

# Stability of the modulated states in $\text{ErNi}_2\text{B}_2\text{C}$ : A neutron diffraction study of chemically doped samples

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Neutron-diffraction measurements on  $\text{Er}_{0.8}\text{R}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  ( $R=\text{Tb}, \text{Lu}$ ) and  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  have been carried out to investigate the influence of chemical doping on the stability of the modulated states in  $\text{ErNi}_2\text{B}_2\text{C}$ . Within the magnetically ordered phase and down to 1.6 K (the lowest measured temperature), the powder diffractograms of Lu and Tb doped compounds are consistent with a transverse sine-modulated (fundamental) mode. A neutron diffraction study on a single-crystal  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  recorded at 1.8 K has confirmed the absence of higher order harmonics. In contrast, the low-temperature patterns of the Co doped compound reveal the presence of all the odd-order harmonics up to the fifth-order satellite, reminiscent of the structure of the undoped  $\text{ErNi}_2\text{B}_2\text{C}$  below the weak-ferromagnetic transition point  $T_{WF}=2.1$  K: a squared-up, noncompensated long-period antiferromagnetic state. The above-mentioned findings are consistent with the conclusions drawn from our previous specific-heat and magnetization measurements on the same compounds: on lowering the temperature further below  $T_N$ , both  $\text{ErNi}_2\text{B}_2\text{C}$  and Co doped compounds undergo a spontaneous transition at  $T_{WF}$ . In contrast, for the Lu and Tb doped compounds, no such second magnetic phase transition was observed down to 1 K; eventually the squaring-up of these structures, as required by entropy arguments, should occur but at temperatures lower than 1 K.

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## I. INTRODUCTION

$\text{ErNi}_2\text{B}_2\text{C}$  is a member of the borocarbide series that manifests very interesting superconducting and magnetic phase diagrams. It crystallizes in the tetragonal ( $I4/mmm$ )  $\text{ThCr}_2\text{B}_2$ -type structure.<sup>1-3</sup> At  $T_c \approx 11$  K, a BCS-type superconductivity sets in. Below  $T_N \approx 6$  K and with no quench of superconductivity, the Er moments order into a transverse spin-density wave (SDW)  $\vec{q} \approx 0.55\vec{a}^*$ , moment aligned along the  $b$  axis.<sup>1-5</sup> Entropy requirements dictate that at very low temperatures, such a sinusoidal state transforms either into an equal-amplitude state (with a different  $\vec{q}$ ) or else into a squared-up state if  $\vec{q}$  is not drastically altered. Indeed, based on single-crystal neutron-diffraction analysis,<sup>4,5</sup> it was inferred that a lock-in to a commensurate, squared-up state occurs at  $T_{WF} \approx 2.2$  K, concomitant with a surge of a weak ferromagnetic (WF) component that coexists with the superconducting state.<sup>6</sup>

Characteristic features are manifested at  $T_{WF}$  in neutron diffraction as well as in various other measurements such as magnetization,<sup>6</sup> specific heat,<sup>7-9</sup> critical current,<sup>10</sup> and high resolution x-ray magnetic scattering.<sup>11</sup> In particular, neutron diffraction studies conducted below  $T_{WF}$  reveal a set of additional even-order magnetic satellites. These satellites together with the weak ferromagnetism are considered to be a

by-product of a lock-in to a long-period noncompensated antiferromagnetic structure.<sup>4,5,12,13</sup> Earlier, Kawano-Furukawa *et al.*<sup>4</sup> and Choi *et al.*<sup>5</sup> proposed that the magnetic structure below  $T_{WF}$  is a 20-unit-cell periodic arrangement wherein, as suggested by the former authors, only the Er moments within the  $z=0$  plane contribute to the weak ferromagnetism. Jensen,<sup>12</sup> in a mean-field treatment reminiscent of the earlier work of Gignoux and Schmitt,<sup>14,15</sup> argued that on lowering the temperature, the magnetic structure undergoes at  $T_{WF}$  a first-order transition into a commensurate, 20-unit-cell period, uncompensated antiferromagnetic state. Alternatively Walker and Detlefs,<sup>13</sup> based on a Ginzburg-Landau theory, argued that at  $T_{WF}$  the system undergoes a second-order lock-in transition into a commensurate, uncompensated antiferromagnetic arrangement with a wave vector satisfying  $\vec{q} = (m/n)\vec{a}^*$  with  $m$  even and  $n$  odd.

The stability of any one of these modulated states (as well as the WF component) against physical and chemical perturbations is an interesting issue, which is expected to shed additional light on their nature as well as on the character of the events around  $T_{WF}$ . Field-dependent neutron diffraction on a single-crystal of  $\text{ErNi}_2\text{B}_2\text{C}$  (Refs. 16-18) showed that, for  $H < H_{sat}$  and  $H \parallel [0\ 1\ 0]$  or  $H \parallel [1\ 1\ 0]$ , the  $a$ -axis modulated, incommensurate states are maintained, though  $|\vec{q}|$  varies with  $H$ . Recently we have studied the effect of substitu-

tion on the magnetic and superconducting properties of  $\text{ErNi}_2\text{B}_2\text{C}$ :<sup>19</sup> a 3d substitution ( $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$ ), a nonmagnetic 4f substitution ( $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ ), and a magnetic 4f substitution ( $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ ). The Co substitution is expected to introduce variations in the Fermi-surface properties and, also, in the features of the localized 4f moment. On the other hand, a substitution by another isoelectronic heavy 4f ion would hardly influence the Fermi-surface properties but would certainly modify the effective de Gennes factor. The results show that 5% Co substitution degrades the superconductivity much stronger than any of the R substitution even though the magnetic features of  $\text{ErNi}_2\text{B}_2\text{C}$  and  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  are very much alike: in good agreement with the case of  $\text{HoNi}_2\text{B}_2\text{C}$  ( $T_N \sim 8$  K,  $T_c \sim 8$  K) wherein a 0.75% Co doping reduces  $T_c$  to below 3 K while the magnetic structure is maintained unchanged.<sup>20,21</sup> Furthermore, the low-field magnetization of  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  reveals at low temperatures a weak ferromagnetic component while no such component is evident in the magnetization of either the Tb or Lu doped compound. In addition, specific heat measurements performed on the same samples show that the Co doped compound manifests the two phase transitions (i.e., at  $T_N$  and  $T_{WF}$ ) just as the ones found for the pure compound while, in contrast, for Tb and Lu doped compounds—measured down to 1 K—only the transition at  $T_N$  is evident. To get further insight into their magnetic properties as well as to evaluate how these substitutions influence the stability of the modulated states of the parent compound, this work investigated the magnetic structures of these doped samples utilizing neutron-diffraction technique.

## II. EXPERIMENT

90% <sup>11</sup>B-enriched polycrystals of  $\text{ErNi}_2\text{B}_2\text{C}$ ,  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ ,  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ , and  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  were prepared by the conventional arc-melt method. These polycrystals were used in powder diffraction measurements and also as feeding rods during the floating-zone synthesis, a process that we used for single crystals growth.<sup>22</sup> The measurements reported in this work were performed on as-grown samples, the physical characterization<sup>19</sup> of which are in good accord with the reported data on  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ ,<sup>23–26</sup>  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ ,<sup>22</sup> and  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$ .<sup>27,28</sup> Specifically, their  $T_N$  and  $T_c$  are given in Table II (see later):  $T_N$  ( $T_c$ ) was taken as the maximum (onset) point in the temperature-dependent isofield magnetization and ac susceptibility curves. There is a slight difference between these  $T_N$  and those of the single crystals,<sup>19</sup> however, there is no extra broadening (as compared to the single crystal counterparts). On the other hand,  $T_c$  of these as-grown polycrystals is almost equal to those of the single crystal counterparts, but there is a considerable broadening in the superconducting transition region. Considering the agreements of our structural analysis on  $\text{ErNi}_2\text{B}_2\text{C}$  with the reported results (see below), we believe that our conclusions regarding the magnetic structures of the doped samples would not be influenced by the slight differences in the magnitude of their  $T_N$  nor the broadening in their  $T_c$ .

Neutron-diffraction measurements were carried out at the Institut Laue-Langevin in Grenoble, France.

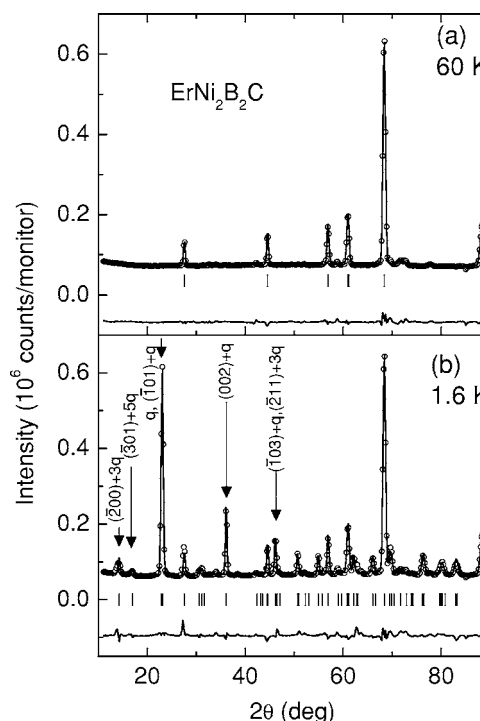


FIG. 1. Neutron powder diffractograms of  $\text{ErNi}_2\text{B}_2\text{C}$  at (a) 60 K and (b) 1.6 K. For each panel, circles represent the total measured diffraction intensity while the solid line gives the calculated pattern. The difference curve is shown at the bottom while the Bragg positions are indicated as short bars (the arrows indicate the low-angle positions of the first, third, and fifth harmonics). The refined parameters are given in Table I for the nuclear pattern while in Table II for the magnetic contribution.

Powder diffraction patterns of  $\text{ErNi}_2\text{B}_2\text{C}$ ,  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$ ,  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ , and  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  were collected using the D1B diffractometer with a selected incident wavelength of 2.52 Å. Measurements on  $\text{ErNi}_2\text{B}_2\text{C}$  were carried out so as to provide a comparison and a reference. For each compound, patterns were collected between 1.6 and 80 K. A single-crystal study on  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  was performed at 1.8 and 20 K on the D10 four-circle diffractometer with  $\lambda = 1.26$  Å.

For powder data analysis, Rietveld refinements of both crystallographic and magnetic structures were carried out using the FULLPROF package of Rodriguez-Carvajal (L. L. B.), based on the original code of Wiles *et al.*<sup>29</sup>

## III. RESULTS AND ANALYSIS

Nuclear diffraction patterns within the paramagnetic phase of  $\text{ErNi}_2\text{B}_2\text{C}$ ,  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$ ,  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ , and  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  are shown in Figs. 1(a), 2(a), 3(a), and 4(a), respectively. Refinements of the crystallographic (and also the magnetic) structure were performed assuming a Pseudo-Voigt peak profile and neglecting preferred orientations, asymmetry, absorption, and strain effects. For the three substitutions, the best refinements (see Table I) were found for the body-centered tetragonal ( $I4/mmm$ ) crystal structure, the same as that of  $\text{ErNi}_2\text{B}_2\text{C}$ ,<sup>1–5</sup> wherein the atomic posi-

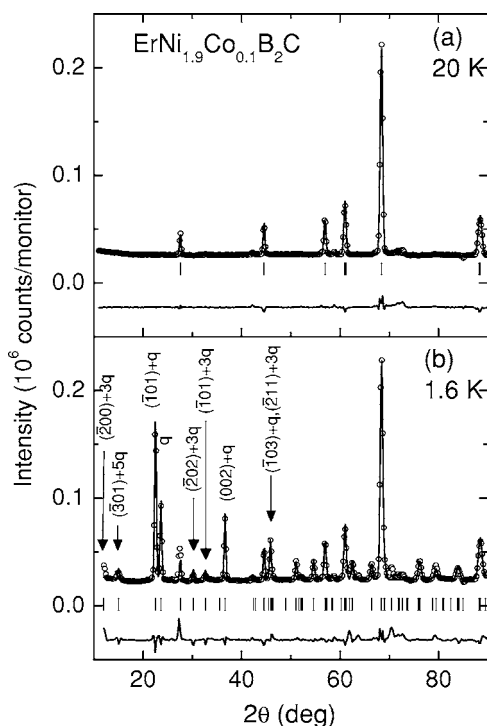


FIG. 2. Neutron powder diffractograms of  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  at (a) 20 and (b) 1.5 K. Circles represent the total measured diffraction pattern while the solid line represents the refinement. The difference curve is shown at the bottom while the Bragg positions are indicated as short bars (the arrows indicate the low-angle positions of the first, third, and fifth harmonics). The refined parameters are given in Table I for the nuclear pattern while in Table II for the magnetic contribution.

tions are taken as:  $4f$  ion  $(0,0,0)$ ,  $3d$  ion  $(1/2,0,1/4)$ , B  $(0,0,z)$ , and C  $(0,0,1/2)$ . The overall features of all the patterns are characteristic of a single-phase sample with, however, a small amount of impurities that are present at the same angles and with almost the same relative intensity.

Based on the reported orthorhombic distortion within the antiferromagnetic phase of  $\text{ErNi}_2\text{B}_2\text{C}$ ,<sup>30,31</sup> we assumed a body-centred-orthorhombic phase ( $Immm$ ) for the crystallographic structure below  $T_N$  and with atomic positions similar to the ones used in the analysis of the paramagnetic phase. We found out that though the calculated difference between the  $a$  and  $b$  parameters remains extremely small (see Table I), nevertheless this undertaking does improve the refinement.

Low-temperature magnetic diffractograms of the studied samples differ appreciably from each other: compared with those of  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  [Figs. 3(b)] and  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  [Figs. 4(b)], patterns of  $\text{ErNi}_2\text{B}_2\text{C}$  [Figs. 1(b)] and  $\text{ErNi}_{1.9}\text{Co}_{0.1}\text{B}_2\text{C}$  [Figs. 2(b)] show additional magnetic satellites, which based on similar analysis of the pure compound,<sup>1-5</sup> are ascribed to the third-order and fifth-order harmonics. A wide “bump” is evident at the bottom of the magnetic peak at  $2\theta \approx 23^\circ$  in the spectra of both  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  and  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ . This feature, attributed to magnetic disorder effects, suggests that the  $4f$  substitution creates relatively more disorder than the  $3d$  substitution.

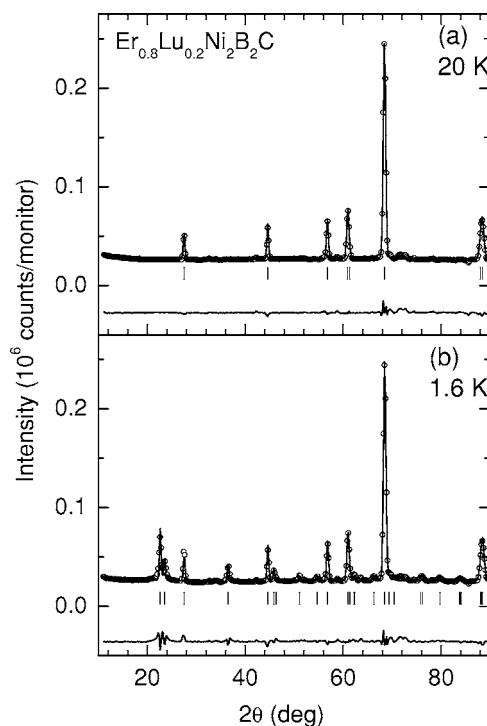


FIG. 3. Neutron powder diffractograms of  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  at (a) 20 and (b) 1.6 K. Circles represent the total measured diffraction pattern while the solid line represents the refinement. The difference curve is shown at the bottom while the Bragg positions are indicated as short bars. The refined parameters are given in Table I for the nuclear pattern while in Table II for the magnetic contribution.

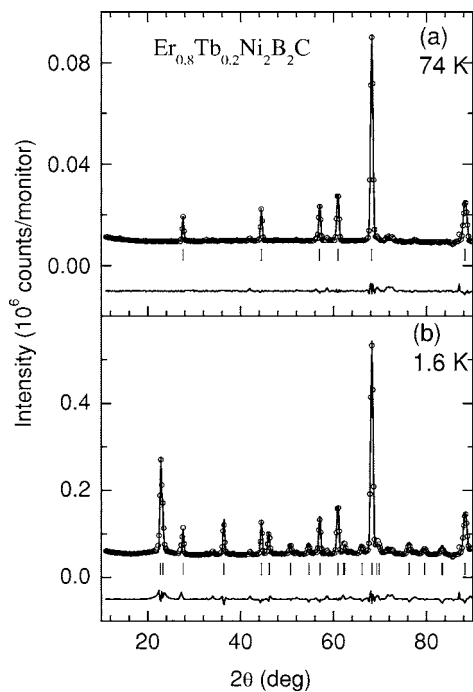


FIG. 4. Neutron powder diffractograms of  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  at (a) 74 and (b) 1.5 K. Circles represent the total measured diffraction pattern while the solid line represents the refinement. The difference curve is shown at the bottom while the Bragg positions are indicated as short bars. The refined parameters are given in Table I for the nuclear pattern while in Table II for the magnetic contribution.

TABLE I. The refined parameters and the reliability factors ( $R_B$  is the Bragg R-factor,  $R_F$  is the RF-factor) of the crystallographic structure within the paramagnetic and the ordered phases of the studied compounds (see text). The temperature of the measurement within the ordered phase is approximately 1.6 K. Note the extremely small change in the lattice parameters above and below the onset of the magnetic order.

Compound	Paramagnetic phase ( $I4/mmm$ )						Ordered phase ( $Immm$ )					
	$T$ (K)	$a$ (Å)	$c$ (Å)	$z$	$R_B$	$R_F$	$a$ (Å)	$b$ (Å)	$c$ (Å)	$R_B$	$R_F$	
ErNi <sub>2</sub> B <sub>2</sub> C	60	3.502(1)	10.581(2)	0.362(1)	5.8	3.3	3.506(1)	3.496(1)	10.577(2)	5.8	4.6	
ErNi <sub>1.9</sub> Co <sub>0.1</sub> B <sub>2</sub> C	20	3.501(1)	10.574(3)	0.361(1)	7.0	3.7	3.504(1)	3.497(1)	10.571(2)	8.1	5.3	
Er <sub>0.8</sub> Tb <sub>0.2</sub> Ni <sub>2</sub> B <sub>2</sub> C	74	3.511(1)	10.560(2)	0.361(1)	3.8	1.8	3.512(1)	3.508(1)	10.560(2)	3.9	2.46	
Er <sub>0.8</sub> Lu <sub>0.2</sub> Ni <sub>2</sub> B <sub>2</sub> C	20	3.497(1)	10.601(2)	0.361(1)	5.9	3.3	3.497(1)	3.493(1)	10.596(2)	4.9	3.2	

Magnetic refinements were undertaken assuming  $\vec{q}_i$  ( $i = 1, 3, 5, \dots$ ) to be along the  $a^*$  direction and the amplitude of the corresponding Fourier component along the  $b$  axis. For the analysis of the magnetic spectra of Er<sub>0.8</sub>Tb<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C, we assumed, in conformance with the so-called “uniform model,” only one effective magnetic moment at all rare-earth sites; in particular, the orientation of the Tb moment in Er<sub>0.8</sub>Tb<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C is taken to follow that of the majority Er moments. The best results of the refinements of the magnetic phases of ErNi<sub>2</sub>B<sub>2</sub>C and ErNi<sub>1.9</sub>Co<sub>0.1</sub>B<sub>2</sub>C are shown in Figs. 1(b) and 2(b), respectively. For this ErNi<sub>2</sub>B<sub>2</sub>C sample, specific heat studies showed  $T_{WF} = 2.1(1)$  K, which is in agreement with the published results. Then, based on the reported single-crystal neutron diffraction analysis,<sup>4,5</sup> the diffractograms collected at 1.6 K should reveal the odd-order as well as the even-order satellites. That the present powder diffractograms reveal up to fifth-order harmonic is an assuring result; higher odd-order or even-order satellites would have much lower intensities that would be within the statistic of our present measurements.

For the case of Tb and Lu doped compounds, the magnetic phase is well reproduced assuming only a simple sine-modulated structure [see Figs. 3(b) and 4(b)].

The fitting parameters of the low-temperature magnetic patterns of the studied samples are given in Table II. The amplitude of the first component  $M_1$  of ErNi<sub>2</sub>B<sub>2</sub>C (similarly that of ErNi<sub>1.9</sub>Co<sub>0.1</sub>B<sub>2</sub>C) is roughly 25% smaller than the corresponding  $M_1$  reported by Lynn *et al.*<sup>1</sup> However, both of these zero-field amplitudes are lower than  $9.9 \mu_B$ , the calculated  $M_1$  based on the field-induced saturated moment re-

ported by Canfield *et al.*<sup>6</sup> assuming that at such temperatures  $M_1 = 4\langle\mu\rangle/\pi$ . On the other hand, the amplitude of the sine wave in Er<sub>0.8</sub>Tb<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C (similarly Er<sub>0.8</sub>Lu<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C) is lower than the corresponding amplitude of ErNi<sub>2</sub>B<sub>2</sub>C; as such the variation of  $M_1$  within the studied compounds does not follow the de-Gennes scaling.

The inference that the higher order harmonics in the diffractograms of Er<sub>0.8</sub>Tb<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C are absent within the probed range of temperatures is consistent with the measurements of Hillier *et al.*<sup>25</sup> who reported that within the experimental resolution and down to 1.5 K, there are no third and higher order harmonics in the magnetic powder diffractograms of Er<sub>1-x</sub>Tb<sub>x</sub>Ni<sub>2</sub>B<sub>2</sub>C ( $x=0.075, 0.9$ ). For Er<sub>0.8</sub>Lu<sub>0.2</sub>Ni<sub>2</sub>B<sub>2</sub>C, this absence is consistent with the results of our neutron diffraction scans of a single crystal of the same compound. These scan were performed at 1.8 K for more than 630 nuclear peaks and 600 magnetic reflections along various directions of the reciprocal space. Two representative results are shown in Fig. 5. The lower panel shows the relative intensity of the (0.3,0,0) peak to be negligibly small. This result should be contrasted with that of the undoped ErNi<sub>2</sub>B<sub>2</sub>C compound wherein the intensity of the third-order harmonic was reported to be almost 20% of that of the first-order peak (see the inset of Fig. 5). Finally, the upper panel of Fig. 5 shows the diffraction events to be composed of only the nuclear ( $h+k+l=2n, k=-2$ ) and the magnetic ( $G \pm \vec{q}_1$ ) spots.

#### IV. DISCUSSION

It is evident that each of the chemical doping imparts a characteristic modification on the modulated states of

TABLE II. Refinement of the magnetic phases of the studied compounds.  $q_n$  and  $M_n$  are, respectively, the refined values of the wave vector (in units of  $a^*$ ) and the Fourier coefficient of the  $(2n+1)$ th harmonic mode.  $R_{mag}$  is the reliability factor for the magnetic refinement. The critical temperatures  $T_c$ ,  $T_N$ , and  $T_{WF}$  were determined from magnetization and ac susceptibility measurements (see Sec. II). For  $T_c$ , the parentheses indicate the width of the superconducting transition region taken as the temperature interval between the 10% and 90% of the diamagnetic signal.

Compound	$T_c$ (K)	$T_N$ (K)	$T_{WF}$ (K)	$ q_1 $ (Å <sup>-1</sup> )	$ q_3 $ (Å <sup>-1</sup> )	$ q_5 $ (Å <sup>-1</sup> )	$M_1$ ( $\mu_B$ )	$M_3$ ( $\mu_B$ )	$M_5$ ( $\mu_B$ )	$R_{mag}$
ErNi <sub>2</sub> B <sub>2</sub> C	11.0(5)	6.7(7)	2.2(1)	0.552(1)	0.345(1)	0.758(1)	7.0(1)	1.9(2)	1.0(2)	14.8
ErNi <sub>1.9</sub> Co <sub>0.1</sub> B <sub>2</sub> C	3.0(5)	6.2(2)	2.0(2)	0.570(1)	0.291(1)	0.848(1)	6.8(1)	1.7(2)	1.4(2)	12.7
Er <sub>0.8</sub> Tb <sub>0.2</sub> Ni <sub>2</sub> B <sub>2</sub> C	3.6(9)	6.9(1)		0.561(1)			5.3(1)			17.7
Er <sub>0.8</sub> Lu <sub>0.2</sub> Ni <sub>2</sub> B <sub>2</sub> C	10.6(9)	5.2(2)		0.566(1)			4.8(1)			32.7

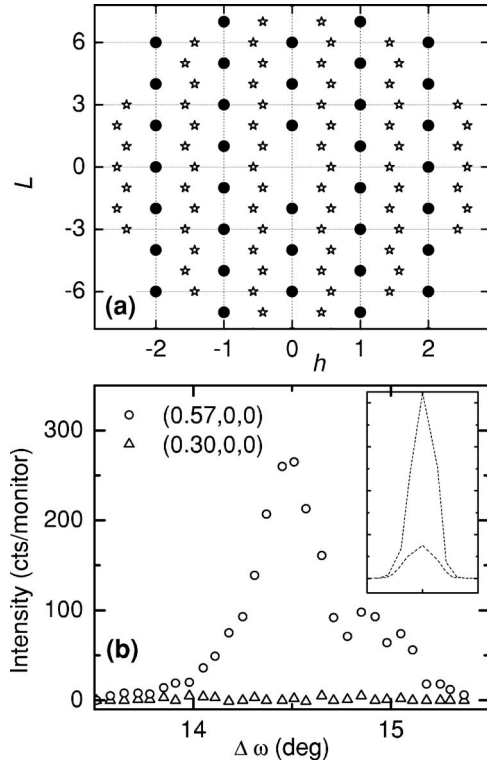


FIG. 5. (a) A representative cross section of the reciprocal space of  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  showing a  $q$  scan of the  $(h, -2, l)$  plane at 1.8 K. The only detected diffraction events are the nuclear (filled circles) peaks  $G \equiv (h, -2, l)$  [ $h+k+l=2n$ ] and the magnetic (stars) satellites ( $G \pm q_1$ ). (b) Neutron scattering profiles ( $\omega$  scan) on a single crystal of  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  at 1.8 K performed around the neighborhood of the positions expected for  $q_1=(0.57,0,0)$  and  $q_3=(0.30,0,0)$  magnetic satellites. The inset shows a similar  $\omega$  scan for  $\text{ErNi}_2\text{B}_2\text{C}$  at 1.3 K, demonstrating the relative intensity of the third-order satellite with respect to the first-order peak (taken from Ref. 5).

$\text{ErNi}_2\text{B}_2\text{C}$ . Below we discuss two of these modifications, namely the change in  $\vec{q}$  (accompanying each substitution) and the reduction in the intensity of the higher order harmonics (accompanying rare-earth substitutions).

Due to the nesting features in the electronic band structure of borocarbides, their wave-vector dependent susceptibility,  $\chi(q)$ , and the Fourier transform of the exchange interactions,  $J(\vec{q})$ , manifests a peak at  $\vec{q} \approx 0.6a^*$ .<sup>32</sup> thus the stabilization of the modulated  $4f$  arrangement with almost the same  $\vec{q}$ . This must also be the case for the alloys studied in this work: the survival of the modulated state for all doping (even for a 20% nonmagnetic substitution) is a testimony that the nesting features are not appreciably modified.

For all the studied substitutions, there is a small but discernible change in  $|\vec{q}|$  (see Table II); the variation being more sizable for the Co-doped sample. Further, none of the obtained  $\vec{q}$  is locked-in to  $(11/20, 0, 0)$ ; though  $\vec{q}$  of  $\text{ErNi}_2\text{B}_2\text{C}$  is very close. It is worth recalling that  $\vec{q}$  is governed by the energy balance between the crystalline electric field, magnetoelastic coupling, and the exchange interactions [through  $J(n\vec{q})$ , the high-order harmonics tend to square-up the structure]. Furthermore, a modification in  $|\vec{q}|$  could arise from a

variation in  $\xi$ , an opening of a superzone gap, or a reduction in the mean free path of the conduction electrons  $\ell$ . Specific to the latter, a variation in  $\ell$  would induce an exponential reduction in the amplitude of the oscillating RKKY function, usually expressed as  $J(r)\exp(-r/\ell)$ .<sup>33,34</sup> Most of these variations are evident in the elemental heavy rare earths and their alloys.<sup>34</sup> For the particular case of borocarbides,  $|\vec{q}|$  does vary with  $\xi$  (see, e.g., Refs. 1 and 11). In addition, as this work shows,  $|\vec{q}|$  varies as well with the chemical doping, which—in turn—influences both  $\xi$  and  $\ell$ . Obviously for the Co substitution, the above mentioned argument concerning the variation of  $\xi$  does not apply; rather the major influence is expected to arise from the induced alteration in the Fermi surface properties, which—as a consequence—would modify the position of the maximum of  $J(\vec{k})$ .

The quench of the intensity of third and higher odd-order harmonic in  $\text{Er}_{0.8}\text{R}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  is reminiscent of the case of  $\text{Er}_{1-x}\text{R}_x$  ( $R=\text{Tb}, \text{Dy}, \text{Y}$ ).<sup>34</sup> Such an effect is usually discussed within a molecular field model (see, e.g., Ref. 34) where  $J(\vec{k})$  is considered to be concentration independent and the crystal field interaction is approximated by a uniaxial anisotropic energy term  $B_2^0[3S_z^2 - S(S+1)]$ . Within this scenario, the decrease in the intensity of the third and higher odd-order satellites is attributed to a decrease in the effective  $|B_2^0|$ . For the  $\text{Er}_{1-x}\text{Tb}_x$  case, the decrease in  $|B_2^0|$  is due to an addition of Tb substituent, which—in contrast to Er—has a positive  $B_2^0$ . Though this argument is valid for temperatures close to  $T_N$ , we suggest that the disappearance of the high-order harmonics in  $\text{Er}_{0.8}\text{R}_{0.2}\text{Ni}_2\text{B}_2\text{C}$  within the range  $1.6 \text{ K} < T < T_N$  is due to a similar substitution-induced change in  $B_2^0$ . It is worth recalling that though the preferred orientation of both  $\text{ErNi}_2\text{B}_2\text{C}$  and  $\text{TbNi}_2\text{B}_2\text{C}$  is along  $\langle 100 \rangle$ , the former compound [with  $B_2^0 = -0.016 \text{ meV}$  (Ref. 35)] orders into a transverse SDW state while the latter one [with  $B_2^0 = 0.098 \text{ meV}$  (Ref. 35)] orders into a longitudinal SDW state.

Finally, it is noted that the  $3d$  doping, in contrast to the case of the Lu and Tb substitutions, does not induce an appreciable decrease in the intensity of the high-order harmonics, possibly because there is no change in  $B_2^0$  and that such a low substitution occurs within the  $\text{Ni}_2\text{B}_2$  layers, the centre of which is  $5 \text{ \AA}$  away from that of the magnetic ErC sheets.

## V. CONCLUSIONS

This work investigated the stability of the modulated states in  $\text{ErNi}_2\text{B}_2\text{C}$  against chemical perturbations. Three different substitutions were carried out namely  $\text{Er}_{0.9}\text{Co}_{0.1}\text{Ni}_2\text{B}_2\text{C}$ ,  $\text{Er}_{0.8}\text{Tb}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ , and  $\text{Er}_{0.8}\text{Lu}_{0.2}\text{Ni}_2\text{B}_2\text{C}$ . The experimental evidences from the neutron powder diffraction analysis reveal that the most important difference between the magnetic structures of the  $3d$ -doped and  $4f$ -doped class of compounds is that, down to 1.6 K, higher order harmonics are evident in the former while absent in the latter. Assuringly, these diffraction results are quite consistent with our previous bulk magnetic and specific heat measurements. Based on the combined evidences of these studies, it is inferred that a 5% Co substitution within the  $\text{Ni}_2\text{B}_2$

planes hardly affects the squaring-up of the sine-modulated magnetic structure or the stabilization of the weak ferromagnetism at and below  $T_{WF}$ . On the other hand, doping with 20% isoelectronic  $4f$  ions stabilizes only the pure sinusoidal harmonic; evidently suppressing the squaring-up process down to temperatures below 1 K.

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