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V.N. Nguyen, F. Tchéou, J. Rossat-Mignod. Evidence for a modulated ordering in the intermetallic compounds TbGe0,2Si0,8 and ErSi of CrB-type. Journal de Physique, 1984, 45 (1), pp.163-167. 10.1051/jphys:01984004501016300. jpa-00209733

HAL Id: jpa-00209733 https://hal.science/jpa-00209733v1

Submitted on 4 Feb 2008

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Evidence for a modulated ordering in the intermetallic compounds TbGe_{0.2}Si_{0.8} and ErSi of CrB-type

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(Reçu le 29 juillet 1983, accepté le 22 septembre 1983)

Résumé. — Les composés TbGe_{0,2}Si_{0,8} et ErSi s'ordonnent antiferromagnétiquement au-dessous des températures de Néel respectives $T_N = 55$ K et 11,5 K. Entre T_N et T_{IC} ($T_{IC} = 43,5$ K et 7,25 K pour TbGe_{0,2}Si_{0,8} et ErSi), les composés étudiés s'ordonnent selon

Entre T_N et T_{IC} ($T_{IC} = 43,5$ K et 7,25 K pour TbGe_{0,2}Si_{0,8} et ErSi), les composés étudiés s'ordonnent selon une double sinusoïde définie par le vecteur de propagation $\mathbf{k} = [k_x, 0, \frac{1}{2}]$. La composante irrationnelle k_x varie avec la température et à T_{IC} a lieu une transition magnétique vers une structure commensurable décrite par le vecteur de propagation $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$. A toute température inférieure à T_N , les moments des ions terbium sont dans le plan (\mathbf{a}, \mathbf{c}) près de l'axe \mathbf{c} , alors que les moments des ions d'erbium sont dirigés selon l'axe \mathbf{a} .

Abstract. — Neutron diffraction experiments have been performed on powder samples of $\text{TbGe}_{0.2}\text{Si}_{0.8}$ and ErSi. They order respectively at $T_N = 55$ K and 11.5 K with a pure sine wave modulation propagating along the [100] direction of the orthorhombic unit cell. The wave vector $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ is incommensurate with the lattice and is temperature dependent down to T_{IC} ($T_{\text{IC}} = 43.5$ K and 7.25 K respectively for TbGe_{0.2}Si_{0.8} and ErSi) at which a magnetic transition to a commensurate phase described, by the wave vector $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$ takes place.

At any temperatures, the erbium magnetic moments are aligned along the **a**-axis, whereas for $TbGe_{0.2}Si_{0.8}$, the terbium magnetic moments are in the (**a**, **c**) plane and make an angle of 20° with the **c**-axis.

1. Introduction.

In order to investigate the magnetic interactions between the rare earth metals and silicon in alloys the equiatomic compounds R-Si (R = rare earth atom) and particularly TbGe_{0.2}Si_{0.8} and ErSi have been studied [1, 2]. From neutron diffraction and magnetization experiments, it has been shown that the compounds TbGe_{0.2}Si_{0.8} and ErSi order antiferromagnetically with Neel temperatures $T_{\rm N} = 56$ K and 11.5 K and that at 1.5 K, the magnetic structures are collinear and described by the wave vector $\mathbf{k} = \begin{bmatrix} \frac{1}{2} & 0 & \frac{1}{2} \end{bmatrix}$. In the compound ErSi, the rare earth magnetic moments are aligned along the a-axis while for $TbGe_{0,2}$ Si_{0.8} the magnetic moments of the terbium atoms lie in the (a, c) plane, close to the c-axis. A crystal field calculation using the point charge model [3] indicates that a strong anisotropy is present for terbium and erbium atoms, giving an antiferromagnetic direction along the c- and a-axis respectively for Tb and Er.

In order to check this point, the thermal evolution of the magnetic structures of these compounds has been undertaken by neutron diffraction measurements from 2.2 K up to $T_{\rm N}$.

As the low temperature magnetic structure of these compounds is not an usual antiferromagnetic structure, i.e. associated with a wave vector corresponding to a symmetry point of the Brillouin zone, we can expect at higher temperature a phase transition towards an incommensurate phase.

2. Experimental.

 $\text{TbGe}_{0.2}\text{Si}_{0.8}$ and ErSi crystallize in the space group Cmcm. At room temperature, the unit cell parameters have been found to be [1, 2] :

$$a = 4.285 \text{ Å}; \quad b = 10.572 \text{ Å}; \quad c = 3.852 \text{ Å}$$

for TbGe_{0.2}Si_{0.8} and a = 4.195 Å; b = 10.380 Å; c = 3.970 Å for ErSi.

The chemical unit cell contains four units of R-Si but, as the space group is face centred, only two R-Si units must be considered. The silicon and the rare earth atoms are both in the same 4c site of C_{2v} (2 mm) symmetry. The rare earth atoms are located in two Bravais lattices : $\mathbf{r}_1 = (0, y, \frac{1}{4})$ and $\mathbf{r}_2 = (0, \overline{y}, \frac{1}{4})$ with y = 0.143 and 0.141 respectively for Tb and Er. Polycrystalline samples of ErSi and TbGe_{0.2}Si_{0.8} were obtained from the terbium, erbium and silicon elements using previously described methods [1, 2]. The single phase nature of these compounds was established by X-ray analysis using FeK_{α} radiation. For both compounds TbGe_{0.2}Si_{0.8} and ErSi, the neutron diffraction experiments were carried out at various temperatures between 2.2 K and T_N using a monochromatic neutron beam with a wave length $\lambda = 1.415$ Å on the DN5 spectrometer installed at the Siloe reactor of the Centre d'Etudes Nucléaires de Grenoble.

3. Neutron diffraction results.

For TbGe_{0.2}Si_{0.8}, the neutron diffraction patterns are shown in figure 1 for temperatures T = 6.9 K and $T_{IC} = 41$ K, and the magnetic peaks $\left[\frac{1}{2}, 0, \frac{1}{2}\right]$, $\left[\frac{1}{2}, 1, \frac{1}{2}\right]$ and $\left[\frac{1}{2}, 2, \frac{1}{2}\right]$ are indexed using the wave vector $\mathbf{k} = \left[\frac{1}{2}, 0, \frac{1}{2}\right]$ characterizing the collinear structure obtained at 1.4 K and 4.2 K [2]. At temperatures higher than $T_{IC} = 41$ K and up to $T_N = 56$ K, a shift in the position of these magnetic peaks was detected. The peaks of type $\left\langle \frac{h}{2}, k, \frac{l}{2} \right\rangle$ for which k is odd shift towards higher Bragg angles which are denoted $\left\langle \frac{h}{2}, k, \frac{l}{2} \right\rangle^+$; whereas, those with k even denoted $\left\langle \frac{h}{2}, k, \frac{l}{2} \right\rangle^{-}$ shift towards smaller angles. This behaviour can be explained by considering that the wave vector $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ becomes incommensurate with $0 < k_x < \frac{1}{2}$. Figure 2 represents the diffraction patterns at T = 57 K (paramagnetic state above $T_{\rm N} = 56$ K) and T = 49 K (between $T_{\rm IC} = 41$ K and $T_{\rm N} = 56$ K).

For ErSi, in the temperature range 2.2 K to $T_{\rm IC} = 7.25$ K, several superlattice magnetic peaks were observed and indexed with the wave vector $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$ as shown in TbGe_{0.2}Si_{0.8} [1, 2]. At temperatures higher than $T_{\rm IC} = 7.25$ K up to $T_{\rm N} = 11.5$ K, the same shift in the magnetic peaks was detected.

The neutron pattern for ErSi differs from that of TbGe_{0.2}Si_{0.8} by the existence of two additional small lines $\langle 0, 0, \frac{1}{2} \rangle$ and $\langle 0, 1, \frac{1}{2} \rangle$ which can be indexed with the wave vector $\mathbf{k} = [0, 0, \frac{1}{2}]$ as was observed for the TmSi and DyGe_{0.2}Si_{0.8} compounds [4]. The thermal evolution of the magnetic peaks $\langle 0, 0, \frac{1}{2} \rangle$ and $\langle \frac{1}{2}, 0, \frac{1}{2} \rangle$ indicates that $T_{\rm N} = 11.5$ K is the same for both wave vectors $\mathbf{k} = [0, 0, \frac{1}{2}]$ and $\mathbf{k}' = [k_x, 0, \frac{1}{2}]$ (Fig. 3). At temperatures below $T_{\rm N} = 11.5$ K, the magnetic moments of erbium atoms associated with the wave vector $\mathbf{k} = [0, 0, \frac{1}{2}]$ are aligned along the **a**-axis, as was observed in TmSi, with a value of $1.5 \mu_{\rm B}$ at T = 2.2 K.



Fig. 1. — Thermal variation of the magnetic intensities of $[\frac{1}{2}, 0, \frac{1}{2}], [\frac{1}{2}, 1, \frac{1}{2}]$ and $[\frac{1}{2}, 2, \frac{1}{2}]$ Bragg peaks.



Fig. 2. — Diffraction powder patterns of $\text{TbGe}_{0.2}\text{Si}_{0.8}$ at T = 56 K and 49 K.

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 $(\frac{1}{2}, 0, \frac{1}{2})$ M

Fig. 3. — Thermal variation of the magnetic intensities of $[0, 0, \frac{1}{2}]$ and $[\frac{1}{2}, 0, \frac{1}{2}]$ Bragg peaks associated with $\mathbf{k}_1 = [0, 0, \frac{1}{2}]$ and $\mathbf{k}_2 = [k_x, 0, \frac{1}{2}]$ respectively for the compound ErSi.

4. Determination of the wave vector.

For TbGe_{0.2}Si_{0.8} and ErSi, the variation of the position of the magnetic reflections $\left\langle \frac{h}{2}, k, \frac{l}{2} \right\rangle$ can be accounted for by considering the wave vector with the following form $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ with $k_x = 0.5 - \varepsilon$. The k_x -values have been calculated from the position of the superlattice peaks $\langle \frac{1}{2}, 0, \frac{1}{2} \rangle^{-}, \langle \frac{1}{2}, 1, \frac{1}{2} \rangle^{+},$ $\langle \frac{1}{2}, 2, \frac{1}{2} \rangle^{-}$. The thermal variation of the k_x -value is reported in table I for $TbGe_{0.2}Si_{0.8}$ and ErSi in the temperature range $T_{\rm IC} < T < T_{\rm N}$. The wave vectors $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$ and $\mathbf{k} = [k_x, 0, \frac{1}{2}]$

which are not symmetry points of the Brillouin zone, are however located on a symmetry line of this zone.

Table I. — Temperature dependence of the k_x -component of the wave vector $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ for TbGe_{0.2}Si_{0.8} and Ersi.

TbGe _{0.2} Si _{0.8}	T (K)	4.2	41	43.5	49	54
$T_{\rm IC} = 43.5 {\rm K}$	k _x	0.5	0.5	0.464	0.444	0.443
ErSi	T (K)	2.2	7.25	7.30	10.2	11.5
$T_{\rm IC} = 7.25 {\rm K}$	k _x	0.5	0.5	0.483	0.470	0.462

5. Magnetic intensities.

In order to calculate the magnetic intensity of a Bragg peak denoted by $\left\langle \frac{h}{2}, k, \frac{l}{2} \right\rangle^{\pm}$, we must not forget to sum all the intensities of scattering vectors giving the same Bragg angle.

On the other hand, the magnetic intensities associated with the magnetic scattering vectors $\mathbf{h}^{\pm} =$

$$\mathbf{H}_{N} \pm \mathbf{k} \text{ are given by :}$$

$$I(\mathbf{H}_{N} \pm \mathbf{k}) = (0.27)^{2} * f_{m}^{2}(h^{\pm}) \times \\ \times \left[\mathbf{F}(\mathbf{h}^{\pm}) * \mathbf{F}^{*}(h^{\pm}) - \frac{(\mathbf{F}(\mathbf{h}^{\pm}).\mathbf{h}^{\pm})(\mathbf{F}^{*}(\mathbf{h}^{\pm}).\mathbf{h}^{\pm})}{\mathbf{h}^{\pm 2}} \right]$$
(1)

where $f_m(\mathbf{h}^{\pm})$ is the magnetic form factor and $\mathbf{F}(\mathbf{h}^{\pm})$ is the geometrical structure factor given by :

$$\mathbf{F}(\mathbf{h}^{\pm}) = \mathbf{F}(\mathbf{H}_{\mathrm{N}} \pm \mathbf{k}) = \sum_{i} m_{k,i} e^{2\pi i (\mathbf{H}_{\mathrm{N}} \pm \mathbf{k}) \cdot \mathbf{r}_{i}}$$
(2)

where the sum must be performed over the two Bravais lattices with the position $\mathbf{r}_i \cdot \mathbf{m}_{k,i}$ is the Fourier component associated with the Bravais lattice i and the wave vector **k**. In order to calculate $\mathbf{F}(\mathbf{h}^{\pm})$, we must consider two types of magnetic ordering :

(i) a helical ordering with :

$$\mathbf{m}_{k} = \frac{m_{0}}{2} \left(\hat{u} - i \hat{v} \right) e^{i\phi} \tag{3}$$

(ii) a sine wave modulation with :

$$\mathbf{m}_{k} = \frac{A}{2} \,\hat{u} \,\,\mathrm{e}^{i\phi} \,. \tag{4}$$

The fact that the x-component of the wave vector is not commensurate implies the magnetic structure to be either a spiral or a sine wave modulation of the magnetic moment. The former structure can be ruled out immediatly in our case owing to the large one-ion anisotropy of terbium ions in C_{2y} local symmetry [3]. The magnetic structure factors $F(h^{\pm})$ have been calculated in the case of a sine wave modulation of the moment, for the two Bravais lattices located in $\mathbf{r}_{I}(0, 0, 0)$ and $\mathbf{r}_{II}(\frac{1}{2}, \frac{1}{2} - 2y, \frac{1}{4})$. The structure factors **F(h)** for $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ are given by :

$$\mathbf{F}(\mathbf{H}_{\mathrm{N}} \pm \mathbf{k}) = \frac{A \cdot \hat{u}}{2} (1 + e^{i\alpha})$$
 (5)

where $\alpha = 2 \pi \mathbf{k} \cdot \mathbf{r}_{II} + \phi$ and ϕ represents the relative phase of the second Bravais lattice with respect to the first one; A is the amplitude of the sine wave modulation. Then, the magnetic ordering is described by a double sine wave modulation with four parameters A, ϕ and the angles θ , Φ defining the moment direction.

	Magneti	ic str	uctu	res c	of TbGe	e _{0.2} Si	D.8	and	ErSi
as	sociated	with	the	wave	vectors	5 k =	$[k_x,$	$0, \frac{1}{2}$]	and
k	$= [\frac{1}{2}, 0,$	$\frac{1}{2}$].							

The moment direction of rare earth atoms and the ϕ -value are given by comparison with the magnetic intensities.

For the compound ErSi, the fact that the intensities of the magnetic Bragg peaks $\langle hkl \rangle \pm k$ are weakly dependent on the index k and $l [I(\langle 000 \rangle + \mathbf{k})] \cong$ $I(\langle 001 \rangle + \mathbf{k})$ and $I(\langle 110 \rangle - \mathbf{k}) \cong I(\langle 130 \rangle - \mathbf{k})$ indicates that the magnetic moments are oriented along the **a**-axis. Then the only remaining parameter to be determined is the coupling between the two Bravais lattices; the best fit of the calculated intensities to the neutron diffraction data has been obtained using $\phi = 0^{\circ}$ or $\phi = 180^{\circ}$. In table II, the amplitude values of the sine wave modulation, in the temperature range T = 2.2 K and T = 11.5 K, are reported for ErSi. In table III, the observed intensities are compared with the calculated intensities (in barn/unit cell) at T = 7.3 K. The thermal variation of the amplitude for the compound ErSi is shown in figure 4.

In the case of TbGe_{0.2}Si_{0.8}, the fact that the intensities of the magnetic Bragg peaks $\langle hkl \rangle \pm \mathbf{k}$ change slowly with the index h and $k [I(\langle 000 \rangle + \mathbf{k}) \neq I(\langle 001 \rangle) + \mathbf{k}]$ indicates that the magnetic moments are aligned near the **c**-axis. On the other hand, the absence of the magnetic Bragg peak $\langle 001 \rangle + \mathbf{k}$ indicates that the magnetic moments of terbium atoms lie in the (101) plane and tilt from the **c**-axis by an angle of about 20°. The best fit of the magnetic intensities to experimental data gives rise to a reliability factor R = 6 % in the temperature range T = 1.5 K and T = 54.0 K, with the ϕ values $\phi = 0^\circ$ or 180° .



Fig. 4. — Thermal variation of the amplitude A(T) of the sine wave modulation for ErSi in the ordered state.

The calculated and observed magnetic intensities (in barn/unit cell) of the compound $\text{TbGe}_{0.2}\text{Si}_{0.8}$ at T = 49 K are given in table V. In table IV, the amplitudes of the sine wave modulation, for the compound $\text{TbGe}_{0.2}\text{Si}_{0.8}$ are given as a function of temperature. The thermal variation of this amplitude is shown in figure 5.

Table II. — Amplitude of the sine wave modulation as a function of temperature for the compound ErSi.

T (K)	2.2	4.2	5.1	6.9	7.25	7.30	8.8	10.2	11.5
<i>A</i> (μ _B)	8.30	8.24	8.20	7.70	7.46	6.80	5.97	5.4	3.03

Table III. — Observed and calculated values of the magnetic intensities (in barn/unit cell) at T = 7.3 K for ErSi ($A = 6.8 \mu_{\rm B} \pm 0.1 \mu_{\rm B}$).

$\langle h_{\rm m}, k_{\rm m}, l_{\rm m} \rangle$	I _{calculated}	Iobserved
$\langle \frac{1}{2}, 0, \frac{1}{2} \rangle^{-}$	14.10	13.40
$\langle \frac{1}{2}, 1, \frac{1}{2} \rangle^+$	31.05	31.30
$\langle \frac{1}{2}, 2, \frac{1}{2} \rangle^{-}$	30.80	37.30
$\langle \frac{1}{2}, 3, \frac{1}{2} \rangle^+$	35.00	38.30
$ \begin{array}{c} \left< \frac{1}{2}, 4, \frac{1}{2} \right>^{-} \\ \left< \frac{1}{2}, 1, \frac{3}{2} \right>^{+} \\ \left< \frac{3}{2}, 2, \frac{1}{2} \right>^{+} \end{array} \right\} $	$ \begin{array}{c} 31.8\\ 31.7\\ 10.3 \end{array} $ 73.8	72.5
$ \begin{array}{c} \left\langle \frac{3}{2}, 1, \frac{1}{2} \right\rangle^{-} \\ \left\langle \frac{3}{2}, 0, \frac{1}{2} \right\rangle^{+} \end{array} $	} 9.0	10.0
$\langle \frac{1}{2}, 0, \frac{3}{2} \rangle^{-}$	13.4	13.8

Table IV. — Amplitude of the sine wave modulation as a function of temperature for $TbGe_{0,2}Si_{0,8}$.

T (K)	7	20	41	43.5	49	54
$A(\mu_{\rm B})$	12.5	12.5	10.6	8.5	7.0	4.6

Table V. — Observed and calculated values of magnetic intensities (in barn/unit cell) at T = 49 K for TbGe_{0.2}Si_{0.8} ($A = 7.0 \mu_B \pm 0.1$).

$\langle h_{\rm m}, k_{\rm m}, l_{\rm m} \rangle$	I _{calculated}	Iobserved
$\langle \frac{1}{2}, 0, \frac{1}{2} \rangle^{-}$	20.0	18.4
$\langle \frac{1}{2}, 1, \frac{1}{2} \rangle^+$	32.6	39.4
$\langle \frac{1}{2}, 2, \frac{1}{2} \rangle^{-}$	29.6	27.0
$\langle \frac{1}{2}, 3, \frac{1}{2} \rangle^+$	33.4	30.0
$ \begin{array}{c} \left\langle \frac{1}{2}, 4, \frac{1}{2} \right\rangle^{-} \\ \left\langle \frac{1}{2}, 1, \frac{3}{2} \right\rangle^{+} \\ \left\langle \frac{3}{2}, 2, \frac{1}{2} \right\rangle^{+} \end{array} \right\} $	$ \left. \begin{array}{c} 34.8 \\ 7.3 \\ 34.9 \end{array} \right\} 77.0 $	76.0
$ \left[\begin{array}{c} \left\langle \frac{3}{2}, 1, \frac{1}{2} \right\rangle^{-} \\ \left\langle \frac{3}{2}, 0, \frac{1}{2} \right\rangle^{+} \end{array} \right] $	29.5 11.3 40.8	43.5
$\langle \frac{1}{2}, 0, \frac{3}{2} \rangle^{-}$	0.2	n.0



Fig. 5. — Thermal variation of the amplitude A(T) of the sine wave modulation for TbGe_{0.2}Si_{0.8} in the ordered state.

Then, $\text{TbGe}_{0.2}\text{Si}_{0.8}$ orders with a double transverse sine wave modulation propagating along the [100] direction with magnetic moments oriented near the **c**-axis, while the compound ErSi orders with a double longitudinal sine wave modulation propagating along the [100] direction with the magnetic moments aligned along the **a**-axis. The magnetic structure of the compound TbGe_{0.2}Si_{0.8} is represented in figure 6.

6. Conclusions.

The observed magnetic ordering, in agreement with a crystal field calculation using the point charge model, indicates that the ground state has an Ising like anisotropy along the c- and a-axis respectively for TbGe_{0.2}Si_{0.8} and ErSi [3]. This strong Ising anisotropy together with competitive exchange interactions give rise to a sine wave modulation below T_N . In figures 4 and 5, which show the thermal variation of the amplitude of the sine wave modulation for TbGe_{0.2}Si_{0.8} and ErSi, two temperature ranges can be distinguished :

(i) In the temperature range $T_{\rm IC} < T < T_{\rm N}$, the ordering characterized by the wave vector $\mathbf{k} = [k_x, 0, \frac{1}{2}]$ is a double sine wave modulation. The k_x -value is incommensurate with the lattice and is temperature dependent down to $T_{\rm IC}$. The amplitude of the sine wave modulation follows a Brillouin law



Fig. 6. — Variations of the magnetic moments describing the sine wave modulation of the magnetic structure along the three directions \mathbf{a} , \mathbf{b} and \mathbf{c} . Only the magnetic moments of the first Bravais lattice located at (000) (number 1) are visualized.

(Figs. 4 and 5) and the array of magnetic moments within the two Bravais lattices (I) and (II) is given by :

$$\mathbf{m}_n(\mathbf{I}) = A \cdot \hat{u} \sin(\alpha_0 + 2 \pi \mathbf{k} \cdot \mathbf{R}_n)$$

$$\mathbf{m}_n(\mathbf{II}) = A \cdot \hat{u} \sin(\alpha_0 + 2 \pi \mathbf{k} \cdot \mathbf{R}_n + \phi).$$

(ii) Below T_{IC} , the magnetic structure changes to a commensurate antiferromagnetic structure, described by the wave vector $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$, of the type previously reported for ErSi[1]. The magnetic structure associated with the wave vector $\mathbf{k} = [\frac{1}{2}, 0, \frac{1}{2}]$ corresponds to a peculiar modulated ordering for which it is possible to get the same moment value on each rare earth ion by locking the phase α_0 to be $\pm \pi/4$. In the compounds TbGe_{0.2}Si_{0.8} and ErSi, for decreasing temperature, we observe a continuous change in the value of the wave vector instead of the appearance of higher order harmonics as in rare earth metals.

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