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## Interpretation of the magnetic field dependence of the first-order AF–HX phase transition in NiBr<sub>2</sub>

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**Abstract.** With decreasing temperature NiBr<sub>2</sub> undergoes successively a second-order para-antiferromagnetic ( $T_N = 52 \pm 1$  K) and then a first-order antiferro–helimagnetic ( $T_{ic} = 22.8$  K) phase transition. In both ordered phases, the individual moments lie in a plane perpendicular to the crystal  $c$  axis. Using a model described in a previous paper, we interpret the variation of  $T_{ic}$  when the magnetic field is applied in the plane of the spins. This study allows us also to confirm the values of some model parameters. However, in the presence of a strong magnetic field the model must be extended to take into account the strains created by the field.

### 1. Introduction

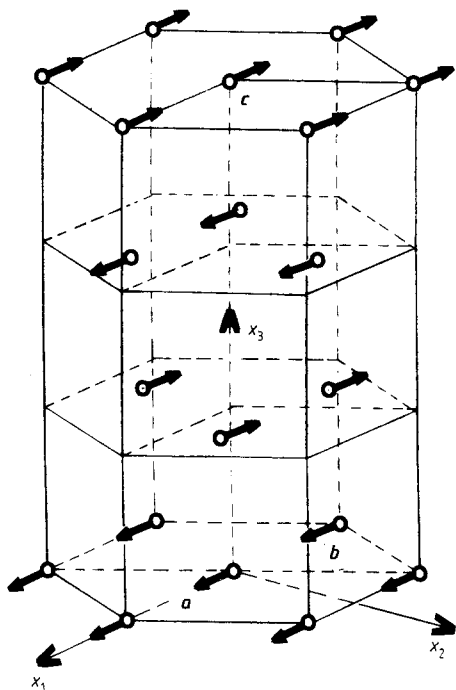
Anhydrous nickel dibromide, NiBr<sub>2</sub>, has a hexagonal layer structure of the CdCl<sub>2</sub> type with a high symmetry axis,  $c$  axis. When the temperature decreases the compound displays successively a second-order para (P)–antiferromagnetic (AF) phase transition ( $T_N = 52 \pm 1$  K, Day *et al* 1976), and then a first-order antiferro–helimagnetic (HX) transition at  $T_{ic} = 22.8$  K (Adam *et al* 1980a, Day and Ziebeck 1980). In both magnetic structures the individual spin directions are orthogonal to the  $c$  axis.

In order to investigate the first-order transition and the low-temperature magnetic phase, many experimental studies have been carried out by varying one or more of the following parameters: temperature  $T$ , uniform magnetic field  $h$  and hydrostatic pressure (Adam *et al* 1980a, Day and Vettier 1981, Adam *et al* 1981, Regnault *et al* 1982, Day *et al* 1982, Pollard *et al* 1982, Katsumata *et al* 1983, Day *et al* 1984, Tuchendler and Katsumata 1985).

In a previous paper (Bettachy and Nasser 1989, referred to as P1), we have presented a model which interprets the first-order (AF)–(HX) phase transition in zero applied field at atmospheric pressure. In this article we use this model to interpret the effects of an applied magnetic field on this phase transition, the field being applied within the easy magnetisation plane (in-plane magnetic field). One of the topics of this article is the study of the magnetic field–temperature phase diagram. Another goal is to obtain a new determination for the values of some model parameters. Comparing these new values with the corresponding values obtained by another method (P1) allows the model to be checked and its limits of validity to be defined.

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**Figure 1.** Crystal and AF structure of  $\text{NiBr}_2$ .  $a$ ,  $b$  and  $c$  are the crystalline axes of the hexagonal cell containing 3 formula units:  $c$  is the high-symmetry axis (3). Open circles, nickel ions; bromide ions are not represented. The AF structure consists of ferromagnetic sheets coupled antiferromagnetically (sequence  $+ - + -$  along the  $c$  axis), the spins lie within the sheets (easy magnetisation plane).  $T_N = 52.1$  K.

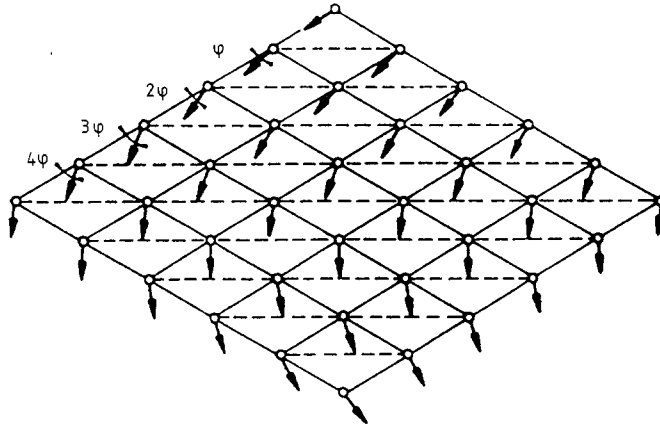
In the following section we give a summary of the experimental data obtained in  $\text{NiBr}_2$  below  $T_N$  at atmospheric pressure in the presence of an applied field. In § 3 we give a theoretical interpretation of these data. The last section is devoted to the conclusion.

## 2. In-plane magnetic field: experimental phase diagram at atmospheric pressure

The primitive nuclear cell of  $\text{NiBr}_2$  is rhombohedral with one molecule per unit cell. In this study it is more convenient to use a hexagonal cell with 3 formula units per unit cell. This hexagonal cell and the crystalline axes  $a$ ,  $b$  and  $c$  are illustrated in figure 1. The following wavevectors  $q$  are labelled in the reciprocal axes  $a^*$ ,  $b^*$  and  $c^*$ .

The AF structure of  $\text{NiBr}_2$  is similar to that of  $\text{NiCl}_2$ . It consists of ferromagnetic planes which are coupled antiferromagnetically as shown in figure 1 (Day *et al* 1976). The wavevector of this structure is  $(0, 0, \frac{2}{3})$ . Below  $T_{ic}$  spins are rotated in the hexagonal planes giving an HX structure with a wavevector  $(\tau, \tau, \frac{2}{3})$ . The HX structure of a hexagonal sheet is illustrated in figure 2, two consecutive hexagonal sheets being antiferromagnetically oriented. Both magnetic structures are close in the sense that the parameter  $\tau$  which is zero in the AF phase takes the value  $8.96 \pm 0.33 \times 10^{-3}$  at  $T_{ic}$  and then increases with decreasing temperature becoming 0.027 at 4.2 K. The thermal variations of the parameters  $\tau$  and  $m^2$ , the square of the magnetisation per ion, have been measured between  $T_N$  and 4.2 K (Adam *et al* 1980a).

An in-plane applied magnetic field reduces the values of the transition temperature  $T_{ic}$ . The magnetic field–temperature phase diagram has been determined at different pressure values (Adam *et al* 1981). At atmospheric pressure,  $T_{ic}$  is  $22.4 \pm 0.5$  and  $20.8 \pm 0.5$  K when the magnetic field is respectively 0.5 and 1.0 T. The extrapolation at zero magnetic field of the transition line gives the value  $23.8 \pm 0.5$  K, which is slightly



**Figure 2.** HX structure of NiBr<sub>2</sub> from Regnault *et al* (1982). Below  $T_{ic} = 22.8$  K, each ferromagnetic sheet becomes helimagnetic as shown in the figure. The sequence  $+ - + -$  along the  $c$  axis is maintained. The helimagnetic angle  $\varphi$  is related to the parameter  $\tau$  by  $\varphi = 2\pi\tau$  and varies with the temperature. At 4.2 K the angle  $\varphi$  is close to  $10^\circ$ .

different from the value 22.8 K directly obtained by Adam *et al* (1980a) for  $T_{ic}$  in zero applied field. At 4.2 K, a  $2.74 \pm 0.02$  T applied field value induces a first-order HX-AF phase transition.

### 3. Theoretical interpretation

We use the Hamiltonian  $\mathcal{H}_t$ :

$$\mathcal{H}_t = \mathcal{H}_0 + \mathcal{H}_1 \quad (1)$$

where  $\mathcal{H}_1$  is considered as a perturbation to  $\mathcal{H}_0$ . The principal Hamiltonian  $\mathcal{H}_0$  contains the exchange interactions, while the perturbing term  $\mathcal{H}_1$  contains in-plane anisotropy, dipolar and Zeeman interactions.

All the above interactions, except the Zeeman term, have been studied in a previous article (P1). Here we only give explanations for the Zeeman interaction.

#### 3.1. Study of the principal Hamiltonian $\mathcal{H}_0$

We take for  $\mathcal{H}_0$  the expression

$$\mathcal{H}_0 = \sum_{\langle R, R' \rangle} -2j_{RR'} \mathbf{S}_R \cdot \mathbf{S}_{R'} \quad (2)$$

where  $\mathbf{S}_R$  is the spin operator at the site  $R$  ( $S = 1$ );  $\sum_{\langle R, R' \rangle}$  means the sum over pairs of spins. Concerning the exchange interactions  $j_{RR'}$ , we take into account the first-, second- and third-neighbour in-plane exchange integrals  $j_1, j_2$  and  $j_3$  respectively together with the interplane exchange integral  $j_4$  (Regnault *et al* 1982, P1).

In our model, the exchange parameters are taken to be linear functions of the components  $\varepsilon_1, \varepsilon_2$  and  $\varepsilon_3$  of the crystal strain tensor defined in the orthogonal frame  $\{0\mathbf{x}_i, i = 1, 2, 3\}$  such that  $0\mathbf{x}_3$  is parallel to the  $c$  axis and  $0\mathbf{x}_2$  is contained in the mirror plane  $m$  of the Ni<sup>2+</sup> ion point group (figure 1). We insist on the fact that, in our model, the only strains that are considered are those related to the magneto-elastic coupling (the background thermal dilatation is neglected).

By symmetry arguments we have

$$j_i = j_i^0 + (\partial j_i / \partial \varepsilon_1)(\varepsilon_1 + \varepsilon_2) + (\partial j_i / \partial \varepsilon_3)\varepsilon_3 \quad (i = 1 \text{ to } 4). \quad (3)$$

By symmetry, we always have  $\varepsilon_1 = \varepsilon_2$ ; so we never mention the strain component  $\varepsilon_2$ . The parameters  $j_i^0$ ,  $\partial j_i / \partial \varepsilon_1$  and  $\partial j_i / \partial \varepsilon_3$  are constant. The parameter  $j_i^0$  corresponds to the value of the exchange integral  $j_i$  in the P phase.

We look for solutions of the form

$$\langle S_R \rangle = m \exp(i\mathbf{q} \cdot \mathbf{R})\mathbf{u}$$

where  $\langle S_R \rangle$  is the thermal value of the spin  $S_R$  at thermodynamic equilibrium,  $\mathbf{u}$  is a constant unit vector contained in the spin rotating plane,  $\mathbf{q}$  is a wavevector of the form  $(\tau, \tau, k)$  and  $m$  is a scalar parameter which does not depend on the site—the absolute value of  $m$  is the magnetisation per site ( $|m| \leq 1$ ).

To obtain the values of the parameters  $m$ ,  $\mathbf{q}$ ,  $\varepsilon_1$  and  $\varepsilon_3$  we have to solve a system of equations composed by the self-consistent mean field equation and by the equations obtained by minimising a variational free energy  $\tilde{F} = \tilde{F}(T, \tau, k, \varepsilon_1, \varepsilon_3)$  which contains the magnetic and the elastic free energy of the system.

The minimisation of the Fourier transform of the exchange interactions leads to the two following solutions (P1, Regnault *et al* 1982):

$$\mathbf{q}_\alpha = (0, 0, \frac{3}{2})$$

and

$$\mathbf{q}_\beta = (\tau, \tau, \frac{3}{2})$$

with

$$\cos 2\pi\tau = 1 - (2\pi\tau)^2/2 = J_1/J_2 \quad (4)$$

where the parameters  $J_1$  and  $J_2$  are linear combinations of the exchange integrals, and are defined by

$$J_1 = j_1 - 15j_2 - 32j_3 - j_4 \quad (4a)$$

$$J_2 = -2(j_1 + 12j_2 + 22j_3). \quad (4b)$$

In the same way as the exchange integrals  $j_i$ , the linear combinations  $J_1$  and  $J_2$  are linear functions of the strain components  $\varepsilon_1$ ,  $\varepsilon_2$  and  $\varepsilon_3$ . So we have

$$J_i = J_i^0 + (\partial J_i / \partial \varepsilon_1)(\varepsilon_1 + \varepsilon_2) + (\partial J_i / \partial \varepsilon_3)\varepsilon_3 \quad (i = 1 \text{ and } 2). \quad (5)$$

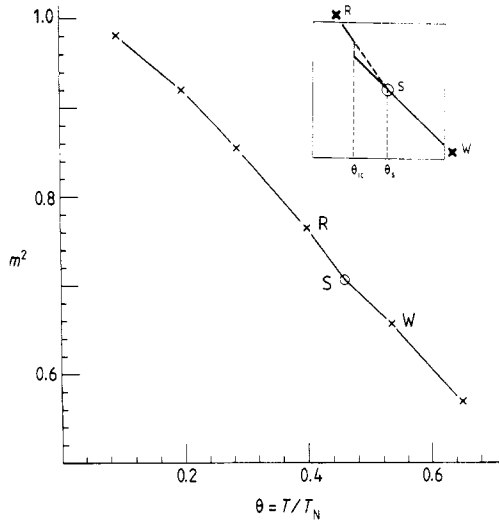
The wavevectors  $\mathbf{q}_\alpha$  and  $\mathbf{q}_\beta$  describe respectively the AF structure (here also called  $\alpha$  solution) and the HX structure ( $\beta$  solution). Both solutions have been studied in our previous paper (P1). Here we only give the results which are necessary for the following.

The relation (4) shows that the  $\beta$  solution does not exist when the ratio  $J_1/J_2$  is negative or is bigger than unity. The fundamental hypothesis of our model is

$$J_1^0/J_2^0 > 1.$$

With this hypothesis, the  $\alpha$  solution exists between  $T_N$ , the Néel temperature, and 0 K, while the  $\beta$  solution exists only below a threshold temperature value  $T_s$  which depends on the ratio  $J_1^0/J_2^0$ , with  $T_s \leq T_N$ . At  $T_s$ ,  $J_1 = J_2$  and both solutions are the same.

By fitting the thermal variations of the parameters  $\tau$  and  $m^2$ , the square of the magnetisation per site (Adam *et al* 1980a), we have obtained the following values:  $m_s^2 = 0.706 \pm 0.001$  and  $T_s = 23.9 \pm 0.4$  K (P1), where  $m_s^2$  is the value of  $m^2$  at  $T_s$ .



**Figure 3.** Thermal variation of  $m^2$  in the HX phase and for the two lowest temperature values of the AF phase:  $\theta$  is the reduced temperature  $\theta = T/T_N$ . The crosses represent experimental data in zero applied field. The coordinates of the open circle S are  $(m_s^2, \theta_s)$ . The full curve corresponds to a linear interpolation between data. This figure displays the slope discontinuity of the curve  $m^2(\theta)$  at S. The inset shows the magnetisation discontinuity at the transition temperature  $\theta_{ic}$ : the broken curve between  $\theta_{ic}$  and  $\theta_s$  cannot be observed. The difference  $\theta_s - \theta_{ic}$  depends on the sum of the in-plane anisotropy, the dipolar and the Zeeman interactions.

For the following we introduce the reduced temperature  $\theta = T/T_N$ , assuming no error on the denominator. We also suppose that  $T_s$ ,  $m_s^2$  and the related parameters have no associated uncertainties. So we have  $m_s^2 = 0.706$ ,  $B_s = (m_s^2)^{1/2} = 0.840$ ,  $x_s = 1.934$ ,  $B'_s = 0.170$  and  $\theta_s = 0.460$ . The value  $x_s$  is obtained by solving the equation  $B(x_s) = B_s$ , where  $B$  is the Brillouin function for a spin  $S = 1$ ; the value of  $B'_s$  is the value of the Brillouin function derivative at  $x_s$ ; and  $\theta_s$  is the value of the threshold reduced temperature.

Figure 3 displays the thermal variation of the parameter  $m^2$ . The crosses correspond to experimental data (Adam *et al* 1980a). The point W belongs to the AF phase, and the point R to the HX phase. The coordinates of the point S are  $m_s^2$  and  $\theta_s$ . We call the point S the threshold point. As predicted by the model, the curve  $m^2(\theta)$  displays a slope discontinuity at the point S.

Below  $\theta_s$  and near  $\theta_s$ ,  $F_\alpha - F_\beta$ , the difference of the  $\alpha$  and  $\beta$  free energies related to  $\mathcal{H}_0$  is given by (P1):

$$F_\alpha - F_\beta = \frac{1}{2}K_s(\theta - \theta_s)^2 \quad (\theta \leq \theta_s). \quad (6)$$

The parameter  $K_s$  is proportional to the slope discontinuity of the magnetisation at  $\theta_s$ . We obtain for  $K_s$  a central value close to  $3.5 \times 10^{11}$  Hz.

At 0 K,  $F_\alpha - F_\beta$  is given by (P1):

$$F_\alpha - F_\beta = \frac{1}{2}K_0\varphi^2 \quad (T = 0 \text{ K}) \quad (7)$$

where  $\varphi$  is the value of the helimagnetic angle at 0 K. This value is close to  $\varphi_0$ , the value at 4.2 K. From the data obtained by Adam *et al* (1980a) we deduce  $\varphi_0^2/2 = 0.0144 \pm 0.001$ . The parameter  $K_0$  is the sum of three terms, the main one being proportional to the magnetisation slope discontinuity. We obtain for  $K_0$  a central value close to  $5.3 \times 10^{12}$  Hz.

We now have to take into account the perturbing term  $\mathcal{H}_1$ .

### 3.2. Total free energies difference for both $F_{i\alpha} - F_{i\beta}$

Calling  $F_{i\alpha}$  and  $F_{i\beta}$  the free energies related to the total Hamiltonian  $\mathcal{H}_i$  for the respective solutions  $\alpha$  and  $\beta$ , we have

$$F_{i\alpha} - F_{i\beta} = F_\alpha - F_\beta - U \quad (8)$$

where the free energy  $U$  is obtained by considering  $\mathcal{H}_1$  as a perturbation compared to  $\mathcal{H}_0$ .

For  $\mathcal{H}_1$  we take

$$\mathcal{H}_1 = \mathcal{H}_a + \mathcal{H}_d + \mathcal{H}_Z$$

where  $\mathcal{H}_a$  is a one-ion in-plane anisotropy interaction coming from the crystalline field acting on the  $\text{Ni}^{2+}$  ion,  $\mathcal{H}_d$  is the dipolar interaction and  $\mathcal{H}_Z$  is the Zeeman interaction.

We deduce  $U_a$ ,  $U_d$  and  $U_Z$  in the following way:

$$\begin{aligned} -U_a &= \langle \mathcal{H}_a \rangle_{0\alpha} - \langle \mathcal{H}_a \rangle_{0\beta} \\ -U_d &= \langle \mathcal{H}_d \rangle_{0\alpha} - \langle \mathcal{H}_d \rangle_{0\beta} \\ -U_Z &= -\frac{1}{2}(\chi_\alpha - \chi_\beta)h^2 = -\bar{U}_Z h^2 \end{aligned}$$

where the mean values, denoted by angular brackets, are made with the unperturbed  $\alpha$  and  $\beta$  solutions,  $h^2$  is the square of the norm of the in-plane applied field  $\mathbf{h}$ , and  $\chi_\alpha$  and  $\chi_\beta$  are the magnetic susceptibilities for the respective solutions  $\alpha$  and  $\beta$ . For  $U$  we then have the relation

$$U = U_a + U_d + U_Z.$$

We neglect the strains related to the interactions contained in  $\mathcal{H}_1$ . In our previous paper (P1) we have studied  $U_a$  and  $U_d$ .

**3.2.1. In-plane anisotropy contribution  $U_a$ .** For this contribution we have

$$\begin{aligned} U_a(\theta) &= U_a(\theta_s^-) = 0.175\delta \cos 2\psi \quad (\text{near } \theta_s) \\ U_a(0 \text{ K}) &= 0.25\delta \cos 2\psi. \end{aligned}$$

The parameter  $\delta$ , which is the strength of the anisotropy interaction, has a positive sign. The angle  $\psi$  is the angle between the spin direction in the AF structure  $\mathbf{u}$ , and the easy axis related to the in-plane anisotropy interaction. In a zero applied field  $\psi = 0$ ; but in the presence of a sufficiently strong in-plane applied field the AF spin direction becomes orthogonal to the applied field direction, and then it depends on the field direction.

**3.2.2. Dipolar contribution  $U_d$ .** In a previous article, we calculated the dipolar contribution at different temperatures. Calling  $U_d(T)$  the dipolar contribution to the temperature value  $T$ , we established that  $U_d(T_s) = 0$ ,  $U_d(22.8) = 6.78 \pm 0.17$ ,  $U_d(22.4) = 9.47 \pm 0.24$ ,  $U_d(20.8) = 19.78 \pm 0.51$  and  $U_d(4.2) = 697 \pm 166$  MHz. The dipolar contribution depends on the temperature via the helimagnetic angle  $\varphi$ .

**3.2.3. Zeeman contribution  $U_Z$ .** We have to calculate the magnetic susceptibilities of the AF and HX structures  $\chi_\alpha$  and  $\chi_\beta$  respectively. For  $\chi_\alpha$  we limit the study to the antiferromagnetic perpendicular susceptibility  $\chi_\perp$ .

For  $\chi_\alpha$  and  $\chi_\beta$  we have the following expressions:

$$\chi_\alpha = -(g_\alpha \mu_B)^2 / s_3 \quad (9)$$

and

$$\chi_\beta = -\frac{1}{2}(g_\beta \mu_B)^2 (s_1 + s_3 - 2s_2) / s_1 s_3 - s_2^2 \quad (10)$$

with

$$\begin{aligned} s_1 &= \tilde{j}(0) + \tilde{j}(2\mathbf{q}) + g\mu_B(\partial m / \partial h)^{-1} \\ s_2 &= \tilde{j}(0) - \tilde{j}(2\mathbf{q}) \end{aligned}$$

$$s_3 = \tilde{j}(0) + \tilde{j}(2q) + g\mu_B h/m$$

and

$$g\mu_B h = -2m\tilde{j}(q) \quad g\mu_B(\partial m/\partial h)^{-1} = -kT(B'(x))^{-1}.$$

In the above relations,  $\tilde{j}(q)$  is the Fourier transform of the exchange interactions. We use  $q_\alpha$  for  $\chi_\alpha$  and  $q_\beta$  for  $\chi_\beta$ ; for the  $\alpha$  solution  $\tilde{j}(0) = \tilde{j}(2q_\alpha)$ ;  $\mu_B$  is the Bohr magneton;  $g_\alpha$  and  $g_\beta$  are the spectroscopic factors for the  $\alpha$  and the  $\beta$  solutions respectively; the parameter  $x$ , which is the Brillouin function argument, depends on the solution: we use  $x_\alpha$  for  $\chi_\alpha$  and  $x_\beta$  for  $\chi_\beta$ ;  $B'(x)$  is the derivative with respect to  $x$  of the Brillouin function for  $S = 1$ .

We can show that  $s_1$ ,  $s_2$  and  $s_3$  verify the following relations:

$$\left. \begin{aligned} s_1 &= 24j_4 + 2(J_3 + J_4) - s_2 + g\mu_B(\partial m/\partial h)^{-1} \\ s_2 &= 4(J_4 + 8j_4)\varphi^2/2 \\ s_3 &= 24j_4 + 2J_4(\varphi^2/2) - s_2 \end{aligned} \right\} \quad (11)$$

In the relations (11), the parameters  $J_3$  and  $J_4$  are linear combinations of the exchange integrals, and they are temperature dependent.

For  $g_\alpha$  we use the value  $2.27 \pm 0.02$  over the temperature range from  $\theta_s$  to 0 K; for  $g_\beta$  we use the value  $2.27 \pm 0.02$  at  $\theta_s$  and near  $\theta_s$ , and the value  $2.06 \pm 0.02$  at 4.2 K (Adam *et al* 1980b, 1985, P1).

We calculate  $\chi_\alpha$  at 4.2 K by using, in relations (9) and (11), the value zero for  $\varphi^2$  and for  $j_4$  its experimental value at 4.2 K,  $2j_4 = 0.042(5)$  THz (Day *et al* 1984). We obtain for  $\chi_\alpha$  at 4.2 K the value  $20.039 \pm 2.739$  Hz G<sup>-2</sup>. Neglecting the thermal variations of the parameters  $j_4$  and  $g_\alpha$ ,  $\chi_\alpha$  has this last value over the temperature range from 4.2 K to  $\theta_s$ .

At low temperature the parameters  $g\mu_B(\partial m/\partial h)^{-1}$  are very big:  $g\mu_B(\partial m/\partial h)^{-1} \rightarrow \infty$  for  $x \rightarrow \infty$ . So, from relation (10) we deduce for  $\chi_\beta$  at 0 K the expression

$$\chi_\beta = -\frac{1}{2}(g_\beta\mu_B)^2/s_3. \quad (12)$$

We use relation (12) for calculating the value of the parameter  $\chi_\beta$  at 4.2 K. On replacing, in relations (12) and (11),  $\varphi^2/2$  by  $\varphi_0^2/2$ ,  $j_4$  by its experimental value and  $J_4$  by the value obtained by using the relation  $J_4 = -4J_2 \varphi_0^2/2$  with  $J_2 = 1.883 \pm 0.171$  THz (P1), we obtain for the ratio  $\chi_\beta\chi_\alpha^{-1}$  the value  $0.434 \pm 0.002$  at 4.2 K. This value is different from the 0.58 mentioned by Regnault *et al* (1982). Using the calculated values obtained for  $\chi_\alpha$  and for the ratio  $\chi_\beta\chi_\alpha^{-1}$ , we obtain the value  $\tilde{U}_Z(4.2) = 5.67 \pm 0.80$  Hz G<sup>-2</sup>.

At  $\theta_s^-$ ,  $\varphi$  is equal to zero, and from relations (10) and (11) we deduce the relation

$$\chi_\beta = -\frac{1}{2}(g_\alpha\mu_B)^2 \left( \frac{1}{s_1} + \frac{1}{s_3} \right)_s \quad (13)$$

with

$$\begin{aligned} (s_1)_s &= 24j_4 + 2(J_3 + J_4)_s - kT_s(B'_s)^{-1} \\ (s_3)_s &= 24j_4 \end{aligned}$$

where  $(J_3 + J_4)_s$  is the value at  $\theta_s$  of the parameters  $(J_3 + J_4)$ . We can replace  $(J_3 + J_4)_s$  by  $kT_s x_s B_s^{-1}$ . Using for  $j_4$  the experimental value obtained at 4.2 K, at  $\theta_s^-$  we obtain for the ratio  $\chi_\beta\chi_\alpha^{-1}$  the value  $0.61 \pm 0.01$  and for  $\tilde{U}_Z$  the value  $\tilde{U}_Z(\theta_s^-) = 3.91 \pm 0.63$  Hz G<sup>-2</sup>.

### 3.3. AF-HX phase transitions near $\theta_s$

For the AF-HX phase transition which occurs near  $\theta_s$ , the reduced transition temperature  $\theta_{ic}$  is obtained by solving the following equation:



$$\frac{1}{2}K_s(\theta_{ic} - \theta_s)^2 - U(\theta_{ic}) = 0.$$

In a zero applied field we have  $U = U_a + U_d$ , and  $\theta_{ic} = 0.439 \pm 0.002$ . Using the approximate value  $-0.175\delta$  for  $U_a(\theta_{ic})$  and the value  $U_d(22.8)$  for  $U_d(\theta_{ic})$  we obtain the relation

$$\frac{1}{2}K_s(\theta_{ic} - \theta_s)^2 - 0.175\delta - U_d(22.8) = 0. \quad (14)$$

In a 0.5 T applied field we have  $U = U_a + U_d + U_z$ , and  $\theta_{ic} = 0.431 \pm 0.01$ . Using  $U_a = 0.175\delta \cos 2\psi$ ,  $U_d = U_d(22.4)$  and  $U_z = \tilde{U}_z(\theta_s^-)h^2$  we obtain

$$\frac{1}{2}K_s(\theta_{ic} - \theta_s)^2 - 0.175\delta \cos 2\psi - U_d(22.4) - \tilde{U}_z(\theta_s^-)h^2 = 0. \quad (15a)$$

In the same way, in a 1.0 T applied field we obtain

$$\frac{1}{2}K_s(\theta_{ic} - \theta_s)^2 - 0.175\delta \cos 2\psi - U_d(20.8) - \tilde{U}_z(\theta_s^-)h^2 = 0 \quad (15b)$$

with  $\theta_{ic} = 0.4 \pm 0.0096$ . We suppose that the angle  $\psi$  is the same in equations (15a) and (15b). For  $\theta_s$  we have the value 0.460.

From relations (15a) and (15b) we deduce for  $K_s$  a positive mean value close to  $2.2 \times 10^{11}$  Hz, and for  $0.175\delta \cos 2\psi$  a negative mean value close to  $-1.2 \times 10^7$  Hz. Using this last value for  $K_s$  in relation (14), we obtain for  $0.175\delta$  a positive mean value close to  $4.6 \times 10^7$ . From the values obtained for  $0.175\delta$  and for  $0.175\delta \cos 2\psi$ , we deduce for  $\delta$  a positive mean value close to  $2.6 \times 10^8$  Hz and for  $\cos 2\psi$  a negative mean value close to  $-0.25$  corresponding to an angle  $\psi$  close to  $52^\circ$ .

The negative value obtained for  $\cos 2\psi$  means that, due to the applied field, the spin direction in the AF phase is not the anisotropy easy axis. So in the experimental study of Adam *et al* (1981) the anisotropy contribution does not favour the (AF) structure below  $\theta_s$ . So, if we set  $h = 0$  in the relations (15a) and (15b) we obtain for the total free energy difference,  $F_{t\alpha} - F_{t\beta}$ , below and near  $\theta_s$  a positive value given by

for  $\theta \leq \theta_s$ :

$$F_{t\alpha} - F_{t\beta} = \frac{1}{2}K_s(\theta - \theta_s)^2 - 0.157\delta \cos 2\psi - U_d(\theta) > 0.$$

So  $F_{t\alpha} - F_{t\beta}$  can be equal to zero only for  $\theta = \theta_s$ , as then the dipolar and the anisotropy contributions are both equal to zero, which means that the HX phase appears for  $\theta_{ic} = \theta_s$ . Extrapolation of the experimental data for  $h = 0$  in the  $(h, T)$  phase diagram gives the temperature value  $23.8 \pm 0.5$  K (Adam *et al* 1981). In their paper, the authors consider that this value is in agreement with the value 22.8 K obtained for the temperature transition value by neutron diffraction (Adam *et al* 1980a) in zero applied field. In our model both values, 22.8 and  $23.8 \pm 0.5$  K, correspond to two different notations: the first value corresponds to the actual phase transition temperature value for a zero applied field while the second one corresponds to the threshold temperature value  $T_s$ , previously found equal to  $23.9 \pm 0.4$  (P1). So the temperature value obtained by extrapolation at  $h = 0$  in the phase diagram  $(h, T)$  corresponds to a measurement of the threshold temperature  $T_s$  when  $\cos 2\psi$  is negative.

The value  $\sim 10^8$  Hz found for  $\delta$ , the in-plane anisotropy, is much smaller than the values obtained for the exchange integrals  $j_1, j_2, j_3$  and  $j_4$  and much smaller than the value obtained for the anisotropic interaction which maintains the spin direction within the  $(0, 0, 1)$  planes (Day *et al* 1984). Up to now, no value has been proposed for  $\delta$ , but many authors have affirmed that  $\delta$  is very small (Katsumata and Date 1969, Adam *et al* 1980b). Moreover the value obtained here for  $\delta$  is compatible with that obtained in our previous paper (P1):  $4.8 \times 10^8$  Hz.

The present value obtained for  $K_s$ ,  $\sim 2.2 \times 10^{11}$  Hz, is compatible with that obtained previously,  $\sim 3.5 \times 10^{11}$  Hz (P1). However, the  $K_s$  values of 2.2 and  $3.5 \times 10^{11}$  Hz lead

to values close to  $0.09R$  and  $0.15R$  respectively for  $L$ , the transition latent heat. Since the experimental value proposed for  $L$  is  $\sim 0.013R$  (White and Staveley 1982), it seems preferable to use the value  $2.2 \times 10^{11}$  Hz for  $K_s$ . We must note that the latent heat experimental value is probably not very accurate.

### 3.4. HX-AF phase transition at 4.2 K

The magnetic field value  $h$  which induces an HX-AF phase transition at 4.2 K approximately verifies the relation

$$\frac{1}{2}K_0\varphi^2 - 0.25\delta \cos 2\psi - U_d(4.2) - \tilde{U}_z(4.2)h^2 = 0$$

with  $h = 2.74 \pm 0.02 \times 10^4$  G, the angle  $\psi$  having the same value as in the equations (15). Using the values obtained for  $\delta \cos 2\psi$ ,  $U_d(4.2)$  and  $\tilde{U}_z(4.2)$  we deduce a mean value for  $K_0\varphi^2/2$  close to  $4.9 \times 10^9$  Hz.

Replacing  $\varphi$  by  $\varphi_0$ , we obtain a mean value for  $K_0$  close to  $3.4 \times 10^{11}$  Hz. This value is fifteen times lower than the value  $5.3 \times 10^{12}$  Hz deduced from experimental data obtained in zero applied field. We consider that this discrepancy between both values of  $K_0$  is due to the fact that we do not take into account the strain tensor created by the applied magnetic field. In the HX-AF transition at 4.2 K, the applied magnetic field is big (2.74 T) and the strains created by this field cannot be neglected. We think that these strains modify the value of the helimagnetic angle  $\varphi$  in the same way that the application of a hydrostatic pressure reduces the value of this angle (Day and Vettier 1981). If we take the value  $\sim 53 \times 10^{11}$  Hz for  $K_0$  obtained in zero applied field, we deduce from the result  $\frac{1}{2}K_0\varphi^2 \sim 49.4 \times 10^8$  Hz, the value  $9.3 \times 10^{-4}$  for the parameter  $\varphi^2/2$  in a 2.74 T applied field. Comparing this value to the value  $\varphi_0^2/2 \sim 0.01439$ , we conclude that the relative variation of the parameters  $\varphi$  as a function of the applied magnetic field is  $\Delta\varphi/\varphi \sim -1.7 \cdot 10^{-2} \text{ kG}^{-1}$  at 4.2 K. This prediction could easily be tested by experiment.

## 4. Conclusion

In this article, using a model described in our previous paper, we have interpreted the experimental data obtained in NiBr<sub>2</sub> concerning the variation of the AF-HX phase transition temperature as a function of an applied magnetic field. We have shown that the transition temperature value  $23.8 \pm 0.5$  K obtained by extrapolation of the AF-HX transition line to zero field does not give the real zero applied field AF-HX transition temperature ( $T_{ic} = 22.8$  K), but rather it corresponds to  $T_s$ , the threshold temperature value of our model. Moreover this study allows us to confirm the values of some model parameters.

Only the HX-AF phase transition observed at 4.2 K in the presence of a 2.74 T applied magnetic field is not quantitatively interpreted by our model. However, we think that this is due to the fact that we have neglected the strains created by the field. We are presently examining this problem.

We believe that some experimental studies, done in the presence of a magnetic field applied within the spin rotating plane, can give interesting new results. For example: (i) at 4.2 K we predict a reduction of the order of 2% per kG for the helimagnetic angle; (ii) near 22.8 K a study of the AF-HX phase transition as a function of the magnetic field direction will provide a direct determination of the in-plane anisotropy.

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