eqs.8,9,11). Based on such a scheme, one is capable of rationalizing the vast varieties of the low-temperature thermal evolution of thermodynamical quantities (such as \(C_M(T)\)) encountered in these (and any series similar to) borocarbides. It is reminded that this analysis is not adequate for the description of the contribution of the modulated state since the involved ordered components do not have equal magnitudes. Finally, the above zero-field treatment is obviously not appropriate for the description of the field-induced metamagnetic phases.

III. EXPERIMENTAL

Single crystals of \(RNi_2B_2C\) (\(R=\text{Er-Gd}\)) were grown by floating zone method \(17\). Physical characterization were carried out utilizing a dc magnetometer [\(H < 50 \text{ kOe}, 2 \text{ K} < T < 20 \text{ K}\)], a mutual-induction ac-susceptometer [\(\nu = 250-500 \text{ Hz}, H_{pp} \approx 1 \text{ Oe}, 30 \text{ mK} < T < 20 \text{ K}\)], and a dc resistivity
set-up \([H<50 \text{ kOe}, 2 K < T < 20 K]\). Structural, magnetic and transport characterizations are in agreement with published results.

The temperature-dependent specific heat was measured on a semi-adiabatic calorimeter \([80 \text{ mK} < T < 25 K, \text{precision better than} 4\%]\). The total specific heat curves measured above 2 K are in agreement with the reported data.\[^8, 18, 19, 20\] However, we observed some discrepancy between the absolute values of \(C_M(T)\) of single crystal and polycrystalline samples: though both specific heats were found to be given by approximately the same functional form, the absolute values of the fit parameters (\(\theta\) and \(\Delta\)) differ by as much as 40%.

For each compound, the total specific heat, \(C_{\text{tot}}\), was analyzed as a sum of an electronic \(C_e\) (\(C_S\) when superconductivity is to be emphasized), a Debye \(C_D\) (\(= \beta T^3\)), a nuclear \(C_N\), and a magnetic contribution \(C_M\) from the only magnetically active \(R\)-sublattice. At temperatures of interest, \(C_e\) and \(C_D\) were estimated based on our specific heat characterization of single crystal \(\text{YNi}_2\text{B}_2\text{C}\) \((\gamma = 17.5 \text{ mJ/moleK}^2\) and \(\beta = 0.12 \text{ mJ/moleK}^3\) which had been synthesized by the very same procedures as the one used for the other single crystals. Thus for each compound, \(\gamma\) was taken to be the same value as that of \(\text{YNi}_2\text{B}_2\text{C}\) while \(\beta\) was evaluated by normalizing the \(\beta\) of \(\text{YNi}_2\text{B}_2\text{C}\) using the approximation \(\beta \propto m^2\)\(^5\) (see Tab.II).

Within the superconducting region, \(C_S\) was evaluated as \(3\gamma T^3/T_e^2\). At any rate, for all the studied compounds, \(C_M(T)\) is much larger than the sum of \(C_e\) and \(C_D\). Consequently, even if \(C_e\) and \(C_D\) are taken as the bare values of \(\text{YNi}_2\text{B}_2\text{C}\), \(C_M(T)\) would not be noticeably modified, ensuring that our conclusions would not be influenced.

\(C_N(T)\) of \(\text{RNi}_2\text{B}_2\text{C}\), when available, is of dominant importance only at very low temperatures and was evaluated by least-square fit using the appropriate hyperfine Hamiltonian; the obtained parameters (shown in Tab.II) compare favorably with those of the corresponding \(R\)-metal\[^{22}\] and \(\text{RCO}_2\text{B}_2\text{C}\) isomorphs\[^{23}\].

IV. RESULTS AND ANALYSIS

From the general feature of \(C_M(T)\) curves (Figs.1-3, 5-6), one distinguishes four temperature regions: (i) a paramagnetic region, \(T > T_N\), wherein \(C_M(T)\) is predominantly due to a change in the population of the CEF levels, (ii) a critical region, \(T \approx T_N\), wherein \(C_M(T)\) is related to the critical phenomena, (iii) an intermediate region wherein \(C_M(T)\) reflects the magnetic character of the spiral/modulated states, and (iv) the low-temperature AF/squared-up states, of prime interest to this work, wherein the measured \(C_M(T)\) is to be confronted with Eqs.\[^6, 11\] and therefrom \(\theta\) and \(\Delta\) are to be extracted.

Before we discuss the features of \(C_M(T)\) for each compound, a word of caution is in order: just as in the case of \(R\)-metals\[^{22}\], the propagation of errors due to successive subtraction of \(C_e(T)\), \(C_D(T)\) and \(C_N(T)\) would eventually influence the absolute value of \(C_M(T)\).

A. \(\text{GdNi}_2\text{B}_2\text{C}\)

Below \(T_N\), the zero-field magnetic structure\[^2\] is a transverse sine-modulated type with \(\vec{k}_a\) that changes from \(0.551^\ast a\) at \(T_N\) to \(0.550^\ast a\) at \(T_R\), where a spin reorientation process sets-in. Below \(T_R\), \(\vec{k}_a\) reverts course and increases monotonically till reaching \(0.553^\ast a\) at 3.5 K.

\(C_M(T)\) of \(\text{GdNi}_2\text{B}_2\text{C}\) (Fig.1) reveals the onset of the magnetic order at \(T_N = 19.5\) K and the spin reorientation process at \(T_R = 13.5\) K, in agreement with earlier studies.\[^{2, 15, 26, 27, 28}\] The thermal evolution of \(C_M(T)\) within the amplitude-modulated state is distinctly different from that within the equal-amplitude, low-temperature state (see below). As mentioned above, within the modulated
region the linearized spin-wave analysis is not applicable and one should resort to the findings of Schmitt and co-workers. In particular, these authors demonstrated that $C_M(T_N)$ of such a state suffers a strong reduction (almost 1/3) in comparison with the value (20.15 J/moleK) expected for an equal-amplitude AF state (our results are in excellent agreement with this statement).

![Graph](image-url)

FIG. 1: log-log plot of $C_{tot}(T)$ (triangles), $C_c(T) + C_D(T)$ (dotted), and $C_M(T)$ (circles) curves of single crystal GdNi$_2$B$_2$C. The continuous line represents Eq.6 with $\theta = 12.5\pm0.2$ K and $\Delta = 1.9\pm0.3$ K. For $\Delta < T < 4$ K, $C_M(T)$ follows $0.058T^3$ J/moleK (dashed line) which is the high-T limit of Eq.6 (see text).

On the other hand, Fig.1 shows that for temperatures below 3.5 K, $C_M(T)$ follows faithfully Eq.6 with $\theta = 12.5\pm0.2$ K and $\Delta = 1.9\pm0.3$ K. The numerical value of $\theta$ (for $\Delta$ see Sec.V) is physically acceptable as can be seen from the following arguments. First, the substitution of this $\theta$ into Eq.7 yields $J_{eff} = 0.58\pm0.2$ K, which is in close agreement with the value reported for HoNi$_2$B$_2$C (Ref.7, 30) and TmNi$_2$B$_2$C (Ref.12). Secondly, the substitution of $\theta$ into Eq.9 predicts correctly the high temperature limit, namely $C_M(T > \Delta) = 0.058T^3$ J/moleK (see Fig.1). Thirdly, the substitution of $\theta$ into the Molecular Field relation

$$T_N = \frac{1}{3}\theta(S + 1)$$

(12)
gives $T_N = 18.8\pm0.3$ K which is in reasonable agreement with the experimentally determined value of $T_N$.

B. TbNi$_2$B$_2$C

A longitudinal SDW, accompanied by an orthorhombic distortion, sets-in at $T_N \approx 8$ K. The magnitude of the modulation vector decreases from $0.551a$ near $T_N$ to $0.545a$ at 2.3 K. A weak ferromagnetic component develops below $T_{WF} \approx 8$ K and at lower temperature a squaring-up of the modulated state was observed.31
$C_M(T)$ of single crystal TbNi$_2$B$_2$C (Fig.2) shows the magnetic ordering at $T_N = 14.5$ K and the WF-associated anomaly that peaks around 5.5 K. These features are in agreement with those reported by Tommy et al.$^{20}$ Similar to the case of GdNi$_2$B$_2$C, no attempt was made to analyze $C_M(T)$ within the amplitude modulated state spanning the range $5$ K $< T < T_N$. Below 5 K, where the orthorhombic-distorted squared-up state is expected, $C_M(T)$ follows convincingly the prediction of Eq.6 with $\theta = 21.5 \pm 0.2$ K and $\Delta = 7.0 \pm 0.5$ K.

![FIG. 2: log-log plot of the $C_{tot}(T)$ (triangle), $C_e(T)+C_D(T)$ (dotted), $C_N(T)$ (dashed) and $C_M(T)$ (circle) of single crystal TbNi$_2$B$_2$C. The continuous line represents Eq.6 with $\Delta = 7.0 \pm 0.5$ K and $\theta = 21.5 \pm 0.2$ K.](image)

C. DyNi$_2$B$_2$C

This compound develops a commensurate AF structure below $T_N$ with moments arranged in an identical manner as that of HoNi$_2$B$_2$C.$^{1,33}$ Moreover, superconductivity coexists with this AF order below $T_c \simeq 6$ K. In contrast to other Ni-based AF superconducting borocarbides, DyNi$_2$B$_2$C presents the following distinct features: (i) no zero-field incommensurate or modulated state was reported.$^{1,33}$ However for $T < 2$ K, anomalously large hysteresis and pronounced reentrant effects were observed for the field range $1$ kOe $\leq H \leq 5.3$ kOe.$^{34}$ (ii) the superconductivity emerges within a well developed AF order ($T_c < T_N$) and that $T_c$ is extremely sensitive to nonmagnetic doping.$^{35}$

$C_{tot}(T)$ (see Fig.3) reveals the onset of the AF order at $T_N = 9.5 \pm 0.2$ K. Within the accuracy of our measurement, the superconducting jump at $T_c \simeq 6$ K is too small to be resolved. On carrying out the analysis of $C_{tot}$ into its components ($C_e$, $C_D$, $C_N$, and $C_M$), we observed an anomalous contribution peaking at 1.2 K and having features reminiscent of a Schottky-like contribution. Accordingly, it was approximated by the standard two-level relation:

$$C_{sch}(T) = R\left(\frac{\delta}{T}\right)^2 \exp\left(\frac{\delta}{T}\right) \left[1 + \exp\left(\frac{\delta}{T}\right)\right]^2$$

(13)
where $\delta$ is the energy separation. It was found out (see the inset of Fig.3) that $\delta = 2.9$ K and that only 0.062 molar fraction was involved. Moreover, the fit was satisfactory for the high temperature tail but not so good at the lower temperature part, suggesting that a multi-level Schottky contribution might be more appropriate. However, for the present discussion, the above two-level approximation is sufficient. It is highly possible that such a contribution is due to 6% defect/impurity which is beyond the limit of detection of our X-ray structural characterization. Coincidently, anomalous hystereses effects were observed in the magnetostriction curves that were measured within the same temperature range. At any case, for $T > \delta$, both $C_{sch}(T)$ and $C_N(T)$ are smaller than $C_M(T)$ (see inset of Fig.3). Nevertheless, we considered $C_M(T) = C_{tot}(T) - C_S(T) - C_D(T) - C_N(T) - C_{sch}(T)$ (see Fig.3).

The thermal evolution of $C_M(T)$ is shown in a log-log plot in Fig.3 and as ln($C_M(T)$) versus $1/T$ plot in Fig.4. In both figures, the comparison with Eq.6 (solid line) is also presented. Evidently over a wide range of temperatures, $C_M(T \leq T_c)$ follows excellently Eq.6 with $\theta = 19.3 \pm 0.2$ K and $\Delta = 8.3 \pm 0.3$ K.

![FIG. 3: A log-log plot of $C_{tot}(T)$ (triangle), $C_e(T) + C_D(T)$ (dotted), $C_N(T)$ (dashed), and $C_M(T)$ (circles) of single crystal DyNi$_2$B$_2$C. The solid line represents Eq.6 (see text). The inset shows the individual contribution of $C_{tot}(T)$ (symbol), $C_N(T)$ (dashed), the magnetic fit (thin solid line), $C_{sch}(T)$ (dash-dot, see Eq.13), and the thick solid line is the sum of all contribution.](image)

D. HoNi$_2$B$_2$C

$C_{tot}(T)$ of single crystal HoNi$_2$B$_2$C (shown in Fig.5) reveals a cascade of three transitions that are usually attributed to magnetic transformations. The signature of the onset of superconductivity is too weak to be observable in our present measurements. On the other hand, for $T < 5$ K, $C_M(T)$ follows the description of Eq.6 with $\theta = 9.7 \pm 0.2$ K and $\Delta = 8.3 \pm 0.3$ K.
FIG. 4: \( \ln(C_M(T)) \) versus \( 1/T \) curve of single-crystal DyNi\(_2\)B\(_2\)C. The data (circles) are compared to Eq. 6 (solid line) giving \( \Delta = 8.3 \pm 0.3 \) K and \( \theta = 19.3 \pm 0.2 \) K. The inset shows on a linear scale, \( C_M(T) \) (circle) together with the comparison to Eq. 6 (see text).

FIG. 5: A log-log plot of \( C(T) \) versus \( T \) of single crystal HoNi\(_2\)B\(_2\)C showing \( C_{tot}(T) \) (triangle), \( C_e(T) + C_D(T) \) (dotted), \( C_N(T) \) (dashed), and \( C_M(T) \) (circle) contributions. The magnetic contribution (circles) are compared to Eq. 6 (solid line) giving \( \Delta = 8.3 \pm 0.3 \) K and \( \theta = 9.7 \pm 0.2 \) K (see text).
Two intriguing features of the $H - T$ phase diagram of ErNi$_2$B$_2$C are $^{37,38,39}$: the onset of the incommensurate transversely polarized SDW state with $k_a = 0.553a^*$ at $T_N = 5.94$ K, and the onset of weak ferromagnetism (WF) at $T_{WF} = 2.2$ K. These two events (none is able to quench superconductivity, $T_c = 10.52$ ± 0.05 K) are well evident in $C_M(T)$ of Fig.6. $T_{WF}$, in particular, is evident as a change of slope that separates two distinct thermal evolutions $^{40}$ $C_M(T)$ within the amplitude-modulated state $T_{WF} < T < T_N$ and that within the squared-up state at $T < T_{WF}$. In the latter region, $C_M(T < T_{WF})$ is well described by Eq.6 with $\theta = 7.4\pm 0.2$ K and $\Delta = 5.4\pm 0.3$ K.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{A log-log plot of $C_{tot}(T)$ (triangle), $C_e(T) + C_D(T)$ (dotted), $C_N(T)$ (dashed), and $C_M(T)$ (circle) of single crystal ErNi$_2$B$_2$C. The solid line is a comparison to Eq.6 with $\theta = 7.4\pm 0.2$ K and $\Delta = 5.4\pm 0.3$ K (see text).}
\end{figure}

\section{F. TmNi$_2$B$_2$C}

Superconductivity sets-in at 11 K and, below $T_N = 1.52 \pm 0.05$ K, coexists with a transversely polarized SDW state wherein the spins are pointing along the c-axis and the modulation vector is $(0.093,0.093,0)^{12}$ At lower temperatures, the incommensurate SDW state squares up $^{11}$ The magnetic specific heat of single crystal TmNi$_2$B$_2$C was measured by Movshivich et al $^{12}$ and was shown to follow Eq.11 with $J_\perp \approx 0.8$ K, $J_\parallel \approx 0.2$ K.

\section{V. DISCUSSION AND CONCLUSIONS}

On a linearized spin-wave (non interacting magnon gas) approach, one is always limited to the low-temperature region of the ordered state which in the case of the borocarbides amounts to being restricted to below the liquid Helium temperatures. Within that temperature range, the
model describes very successfully the magnetic contribution of the studied compounds indicating that \( C_M \) can be safely associated with the gapped collective excitations that propagate within the orthorhombic-distorted 3d AF (-type) structure.

The successful applicability of the model to the commensurate collinear AF ground structures of \( R = \text{Ho, Dy} \) is understandable. To justify its applicability to the cases of \( R = \text{Tm, Er, Tb, Gd} \), it is sufficient to show that their ground structures are well squared-up and that all the moments do have equal amplitudes and orient (or bunch) along a specific direction. The state of ErNi\(_2\)B\(_2\)C below \( T_{WF} \) and that of GdNi\(_2\)B\(_2\)C below 3.5 K provide the best illustrations of the fulfillment of this requirement. The colinear, equal-amplitude, and squared-up character of the state of ErNi\(_2\)B\(_2\)C below \( T_{WF} \) was elegantly revealed in the neutron diffraction studies of Choi et al\(^{37}\) and Kwano-Furukawa et al\(^{38}\). It is remarked that the presence of weak ferromagnetism (reflected as kink that separate oppositely oriented domains) would hardly modify this picture since the excitation energy of the kink is much higher than that of the magnon. Let us now discuss the character of the state of GdNi\(_2\)B\(_2\)C below 3.5 K. A recent magnetoelastic study\(^{18}\) on single crystal GdNi\(_2\)B\(_2\)C demonstrated the presence of substantial magnetoelastic and anisotropic exchange interactions, in particular below \( T < T_R \). The magnitude of the \( c^\| \) strain mode (which leads to symmetry lowering from four-fold tetragonal to two-fold orthorhombic) is very large and increases with decreasing temperature leading progressively to an orthorhombic-distorted magnetic state wherein the moments, due to entropy arguments, approach equal amplitudes. Based on the suggestion of Detlefs et al\(^{22}\), the magnetic structure below \( T_R \) is either a transverse modulated state with moment orientation away from the b-axis in the bc-plane or a modified spiral-like structure which, due to the low symmetry (2mm) of the [100] direction, is likely to suffer fanning or bunching (becoming more stronger as the temperature is decreased). Tomala et al\(^{26}\), on exploring these two structural possibilities, argued that the \(^{155}\)Gd Mossbauer spectra at 4.2K \( < T < T_R \) were better fitted with a bunched spiral-like state. In this Mossbauer analysis, the c-component was found to be almost equal (\( \approx 75\% \) at 4.2 K) to the b-component, a result that does not reflect the 2mm-symmetry of the [100] direction. Considering these and the above observations together with entropy arguments, one concludes that the low-temperature structure is either a squared-up, equal-amplitude, and collinear state or - as supported by Mossbauer analysis- a strongly bunched, squared-up, and equal-amplitude state. As far as the magnon specific heat is concerned, the interactions in both structures can be represented by the Hamiltonian of Eq.1 (allowing for a unimportant difference in the direction of the axis of quantization).

Figs.1-6 and Tab.I demonstrated convincingly that, based on only \( \Delta \) and \( \theta \), the diverse functional form of the measured \( C_M(T) \) can be systematized: when both \( \Delta \) and \( \theta \) are large, \( C_M(T < \Delta) \) reflects a magnon contribution from an anisotropic magnetic structure as in \( R = \text{Er, Ho, Dy, Tb} \). When \( \Delta \) is relatively small but \( \theta \) is large, \( C_M(T > \Delta) \) reflects a magnetic contribution from a quasi-isotropic magnetic structure as in GdNi\(_2\)B\(_2\)C. For a weak \( \Delta \) and \( J^{\|} \), \( C_M(T) \) reflects a contribution from a quasi-2d structure as in TmNi\(_2\)B\(_2\)C.

The evolution of \( \Delta \) and \( \theta \) across the studied compounds is reasonable. \( \theta \), on the one hand, reflects predominately the evolution of the de Gennes factor (see Eqs.5,7 and Tab.I) as can be appreciated on observing that \( \theta \) scales very well with the de Gennes factors for \( R = \text{Tm, Er, Ho, Dy, Tb} \). That the experimentally determined \( \theta \) of GdNi\(_2\)B\(_2\)C is a factor of three lower than the one expected from de Gennes scaling may be attributed to the additional dependence of \( \theta \) on \( H_a \) which for GdNi\(_2\)B\(_2\)C is the lowest.

\( \Delta \), on the other hand, reflects the combined influence of the anisotropic forces and interlayer coupling. This is expressed by Eq.3 which for the case of, say, ErNi\(_2\)B\(_2\)C (considering \( H_A \approx 15 \text{ kOe} \) and \( |J^{\|}| \approx 0.1 \text{ K} \) gives a value of 4 K which is close to the experimental value. The observation that \( \Delta \) is nonzero for each of the studied compounds is in agreement with the reported anisotropic features of the magnetic and transport properties.\(^{15}\) The strong anisotropy of each of \( R = \text{Er, Ho, Dy, Tb} \),...
Ho, Dy, Tb is in accord with what is expected from their CEF properties. In contrast, the weak anisotropy observed in GdNi$_2$B$_2$C is most probably due to a combination of anisotropic exchange, dipolar, and two-ion magnetoelastic couplings.

It is interesting to discuss one particular aspect of the interaction between magnons and superconductivity in, say, $R=$ Ho ($T_N < T_c$) and Dy ($T_c < T_N$): the magnon-mediated pairbreaking effect that manifests itself in the thermal evolution of $H_{c2}(T < T_N, T_c)$ [Ref.42]. Noteworthy, the thermal evolution of $H_{c2}(T < T_N, T_c)$ of both HoNi$_2$B$_2$C and DyNi$_2$B$_2$C [Ref.9] are very similar which, considering the above-mentioned similarity in their magnetic properties, suggests that the involved pairbreaking effects (in particular the magnon-mediated one) are similar. This, in turn, suggests that the magnon characteristic (say low-energy magnon spectra) in both compounds must be similar. This is indeed the case: the analysis of Sec.IV.C-D showed that the energy cost for magnon excitation in both compounds is practically equal ($\Delta \approx 8$ K see Tab.I). Therefore doping of Ho into DyNi$_2$B$_2$C (up to 80% but still $T_c < T_N$) would not lead to a softening of $\Delta$. Then, for this concentration range, there should be no variation in $H_{c2}$ and $T_c$ even though the deGennes factor does vary (remember that within the antiferromagnetic state, the Abrikosov-Gorkov pair-breaking mechanism is not valid). This provides an additional experimental confirmation of the hypothesis of Cho et al [35] that the magnon spectrum of $(\text{Dy}_{1-x}\text{Ho}_x)\text{Ni}_2\text{B}_2\text{C}$ is hardly modified for $x < 0.8$. In contrast, for $(\text{Dy}_{1-x}\text{Ho}_x)\text{Ni}_2\text{B}_2\text{C}$ ($x > 0.8$), the onset of superconductivity occurs within the paramagnetic state and consequently the Dy dopant depresses $T_c$ linearly as expected from the Abrikosov-Gorkov theory.

In summary, we were able to reveal the magnon specific heat contribution of the heavy members of the borocarbides and to identify the expressions that describe their thermal evolution. These expressions (given in terms of only two physically accepted parameters) were derived from the spinwave analysis of a simple Hamiltonian that consists of effective exchange couplings and anisotropic interactions. We investigated as well an influence of the magnons on the superconductivity of these AF superconductors. Improvements and extension of this analysis are underway. These include, on the experimental side, probing the magnon contribution in single crystals of $R$Ni$_2$B$_2$C by other (microscopic and macroscopic) techniques and, on the theoretical side, a better and more realistic approximation of $J(k)$, CEF effects, and magnetoelastic, and anisotropic exchange forces.

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A. Tables

TAB.I Some zero-field parameters of selected $RF_{2}B_{2}C$ compounds. Superconducting $T_c$, magnetic $T_N$, magnetic structure, propagation wave vector, and moment direction are taken from Ref.1. Squaring of the modulated SDW state is taken to occur at lower temperatures. The gap, $\Delta$, and characteristic temperature, $\theta$, were determined from the indicated equation and figure. $\theta_{exp}$ of TmNi$_2$B$_2$C is calculated by substituting into Eqs. 5, 7 the fit values ($J_\perp$=.8 K and $|J_\parallel|$=.2) given by Movshivich et al.12. $\theta_{deG}$ is the de Gennes scaling of $\theta$ taking that of $R$=Ho as a reference.

| $R$ | $\gamma$ | $C_s$ | $\theta$ | $\beta$ | $C_N$ | $\theta_{deG}$ | Eq. | Fig. | $mJ/moleK^{2}$ | J/moleK | (mJ/moleK$^{4}$) | $\alpha$ (K) | $P$ (K) |
|-----|----------|-------|-----------|--------|-------|-------------|------|-----|----------------|----------|----------------|-----------|--------|
| Gd  | 17.5     | 392(0.1935) | 0         | 0      | 1    | | |
| Tb  | 17.5     | 391(0.196) | 0.14(2)   | 0.02(1) | 2    | | |
| Dy  | 17.5     | 388 (0.200) | $-0.0396$ | $0.0554$ | 0.009 | 0.01 | 3    | |
| Ho  | 17.5     | 386 (0.203) | 0.29      | 0.009 | 5    | | |
| Er  | 17.5     | 384 (0.206) | 0.045     | $-0.0001$ | 6    | | |
| Tm  | $\sim$18 | $\sim$355($\sim$0.26) | 0.0202 | 0 | 4 in Ref.12 | | |