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Two-step magnetic ordering in as-cast and annealed DyNi₂B₂C samples

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Abstract. Polycrystalline DyNi₂B₂C samples, as-cast and annealed, were prepared and studied by x-ray diffraction (indicating the LuNi₂B₂C-type structure) and by magnetization. The magnetization study (in H = 20 Oe) of both samples indicates initial ordering at $T_N = 16.3(3)$ K, and a second transition, especially in the annealed one, at $T_o = 10.4(3)$ K. Transitions into superconducting states occur at $T_c \cong 6$ K in the annealed sample and at higher T_c in the as-cast sample. The two magnetic transitions agree with available neutron-diffraction data on annealed polycrystalline DyNi₂¹¹B₂C. A two-step magnetic ordering is proposed, involving initial ferromagnetic order in the basal planes, followed by a build-up of the correct antiferromagnetic (alternate) stacking of these planes throughout the samples. It is completed at the lower transition, T_o , only in the annealed sample, leading to its lower T_c .

1. Introduction

The LnNi₂B₂C compounds (Ln = a lanthanide) have been studied intensively in recent years due to their antiferromagnetic (AF) states and the coexistence of these magnetically ordered states with superconductivity for the heavier lanthanides Dy, Ho, Er and Tm [1–16]. A recent review paper by Gupta [1] describes main features of this family of quaternary compounds. These borocarbide compounds, as well as YNi₂B₂C, crystallize in the body-centred tetragonal (BCT) LuNi₂B₂C-type structure (space group *I*4/*mmm*) [3]. This structure is close to the BCT ThCr₂Si₂-type structure of most of the 1:2:2 compounds of the large AT₂X₂ family (A = Ln, U, Th; T = d transition element; X = Si, Ge), with carbon atoms entering the centres of the squares of A atoms in the basal planes, thereby modifying the ThCr₂Si₂-type structure into the LuNi₂B₂C-type structure. Both structures are characterized by a single free parameter, *z* (close to 3/8), determining the positions of the X (Si, Ge) and boron atoms, respectively.

Among the above $LuNi_2B_2C$ -type compounds those with Lu and Y are superconducting (SC) and are nonmagnetic, while those with Pr, Nd, Gd and Tb order with various AF structures but are normal conductors [1]. Out of the four compounds reported to have coexisting AF and SC states, $DyNi_2B_2C$ has been generally claimed [1, 4–13, 16] to be the only compound where

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the onset of an SC state occurs at T_c lower than the AF transition at T_N . This situation is discussed in detail in the recent review paper mentioned above [1].

As part of our magnetic studies of $(Pr_{1-x}Dy_x)Ni_2B_2C$ solid solutions at the National Research Institute for Metals (NRIM) [17], we have recently studied the magnetic properties of the end compounds, $PrNi_2B_2C$ and $DyNi_2B_2C$, and report here in detail our studies of the latter.

The lattice parameters, reported previously for $DyNi_2B_2C$ (in [3, 5, 6, 8, 9, 11, 16], respectively), fall in quite narrow ranges:

a[pm] = 354.2, 353.4, 353.2, 354, 353.2, 353.1, 353.42; c[pm] = 1050.1, 1048.4, 1048.8, 1051, 1048.2, 1048.6, 1048.78; $z_B = 0.3586$ (by x-rays [3], quite unexpected), 0.3584 (by neutrons [16]).

The paramagnetic parameters, reported previously for polycrystalline $DyNi_2B_2C$ [2, 6, 8, 11] or for a crushed $DyNi_2B_2C$ single crystal [5] are more spread out than the lattice parameters:

paramagnetic Curie temperature, θ [K] = +9.8, +1(2), -7, +8.1 ([2, 5, 6, 11], respectively); effective magnetic moment, μ_{eff} [μ_B] = 11.1, 9.85, 10.6, 10.63, 10.6 ([2, 5, 6, 8, 11], respectively).

The dc susceptibility, measured on single crystals [5, 10], is highly anisotropic, yielding paramagnetic values which depend on the direction of the applied magnetic field H with respect to the tetragonal axis c.

For a field *H* parallel to *c* $\theta = -82$ K, $\mu_{eff} = 10.4 \mu_B$ [5]; $\theta = -3$ K, $\mu_{eff} = 9.0 \mu_B$ [10]. For *H* perpendicular to *c* $\theta = +25$ K, $\mu_{eff} = 9.8 \mu_B$ [5]; $\theta = +16.4$ K, $\mu_{eff} = 12.38 \mu_B$ [10].

The agreement between the two sets of paramagnetic values is rather poor. The high effective magnetic moment (12.38 μ_B), obtained for the perpendicular case in the second study [10], is quite unreasonable, as it requires an unrealistic nickel effective magnetic moment of 4.5 μ_B .

Previous studies of the dc susceptibility in medium fields (MF, 100–1000 Oe) [2, 5, 7, 8], of the magnetization in low fields (LF, 2–3 Oe) [6], and of the ac susceptibility [4, 9, 11] conclude unanimously that $DyNi_2B_2C$ undergoes an AF transition around 10.5 K. It is a quite common among borocarbide researchers [1] to quote this temperature as the T_N of $DyNi_2B_2C$.

However, several of the polycrystalline DyNi₂B₂C samples studied in LF by magnetization [6] and ac susceptibility [4, 9, 11], prepared separately and even with systematic changes of some (Dy, C) of the constituents [4], show another magnetic transition at 16–16.5 K. Sample A of Lin *et al* [6], containing <5% Ni₂B impurity phase, seems (in their figure 2) to undergo an initial magnetic transition at 16 K, followed by a second transition at 10.5 K. Three of the samples prepared by Tomy *et al* [4] show (in their figure 2) initial magnetic transitions around 16.5 K: the carbon-excess sample DyNi₂B₂C_{1.1}, containing an impurity phase identified as DyB₂C₂, the dysprosium-deficient sample Dy_{0.09}Ni₂B₂C, with unstated purity, and the stoichiometric DyNi₂B₂C sample, showing no traces of impurities. Also sample A of Sanchez *et al* [9], that exhibits superconductivity below 6 K, undergoes (in the right inset of their figure 2) an initial magnetic transition at 16.5 K, followed by a second transition at 11 K. The two different samples of Hossain *et al* [11] show (in their figure 2) initial magnetic transitions at 16 K, clearly for sample II (that has no SC transition), being attributed by the authors to an impurity phase such as DyB₂C₂, and traces of such a transition for sample I (that has an SC transition).

The magnetization measurements on single crystals of DyNi₂B₂C (with $H \perp c$) could not detect any transition around 16 K because they were done at large temperature steps of 2 K around this temperature: by Tomy *et al* [7] (in their figure 2) at 13, 15, 17 and 19 K, in H = 100 Oe, and by Dervenagas *et al* [12] (in the inset of their figure 1) at 12, 14, 16, 18 and 20 K, in unreported magnetic field.

The weak magnetic transition around 16 K is not readily seen in the reported measurements of electrical resistivity [2, 4, 11] and specific heat [6, 7], but can be traced in the single-crystal electrical-resistivity measurements of Tomy *et al* [7] (in their figure 3).

The magnetic structure of DyNi₂¹¹B₂C has been determined by neutron-diffraction (ND) measurements (in zero applied field, ZAF) [12–16] to be an AF-I-related structure, in which the Dy ordered magnetic moments are aligned along the [110] direction within ferromagnetic basal planes, stacked antiferromagnetically (alternate stacking, + - + -). Of the two ND studies on as-grown single crystals of DyNi₂¹¹B₂C [12, 13], the former [12] has followed the temperature variation of the intensity only for the (001) magnetic reflection and reported that it disappears at 11 K. In the AF-I structure, where the magnetic moments are along the tetragonal axis and are perpendicular to the ferromagnetic basal plane, {001} reflections are absent. The observation of such reflections for the AF-I-related structure of DyNi₂B₂C indicates Dy ordered magnetic moments perpendicular to the tetragonal axis. This observation is supported by the ¹⁶¹Dy Mössbauer-effect (ME) study of an annealed DyNi₂B₂C sample by Sanchez *et al* [9], which suggests also Dy ordered magnetic moment $m = 9.8(1) \mu_B$, and a first-order magnetic transition around 11 K.

The neutron-diffraction measurements on an annealed polycrystalline $DyNi_2^{11}B_2C$ sample by Lynn *et al* [14–16] have shown that the Dy ordered moment at 1.7 K is $m = 8.47(9) \mu_B$, much lower than the ME value (with no magnetic moment on the Ni atoms). In measurements the intensity of the {001} reflection line and the Dy ordered magnetic moment do not vanish at the transition temperature, 10.18 K [14, 15] or 10.6 K [16]. Our present study of polycrystalline DyNi₂B₂C, as part of our crystallographic and magnetic studies of the (Pr_{1-x}Dy_x)Ni₂B₂C solid solutions [17], refers specifically to these neutron-diffraction observations.

2. Experimental details

A polycrystalline sample of DyNi₂B₂C, weighing 1.9 grams, was prepared by arc melting stoichiometric amounts of the constituents in an argon atmosphere. A titanium piece was used as a getter for the oxygen. The weight loss in the melting process was negligible. Part of the as-cast sample, weighing 0.76 grams, was then annealed in vacuum at 1323 K for 168 h. The preparation procedure was quite similar to the one used by Lynn *et al* [16] for preparing their annealed polycrystalline DyNi₂¹¹B₂C sample. Both our as-cast and annealed samples were characterized by x-ray diffraction at room temperature (RT = 295 K), using a Rigaku diffractometer. Mostly single-phase materials were formed, with low content of minority phases, comparable with that inferred from published x-ray diffractograms, in both as-cast and annealed samples. The RT diffractograms of these samples (figure 1) are almost identical.

Magnetization measurements, on a part of the as-cast sample weighing 159 mg, and on a part of the annealed sample weighing 149 mg, were made with a SQUID setup in the temperature range of 1.8–273 K. The measurements were made in magnetic fields of 20 Oe (low field, LF) and 1000 Oe (medium field, MF). The materials were first subjected to zero-field cooling (ZFC) down to 1.8 K and measured in LF up to 20 K. They were then subjected to field cooling (FC) in the LF down to 1.8 K and re-measured up to 273 K. For better determination of the paramagnetic values, the annealed sample underwent later ZFC down to 20 K and measurement in MF up to 273 K, yielding the dc-susceptibility data.

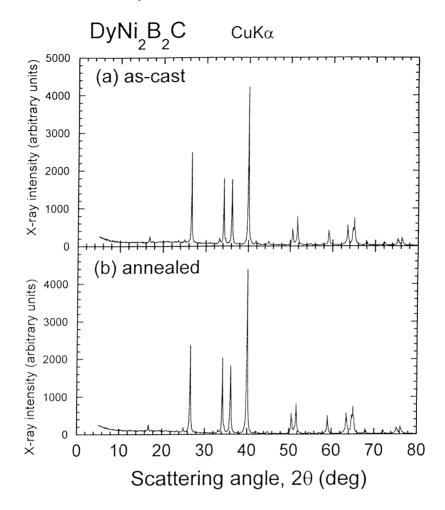


Figure 1. X-ray (Cu K α radiation, $\lambda = 154.178$ pm) diffractograms of polycrystalline samples of DyNi₂B₂C: (a) as cast, prepared by arc melting the constituents in an argon atmosphere, and (b) annealed, in vacuum at 1323 K for 168 h. The diffractograms of both samples are practically identical, with depression of small impurity lines upon annealing. Only {*hkl*} diffraction lines with h + k + l = even are observed, indicating space group *I4/mmm*.

3. Results

The x-ray diffraction measurements at RT confirm the formation of a single-phase $DyNi_2B_2C$ compound, which crystallizes in the body-centred tetragonal $LuNi_2B_2C$ -type structure (space group I4/mmm). The RT lattice parameters, determined from the Rietveld profile analysis of the recorded diffractograms (figure 1), are:

for the as-cast sample: $a = 353.53(2)$ pm,	c = 1049.27(7) pm;
for the annealed sample: $a = 353.61(2)$ pm,	c = 1049.13(8) pm;
$z_B = 0.369(16)$ and 0.371(24), respectively, determined with a rather low accuracy.	

The annealing process introduces minor changes in the lattice parameters, involving a small increase in a and a matching small decrease in c, with no change in the tetragonal-cell volume. The values to be quoted are no doubt those obtained for the annealed sample.

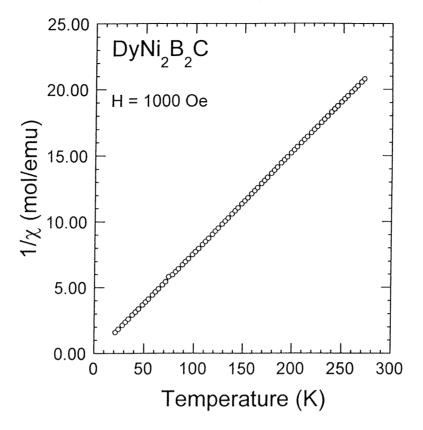


Figure 2. MF (1000 Oe) inverse dc susceptibility of the annealed sample of polycrystalline DyNi₂B₂C in the temperature range of 20–273 K. The straight line yields a paramagnetic Curie temperature $\theta = +0.6$ K and an effective magnetic moment $\mu_{eff} = 10.21 \ \mu_B$.

The MF inverse dc susceptibility of the annealed polycrystalline DyNi₂B₂C sample obeys a Curie–Weiss law yielding a straight line in the entire temperature range 20–273 K (figure 2). From this line we obtain the paramagnetic Curie temperature $\theta = +0.6(5)$ K, and deduce the effective magnetic moment $\mu_{eff} = 10.21(1) \mu_B$. The LF inverse dc susceptibility yields a similar value for θ but a reduced value of μ_{eff} , due to uncertainty in the value of the LF seen by the sample (known to be subjected to compensating fields in the SQUID).

The LF (20 Oe) magnetization measurements on the as-cast polycrystalline DyNi₂B₂C sample in the temperature range of 1.8–20 K are shown in figure 3(a). The ZFC and FC curves coincide above \cong 18 K, slightly above the initial ordering transition at $T_N = 16.3(3)$ K, seen as a peak in the ZFC curve and as a saddle in the FC curve. Both curves show traces of a second transition at \cong 10 K. A third transition on the ZFC curve is associated with the establishment of the negative part of this curve (figure 3(a)), due to the diamagnetic signal from the SC state, known to occur in DyNi₂B₂C. It seems to appear at T_c above the value encountered in annealed samples, \cong 6 K [4–11].

The LF (20 Oe) magnetization measurements on the annealed polycrystalline $DyNi_2B_2C$ sample in the temperature range of 1.8–20 K are shown in figure 3(b). The ZFC and FC curves coincide exactly above the initial ordering transition at $T_N = 16.3(3)$ K, seen as saddles on both curves. These curves indicate a second magnetic transition at $T_o = 10.4(3)$ K, seen as peaks on both curves. A third transition on the ZFC curve is associated with the establishment

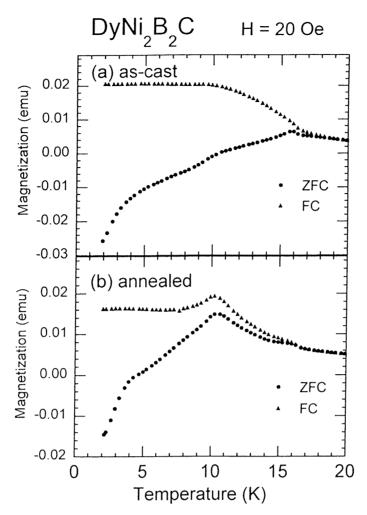


Figure 3. LF (20 Oe) dc magnetization of ZFC and FC polycrystalline DyNi₂B₂C samples, in the temperature range of 1.8–20 K: (a) as-cast sample (159 mg)—the ZFC and FC curves coincide above \cong 18 K, slightly above the initial magnetic-ordering temperature, $T_N = 16.3$ K, and both show traces of another magnetic transition at \cong 10 K; (b) annealed sample (149 mg)—the ZFC and FC curves coincide exactly above the initial magnetic-ordering temperature, $T_N = 16.3$ K, and both indicate a second magnetic transition at $T_o = 10.4$ K. The ZFC curves of both samples are negative at the lower temperatures due to diamagnetic signals from the SC states, occurring below third-transition temperatures T_c , with slightly higher value in the as-cast sample.

of the negative part of this curve (figure 3(b)), due to the diamagnetic signal from the SC state below $T_c \cong 6$ K, as exhibited by previously obtained ZFC curves for annealed DyNi₂B₂C samples [4–11].

4. Discussion

The lattice parameters obtained for our polycrystalline samples of $DyNi_2B_2C$ (the values for the annealed sample being: a = 353.61(2) pm, c = 1049.13(8) pm) agree with the published values [3, 5, 6, 8, 9, 11, 16], thereby suggesting also a similar quality of our samples.

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Our MF paramagnetic values ($\theta = +0.6(5)$ K, $\mu_{eff} = 10.21(1) \mu_B$) fall well in the middle of the published data for the paramagnetic Curie temperature θ , (-7)–(+9.8) K, and for the effective magnetic moments μ_{eff} , 9.85–11.1 μ_B [2, 5, 6, 8, 11]. The paramagnetic Curie temperature in our polycrystalline DyNi₂B₂C, +0.6 K, indicates weak magnetic exchange interactions at the rather low initial ordering temperature, $T_N = 16.3$ K. The effective magnetic moment in our polycrystalline DyNi₂B₂C, 10.21 μ_B , is slightly below the free ion value for Dy³⁺, 10.63 μ_B . It should be mentioned that the Dy ordered magnetic moment, 8.47(9) μ_B , obtained directly by ND on annealed polycrystalline DyNi₂¹¹B₂C [14–16], is well below the free-ion value for Dy³⁺, 10 μ_B , while the value, 9.8(1) μ_B , obtained indirectly by ME on annealed polycrystalline DyNi₂B₂C [9], is close to the free-ion value.

Our LF magnetization, measured on the as-cast sample of polycrystalline DyNi₂B₂C, indicates that initial magnetic ordering occurs at $T_N = 16.3(3)$ K, and is characterized by a peak in the ZFC curve and a saddle on the FC curve (figure 3(a)). The lower-temperature transition is not so developed in this stage of preparation of the present material. The as-cast sample exhibits, nevertheless, the expected SC state at T_c above the common value of $\cong 6$ K, as seen from the negative part of the ZFC curve (figure 3(a)).

The most striking feature of our LF magnetization, measured on the annealed sample of polycrystalline DyNi₂B₂C, is the three transitions observed. The initial magnetic ordering occurs at $T_N = 16.3(3)$ K, and is characterized by saddles on both LF magnetization curves, obtained following ZFC and FC (figure 3(b)). Another transition at a lower $T_o = 10.4(3)$ K is characterized by peaks in both LF magnetization curves, obtained following ZFC and FC (figure 3(b)). It appears like the usual magnetic-transition peaks that one encounters in three-dimensional magnetic ordering. The annealed sample exhibits the anticipated SC state at $T_c \approx 6$ K, as seen from the negative part of the ZFC curve (figure 3(b)).

As already mentioned above, other annealed polycrystalline samples of $DyNi_2B_2C$ seem also to undergo an initial magnetic transition around 16 K [4, 6, 9, 11]. Some of the people studying $LnNi_2B_2C$ systems, such as Hossain *et al* [11], tend to associate this transition with the complicated ferromagnetic-like ordering of an impurity phase DyB_2C_2 at 16.4 K, reported by Sakai *et al* [18] in their early study of the LnB_2C_2 compounds. There are, however, several arguments against association of the higher-temperature transition with this impurity phase:

- (a) The various preparations of $DyNi_2B_2C$ cannot have the same impurity phase (DyB_2C_2), or equivalent amounts of this phase. The stoichiometric sample of Tomy *et al* [4], showing no traces of impurities, still shows the existence of a magnetic transition around 16.5 K.
- (b) Our measurements on the (Pr_{1-x}Dy_x)Ni₂B₂C solid solutions [17] indicate systematically varying saddle-like magnetic transitions, at 16.3(3) K in DyNi₂B₂C, at 15.0(3) K in PrNi₂B₂C, and at 14.2(3) K in (Pr_{0.91}Dy_{0.09})Ni₂B₂C, as well as traces of such a transition at 13.5 K for (Pr_{0.75}Dy_{0.25})Ni₂B₂C. As PrB₂C₂ does not order magnetically down to 3 K [18], it does not cause the other transitions. We therefore believe that also DyB₂C₂ is not behind the transition in DyNi₂B₂C.
- (c) The transition at 16.3 K in DyNi₂B₂C is also consistent with the ZAF powder neutrondiffraction measurements of Lynn *et al* [14–16]. The persistence of the {001} magnetic reflection above 11 K cannot be traced to an impurity phase but to the major phase, since the impurity phase will give totally different magnetic reflections, with discontinuity of the magnetic reflections of the major phase.

In figure 4 we compare our LF magnetization versus temperature curves (figure 4(c)) of the annealed polycrystalline $DyNi_2B_2C$ sample with the temperature dependence of the ZAF neutron-diffraction intensity of the first magnetic reflection {001} (figure 4(a)) [14, 15] and of the deduced Dy ordered magnetic moment (figure 4(b)) [16]. It is rather clear that

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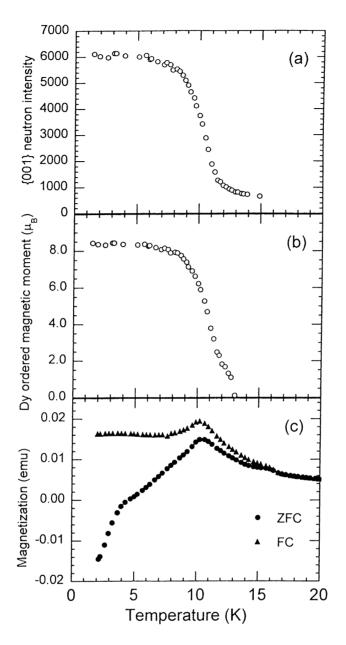


Figure 4. Comparison of (a) the temperature dependence of the ZAF neutron-diffraction intensity of the first magnetic reflection {001} [14, 15], and (b) the deduced Dy ordered magnetic moment [16], with (c) our LF magnetization versus temperature curves, for annealed polycrystalline samples of DyNi₂B₂C. Both neutron-diffraction-based curves clearly persist between the T_o and T_N transitions.

both ND-based curves persist between the T_o and T_N transitions, and that initial ordering of DyNi₂B₂C in the AF-I-related structure occurs at $T_N = 16.3$ K.

A somewhat similar situation, of two consecutive magnetic transitions associated with the same structure (AF-I, mentioned above), has been recently reported for several (A, A') Co_2Ge_2

solid solutions, crystallizing in ThCr₂Si₂-type structure [19–21]. In these solid solutions, namely the systems (U, Tb)Co₂Ge₂ [19], (U, Nd)Co₂Ge₂ [20] and (Nd, Tb)Co₂Ge₂ [21], the magnetic atoms A and A' are randomly distributed in the (A, A') site in the basal planes. The initial ordering, aligning all magnetic moments in the basal planes along the tetragonal axis, antiparallel for the (U, Tb) and (Nd, Tb) cases and parallel for the (U, Nd) case [19– 21], takes place at T_N , due to the strong in-plane ferromagnetic exchange in these materials, especially in the uranium systems. At the initial ordering the alternate sequence required by the AF-I structure is not yet established, due to the random character of the magnetic basal planes, and the weaker interactions (RKKY type) between such consecutive planes (separated by Co_2Ge_2 planes). As the temperature is lowered, the size and number of regions with the correct alternate stacking (fully ordered regions, yielding coherent neutron diffraction) rise, and entail gradual increase in the magnetic (AF-I) diffraction lines. At a lower temperature, T_{o} , the entire sample acquires the alternate structure, giving rise to the full AF-I ordering. We are therefore considering a two-step magnetic ordering. The initial step involves a two-dimensional ordering, due to in-plane ferromagnetic, mostly direct, exchange interactions, which sets in at T_N , accompanied by a build-up of the three-dimensional ordering. The second and final step involves a transition, at a lower T_{o} , to fully ordered (three-dimensional) magnetic (AF-I) state. On lowering the temperature between the two transitions, from T_N to T_o , the susceptibility continues to rise, as full ordering is not yet achieved. Simultaneously, the magnetic neutrondiffraction intensities increase, although at a slower rate with respect to the increase in the fully ordered state below T_{a} .

For the magnetic behaviour of annealed polycrystalline DyNi₂B₂C we propose a similar two-step magnetic ordering. The initial step involves a second-order transition at T_N , where the in-plane ferromagnetic ordering of the Dy moments starts, and it is considerably weakened due to the presence of the carbon atoms in these basal planes. The coupling between the distant consecutive planes is rather weak, being separated by Ni₂B₂ planes (or chains). On lowering the temperature the Dy ordered moments increase, as do the total ferromagnetic moments of the planes, as well as the inter-plane interactions (RKKY type), overcoming the large separation between consecutive planes. The number and size of regions with full AF-Irelated stacking increase, and at the lower transition temperature T_o they extend quite abruptly over the entire sample, having undergone a first-order transition [9, 14, 15]. This is the second and final step in forming the observed magnetic ordering at the lowest temperatures. On lowering the temperature of polycrystalline $DyNi_2B_2C$ from T_N to T_o , the magnetization still increases, due to the existence of non-ordered regions (figures 3(b), 4), and simultaneously the magnetic neutron-diffraction intensities increase (figure 4(a)), though at a rather slow rate with respect to the rate below T_{o} . The present model makes unnecessary the two-dimensional Ising model explanation proposed [14, 15] for the lower transition. The weak initial two-dimensional ferromagnetic ordering is reflected also in the rather small value of θ .

In the as-cast polycrystalline DyNi₂B₂C, the initial in-plane ferromagnetic ordering occurs naturally at the same ordering temperature of the annealed sample, namely at $T_N = 16.3$ K. At the temperature of the second transition the magnetization curves do not depict any peak (figure 3(a)), probably due to the incomplete three-dimensional ordering (i.e. the partial AF-I-related stacking) in this stage of preparation of the material. The higher SC transition temperature in this stage of preparation is also related to the incomplete magnetic order.

A similar magnetic behaviour, with an initial saddle-like transition, is observed also in other polycrystalline $LnNi_2B_2C$ -type materials, namely our $PrNi_2B_2C$ and $(Pr_{0.91}Dy_{0.09})Ni_2B_2C$ samples [17].

The absence of the two-step magnetic ordering in the neutron-diffraction measurements on an as-grown single crystal of $DyNi_2^{11}B_2C$ [12] cannot easily be explained. It is probably

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the annealing procedure, normally not applied to single crystals, that stabilizes the required crystallographic order involved in the initial magnetic ordering in the basal planes.

The appearance of an SC state already in the as-cast stage of $DyNi_2B_2C$ (figure 3(a)), where the absence of full magnetic ordering is related to incomplete crystallographic order in the basal planes, confirms earlier indications about the role of Ni_2B_2 chains (formed by the adjacent Ni and B planes) in the superconductivity [1, 9]. From figure 3 it seems that the SC transition in the as-cast sample occurs at a higher temperature with respect to the transition in the annealed sample, probably due to some depression of T_c by the complete AF-I-related ordering of the latter. Electrical-resistivity measurements are planned in order to confirm this preliminary observation, and to find the T_o/T_c relation for the as-cast DyNi₂B₂C sample.

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