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Reorientation of magnetic moments under mechanical stresses in FeCl_2 at 0 K : theoretical predictions

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Résumé. — Nous montrons dans le cas de FeCl_2 — un composé antiferromagnétique uniaxe pour $T \leq T_N$ (≈ 25 K) — qu'on peut, par application de contraintes mécaniques, modifier la symétrie du champ cristallin des ions Fe^{+2} et par là modifier l'orientation des moments de ces ions. Nous donnons en fonction de la variation du champ cristallin δW , l'expression des composantes du tenseur qui relie la variation du moment de chaque ion Fe^{+2} aux déformations du cristal. Les calculs sont faits pour FeCl_2 à 0 K dans les phases antiferromagnétique et paramagnétique saturée ; cette dernière phase est obtenue en appliquant un champ magnétique suffisamment fort selon l'axe d'alignement des moments. Nos résultats sont conformes à ceux que laissent prévoir les arguments de symétrie. L'ordre de grandeur prévu pour la variation d'aimantation est suffisant pour permettre sa mise en évidence expérimentale. De la mesure des composantes du tenseur, on peut déduire la valeur de certaines dérivées du champ cristallin en fonction des déformations.

Abstract. — We predict in the case of FeCl_2 which is an uniaxial antiferromagnet for $T \leq T_N$ (≈ 25 K), that one can modify the Fe^{+2} ions crystalline field symmetry by applying a mechanical stress and hence modify the moment orientation of these ions. We give as function of the crystalline field variation δW , an expression for the components of the tensor which relates the moment variation of each ion with the crystal strains. 0 K results are obtained for the antiferromagnetic and saturated paramagnetic phases, this latter phase being obtained by application of a strong enough magnetic field along the moment alignment direction. Our results are compatible with symmetry considerations. The size of the predicted moment variation suggests that it could be observed experimentally. From the tensor components measurement one can deduce the value of some of the crystalline field derivatives with respect to strains.

1. Introduction. — The magneto-elastic coupling associated with the crystalline field has been extensively studied in FeCl_2 . Some original dynamic [1-8] and static [9, 10 and 17] effects due to this coupling have been observed and described by different authors. In this article we predict the existence of some additional static effects due to this coupling.

In the paramagnetic phase ($T > T_N \approx 25$ K), the Fe^{+2} ions are located in crystallographic sites of group point symmetry $\bar{3}m$ which can be derived from each other by elementary translations of the Bravais rhomboedral lattice. Let us call $\{\text{Ox}_i, i = 1, 2, 3\}$ a set of rectangular coordinate axes, Ox_3 being parallel to the symmetry axis $\bar{3}$ and Ox_1 being contained in one of the mirror-planes of the $\bar{3}m$ point group. Below T_N , an uniaxial antiferromagnetic two-sublattice structure (A.F.) appears, the Ox_3 axis being the easy magnetization axis. On figure 1, this (A.F.) structure at 0 K is

Fig. 1. — Antiferromagnetic order of FeCl_2 at 0 K and $\mathbf{H} = 0$, with the $\{\text{Ox}_i\}$ set as used in the text.

shown together with the $\{\text{Ox}_i\}$ axes. With an applied magnetic field parallel to Ox_3 and exceeding a critical value H_s (≈ 10 kOe) the structure is that of a saturated

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paramagnetic (S.P.); i.e., all the moments have the direction of the applied field.

Let us call ε_{rs} ($r, s = 1, 2, 3$) the components relative to $\{\text{Ox}_i\}$ of the crystal homogeneous strain tensor. One must distinguish two types of strains : *a*) those which preserve the $\bar{3}m$ symmetry of the crystal (i.e. ε_{33} or ε_{11} and ε_{22} with $\varepsilon_{11} = \varepsilon_{22}$); *b*) those which do not (i.e. ε_{12} , ε_{13} , ε_{23} , or ε_{11} and ε_{22} with $\varepsilon_{11} \neq \varepsilon_{22}$). We show in this paper that when one creates, *via* external stresses, strains of the second type, the moment alignment direction is no longer preserved along Ox_3 in either the (A.F.) or the (S.P.) phases.

This result can be predicted by means of symmetry arguments. Although these arguments do not provide quantitative information about the phenomena to be expected, they remain essential for accepting (or eliminating) the mechanisms invoked as responsible for these phenomena.

In the case of FeCl_2 , the above mentioned moment reorientation, after inspection of symmetry compatibility, is attributed to the variation of the crystal field related to strains of the second type, for two conditions which turn out to be well verified : i) the corresponding variation of the dipolar interaction can be neglected and ii) the quenching of the Fe^{++} orbital moment is not perfect.

Let us call δW the operator associated with the variation of the crystal field related to the strains of the second type. δW contains as parameters some derivatives of the crystal field with respect to strains.

The effects described in [10] are, like those presented here, due to δW . But, whereas in [10] we showed that an applied field perpendicular to Ox_3 creates both strain and original magnetic effects, here we are concerned with the influence of mechanical stress. The experimental study of the influence of such a magnetic field or of some particular stresses would allow the parameters contained in δW to be evaluated. In previous experimental studies [2, 5 and 7] using dynamical effects, this evaluation was either indirectly or incompletely obtained.

In the first part below we study the symmetry of the tensor which relates the Fe^{++} ions magnetic moments components to those of the crystal homogeneous elastic strains. In the second part we describe the microscopical origin of this tensor and we give the expression of its components with respect to the parameters contained in δW . In this paper we neglect the dipolar interactions and the associated magneto-elastic coupling. Calculations are carried out for FeCl_2 at 0 K.

2. Symmetry arguments. — **2.1 (A.F.) PHASE IN ZERO APPLIED FIELD.** — In the (A.F.) phase, in a zero applied field, the point group of the Fe^{++} ions is no longer $\bar{3}m$ but $\bar{3}m$; the underlined mirror indicating that the spatial transformation must be followed by time-inversion. Let us call R the time-inversion opera-

tor. It is obvious that this operator interchanges the two magnetic sublattices.

Let us call $\delta m_i^{(\alpha)}$ the component relative to Ox_i of the magnetic moment variation induced by the applied stresses on a Fe^{+2} ion of the α sublattice ($\alpha = 1, 2$). To first order in ε_{rs} we have

$$\delta m_i^{(\alpha)} = \chi_{irs}^{(\alpha)} \varepsilon_{rs}. \quad (1)$$

The time-inversion operator changes the sign of the $\chi_{irs}^{(\alpha)}$ tensor. So we have the following results

$$R\chi_{irs}^{(\alpha)} = -\chi_{irs}^{(\alpha)} = \chi_{irs}^{(\beta)} \quad \forall i, r, s. \quad (2)$$

In these relations $\alpha = 1$ (resp. 2) and $\beta = 2$ (resp. 1).

From relations (1) and (2) we deduce the following relations (3) and (4) :

$$\delta m_i^{(1)} + \delta m_i^{(2)} = 0 \quad \forall i \quad (3)$$

$$\delta m_i^{(1)} - \delta m_i^{(2)} = 2 \chi_{irs}^{(1)} \varepsilon_{rs} = 2 \chi_{irs} \varepsilon_{rs} \quad (4)$$

where $\chi_{irs}^{(1)} = \chi_{irs}$.

Relations (3) and (4) mean that when we apply stresses the crystal keeps its antiferromagnetic structure but the alignment direction of the moments is no longer necessarily parallel to Ox_3 .

The χ_{irs} tensor is invariant under the $\bar{3}m$ point group symmetry operations. One can find in Birss' Book [11] the symmetry relations of the χ_{irs} . From Birss' results one can obtain :

$$\delta m_1^{(1)} = -\delta m_1^{(2)} = \chi_{111}(\varepsilon_{11} - \varepsilon_{22}) + 2 \chi_{113} \varepsilon_{13} \quad (5a)$$

$$\delta m_2^{(1)} = -\delta m_2^{(2)} = -2 \chi_{111} \varepsilon_{21} + 2 \chi_{113} \varepsilon_{23} \quad (5b)$$

$$\delta m_3^{(1)} = -\delta m_3^{(2)} = \chi_{333} \varepsilon_{33} + \chi_{311}(\varepsilon_{11} + \varepsilon_{22}). \quad (5c)$$

2.2 (S.P.) PHASE. — In this phase just one of the magnetic sublattices is left. The symmetry point group of each Fe^{+2} ion, the physical system including the crystal and the applied field, is again $\bar{3}m$; the symmetry operators $\bar{3}$ and $\underline{m}$ being the same as in the (A.F.) phase.

Let us call $\delta \tilde{m}_i$ the component relative to Ox_i of the magnetic moment variation induced by the applied stresses on a Fe^{+2} ion. To first order in ε_{rs} we have :

$$\delta \tilde{m}_i = \tilde{\chi}_{irs} \varepsilon_{rs}.$$

The $\tilde{\chi}_{irs}$ tensor having the same symmetry relations as the χ_{irs} tensor we have the following relations :

$$\delta \tilde{m}_1 = \tilde{\chi}_{111}(\varepsilon_{11} - \varepsilon_{22}) + 2 \tilde{\chi}_{113} \varepsilon_{13} \quad (6a)$$

$$\delta \tilde{m}_2 = -2 \tilde{\chi}_{111} \varepsilon_{21} + 2 \tilde{\chi}_{113} \varepsilon_{23} \quad (6b)$$

$$\delta \tilde{m}_3 = \tilde{\chi}_{333} \varepsilon_{33} + \tilde{\chi}_{311}(\varepsilon_{11} + \varepsilon_{22}). \quad (6c)$$

Relations (6a) and (6b) show that with some stresses the crystal magnetization is no longer directed along the applied field.

In FeCl₂ the quantities χ_{111} , χ_{113} , $\tilde{\chi}_{111}$ and $\tilde{\chi}_{113}$ are not zero because of the part of magneto-elastic coupling associated with δW . These quantities are related only to δW and not to the other observables, in a way to be described in section 3.

On the other hand the quantities χ_{333} , χ_{311} , $\tilde{\chi}_{333}$ and $\tilde{\chi}_{311}$ are not zero in FeCl₂ because of magneto-elastic coupling coming from exchange and from the crystalline field (except, for the crystalline field, for the part described by δW).

3. Microscopic study. — The microscopic model that we use has been introduced in [10]. We will refer here only to what is necessary for the description of our results.

The magneto-elastic coupling is considered as a perturbation of the rigid crystal model. Ôno *et al.* [14] were the first to propose satisfactory numerical values for the parameters of this model. Following their work, several authors have proposed modifications of these values [15, 16].

3.1 RIGID CRYSTAL MODEL : MAGNETO-ELASTIC COUPLING IS NEGLECTED. — We give some results obtained by Ôno *et al.* [14].

3.1.1 In the (A.F.) phase, in zero applied field, in a two-sublattice molecular field approximation, we get the following results at 0 K :

— The first three energy levels of a Fe²⁺ ion are the same for any of the magnetic sublattice to which the ion belongs. Let us call E_k , the energy value of the k level, with $k = \text{I, II, and III}$ ($E_{\text{I}} < E_{\text{II}} < E_{\text{III}}$). We neglect the energy values above E_{III} .

— The eigen kets associated to the energy values E_{I} , E_{II} and E_{III} are respectively $|1\rangle$, $|0\rangle$ and $|-1\rangle$ if the Fe²⁺ ions belong to the sublattice (1) and respectively $|-1\rangle$, $|0\rangle$ and $|1\rangle$ if they belong to the sublattice (2). These three kets are the eigen kets of s_3 the component relative to Ox_3 of a fictitious spin s ($s = 1$) defined for each Fe²⁺ ion. We give in Appendix I the expressions for these kets with respect to the kets $|L, S, M_L, M_S\rangle$, eigen kets of L^2 , S^2 , L_3 and S_3 ; L and S being the orbital and the spin moments of the free ion Fe²⁺ ($L = 2$ and $S = 2$), and L_3 and S_3 their components relative to Ox_3 .

— Let us call $x_i^{(\alpha)}$ ($\alpha = 1, 2$) the mean value in the (α) sublattice of s_i the component of s relative to Ox_i . We obviously have $x_1^{(\alpha)} = x_2^{(\alpha)} = 0$ and

$$x_3^{(1)} = -x_3^{(2)} = 1.$$

3.1.2 The results at 0 K in the (S.P.) phase, in presence of an applied field with modulus H ($H > H_s$) along Ox_3 , are the following :

— All the Fe²⁺ ions have the same lowest three

energy levels. Let us call $\tilde{E}_k$ the value of energy of the k level with $k = \text{I, II and III}$ ($\tilde{E}_{\text{I}} < \tilde{E}_{\text{II}} < \tilde{E}_{\text{III}}$). We neglect the values of energy above $\tilde{E}_{\text{III}}$.

— The eigen kets associated to the energy values $\tilde{E}_{\text{I}}$, $\tilde{E}_{\text{II}}$ and $\tilde{E}_{\text{III}}$ are respectively $|-1\rangle$, $|0\rangle$ and $|1\rangle$.

— Let us call $\tilde{x}_i$ the mean value of s_i . We have $\tilde{x}_1 = \tilde{x}_2 = 0$ and $\tilde{x}_3 = -1$.

We shall now express the quantities χ_{111} , χ_{113} , $\tilde{\chi}_{111}$ and $\tilde{\chi}_{113}$ as function of the parameters contained in the expression of δW . For this we shall consider δW to be a perturbation to Ôno's model.

The quantities χ_{333} , χ_{311} , $\tilde{\chi}_{333}$ and $\tilde{\chi}_{311}$ do not depend on δW . They depend on the variations of the Ôno model parameters with the strains which preserve the crystal symmetry. Appendix II gives expressions of these quantities as function of the variation of these parameters.

3.2 δW AS A PERTURBATION OF THE RIGID CRYSTAL MODEL. — The expression of δW with respect to L_i , the component of L relative to Ox_i , is invariant under the $\bar{3}m$ point group symmetry operations and can be written

$$\begin{aligned} \delta W = & \varepsilon_{13}(K_1 R + K'_1 U) + \varepsilon_{23}(K_1 S - K'_1 V) + \\ & + (\varepsilon_{11} - \varepsilon_{22})(K_2 U - K'_2 R) \\ & + 2\varepsilon_{12}(K_2 V + K'_2 S) \end{aligned}$$

with

$$\begin{aligned} R &= L_1 L_3 + L_3 L_1 & U &= L_1^2 - L_2^2 \\ S &= L_2 L_3 + L_3 L_2 & V &= L_1 L_2 + L_2 L_1. \end{aligned}$$

In this expression of δW — limited to second order in L_i — K_1 , K'_1 , K_2 and K'_2 are real constants which correspond to some partial derivatives of the crystalline field with respect to the crystal strains tensor.

We admit that the effect of δW is to perturb the results obtained in the rigid crystal model. One can show that to first order theory of perturbation the following results are obtained.

3.2.1 In the (A.F.) phase. — The expression of $(\delta E)_\alpha$, the perturbation of the fundamental energy level of one ion of the (α) sublattice, does not depend explicitly on ε_{13} , ε_{23} , $(\varepsilon_{11} - \varepsilon_{22})$ and ε_{12} . If we call $\delta x_i^{(\alpha)}$ the variation of the mean value of s_i induced by δW in the (α) sublattice, then $(\delta E)_\alpha$ depends only on $\delta x_3^{(1)}$ and $\delta x_3^{(2)}$.

— The perturbed fundamental state $|\varphi^{(\alpha)}\rangle$ of a Fe²⁺ ion of the (α) sublattice can be written :

$$|\varphi^{(\alpha)}\rangle = |m_\alpha\rangle + p_\alpha |0\rangle + q_\alpha |m_\beta\rangle$$

with $\alpha = 1, 2$ and $\beta = 2, 1$, $m_1 = 1$ and $m_2 = -1$.

From this, the self-consistent molecular field equations can be written :

$$x_i^{(\alpha)} + \delta x_i^{(\alpha)} = \langle \varphi^{(\alpha)} | s_i | \varphi^{(\alpha)} \rangle.$$

These equations can be solved by expressing $\delta x_i^{(\alpha)}$ as linear functions of the ε_{pq} , and we obtain the following results :

$$\delta x_i^{(1)} + \delta x_i^{(2)} = 0 \quad \forall i = 1, 2 \text{ and } 3 ;$$

and
$$\delta x_3^{(1)} = \delta x_3^{(2)} = 0$$

to first order in ε_{pq} . Then we can deduce the component $\delta m_i^{(\alpha)}$ relative to Ox_i of the variation of the mean value of the magnetic moment induced by δW in a Fe^{+2} ion of the (α) sublattice by using the relation

$$\delta m_i^{(\alpha)} = -g_i \mu_B \delta x_i^{(\alpha)}$$

with

$$g_1 = g_2 = g_{\perp}, \quad g_3 = g_{\parallel}$$

and μ_B being the Bohr magneton. In the Ôno model $g_{\perp} \approx 3.2$ and $g_{\parallel} \approx 4$.

By comparing the results obtained by this method with that contained in the relations (5a) and (5b) one can write :

$$\begin{aligned} \chi_{111} &= \mu_B \gamma (2\sqrt{2} K_2 - K_2') \\ \chi_{113} &= \frac{1}{2} \mu_B \gamma (K_1 + 2\sqrt{2} K_1') \end{aligned}$$

with
$$\gamma = g_{\perp} \frac{f}{D_{\parallel} - J_{\perp}^{-}},$$

$$D_{\parallel} = E_{\parallel} - E_1, \quad J_{\perp}^{-} = J_{1\perp} - J_{2\perp}$$

and $f = c'd - cd'$. The quantities c , c' , d and d' are defined in the Appendix I and $J_{1\perp} - J_{2\perp}$ correspond to exchange interactions and are defined in [10]. In the Ôno model $f = -0.107$, $D_{\parallel} = 33 \text{ cm}^{-1}$, $J_{\perp}^{-} = 14.53 \text{ cm}^{-1}$ and $\gamma = -2.3 \times 10^{-2} \text{ cm}^{-1}$.

3.2.2 In the (S.P.) phase. — The expression of $\delta \tilde{E}_1$, the perturbation of the fundamental energy level of one Fe^{+2} ion, does not depend explicitly on ε_{13} , ε_{23} , $(\varepsilon_{11} - \varepsilon_{22})$ and ε_{12} . If we call $\delta \tilde{x}_i$ the variation of the mean value of s_i induced by δW , $\delta \tilde{E}_1$ depends only on $\delta \tilde{x}_3$.

— The perturbed fundamental state $|\tilde{\varphi}\rangle$ of a Fe^{+2} ion can be written

$$|\tilde{\varphi}\rangle = |-1\rangle + \tilde{p}|0\rangle + \tilde{q}|1\rangle.$$

As previously described, we deduce $\delta \tilde{x}_i$ as linear function of the ε_{pq} from the molecular field equations. And from this we obtain $\delta \tilde{m}_i$ the component relative to Ox_i of the variation of mean value of the magnetic moment induced by δW in each Fe^{+2} ion. Then, by comparing this result with that of the equations (6a) and (6b) one can write

$$\begin{aligned} \tilde{\chi}_{111} &= -\mu_B \tilde{\gamma} (2\sqrt{2} K_2 - K_2') \\ \tilde{\chi}_{113} &= -\frac{1}{2} \mu_B \tilde{\gamma} (K_1 + 2\sqrt{2} K_1') \end{aligned}$$

with

$$\tilde{\gamma} = g_{\perp} \frac{f}{\tilde{D}_{\parallel} - J_{\perp}^{+}}, \quad \tilde{D}_{\parallel} = \tilde{E}_{\parallel} - \tilde{E}_1$$

and

$$J_{\perp}^{+} = J_{1\perp} + J_{2\perp}.$$

As we neglect dipolar interactions

$$\tilde{D}_{\parallel} = D_{\parallel} + g_{\parallel} \mu_B H.$$

In the Ôno model $J_{\perp}^{+} = 11.63 \text{ cm}^{-1}$, $\tilde{D}_{\parallel} = 35 \text{ cm}^{-1}$ for $H = 10 \text{ kOe}$ and $\tilde{\gamma} \approx -10^{-2} \text{ cm}^{-1}$.

3.3 STRESSES AND STRAINS. — One can obtain the values of $(\varepsilon_{11} - \varepsilon_{22})$, ε_{12} , ε_{13} and ε_{23} created by the applied stresses by minimizing with respect to the ε_{pq} the variational energy $\frac{1}{2} C_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \sigma_{pq} \varepsilon_{pq}$, where, in the $\{Ox_i\}$ frame, C_{ijkl} and σ_{pq} are respectively the components of elasticity tensor per unit volume and the components of the external stress tensor. This result is correct because first all the observables of the magneto-elastic coupling depending on these strains components are, in $FeCl_2$, included in δW and second the magneto-elastic energies $(\delta E_{\nu})_{\alpha}$ and $\delta \tilde{E}_1$, do not depend on these same components. However in the (S.P.) phase this result supposes that we have neglected the torques coming from the interaction of the applied magnetic field with the crystal magnetization variation, in comparison with the external stresses.

In each of the two phases, the symmetry relations between the components C_{ijkl} of the elasticity tensor are those of the paramagnetic phase ($T > T_N$); i.e. for the $3m$ point group. Only the numerical values of the non zero components can depend on the particular magnetic phase present.

4. Conclusion. — This study shows that knowledge of the strain (or stress) dependence of the magnetic moments would allow us to obtain the values of some derivatives of the crystalline field with respect to the strain tensor.

If we take the parameters K_1 , K_1' , K_2 , K_2' to have values of the order of $5 \times 10^3 \text{ cm}^{-1}$ [10] and [7], we see that strains near 2×10^{-3} would create a moment variation of near $10^{-1} \mu_B$ for each Fe^{+2} ion. However this expected value can be above $10^{-1} \mu_B$ if we use the values of Alben [15] or Carrara [16] for the rigid crystal model parameters; with the values of these authors γ and $\tilde{\gamma}$ will be higher than obtained using the values of Ôno *et al.* Such variation can be easily observed in the study of depolarization of a polarized neutron beam (see studies of V. A. Trunov *et al.* [17]). Although $FeCl_2$ crystal has a tendency to cleave according to reticular planes perpendicular to the $\bar{3}$ axis, we think that strains near 2×10^{-3} are still too small to cleave the crystal.

Finally we note that, in our model, $\mathbf{u}$, the relative microscopic displacement of an atom located, in the undeformed crystal, at position $\mathbf{R}_0$ is not necessarily equal to $\vec{\epsilon}\mathbf{R}_0$, where $\vec{\epsilon}$ is the macroscopic strain tensor. But this is not necessary to the coherence of the model.

It is sufficient for our model to be coherent that the microscopic relative displacements of the atoms be uniquely determined by the strain tensor. For i) the microscopic parameters K_1 , K_2 , K'_1 , and K'_2 are phenomenological and not calculated from the atomic positions, and ii) the Fe⁺² ions are located, in the undeformed crystal, in equivalent crystallographic sites. In the elastic strains case we can assume that the relative microscopic displacements of atoms are uniquely determined by the strain tensor.

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APPENDIX I

Expression of the kets $|\pm 1\rangle$ and $|0\rangle$ in the representation $\{|M_L\rangle \otimes |M_S\rangle\}$. — Let us call $|M_L\rangle$ the eigen states of L^2 and L_3 and $|M_S\rangle$ those of S^2 and S_3 . Let us call $|P_{\pm 1}\rangle$ and $|P_0\rangle$ the following linear functions of $|M_L\rangle$:

$$\begin{aligned} |P_1\rangle &= -\frac{1}{\sqrt{3}}[\sqrt{2}|-2\rangle + |1\rangle] \\ |P_0\rangle &= |0\rangle \\ |P_{-1}\rangle &= \frac{1}{\sqrt{3}}[\sqrt{2}|2\rangle - |-1\rangle]. \end{aligned}$$

In the representation $\{|P_i, M_S\rangle, i = \pm 1, 0\}$ the expressions of the kets $|\pm 1\rangle$ and $|0\rangle$ are

$$\begin{aligned} |\pm 1\rangle &= c|P_{\pm 1}, 0\rangle + d|P_0, \pm 1\rangle + e|P_{\mp 1}, \pm 2\rangle \\ |0\rangle &= c'|P_{\pm 1}, \mp 1\rangle + d'|P_0, 0\rangle \end{aligned}$$

with c, d, e, c', d' real constants.

In the Ôno model

$$\begin{aligned} c &= -0.274 & d &= 0.428 & e &= -0.861 \\ c' &= -0.595 & d' &= 0.540. \end{aligned}$$

APPENDIX II

Microscopic study of quantities χ_{333} , χ_{311} , $\tilde{\chi}_{333}$ and $\tilde{\chi}_{311}$. — By symmetry arguments, it can be shown that the spin hamiltonian of the crystal in presence of strains which preserve the crystal symmetry (ϵ_{33} and/or ϵ_{11} and ϵ_{22} , with $\epsilon_{11} - \epsilon_{22} = 0$) can be obtained by considering the Ôno model parameters as function of

the strains. More precisely, if one parameter p is equal to p_0 in the Ôno model, it must be replaced by $p_0 + dp$ in the spin hamiltonian of the deformed crystal with

$$dp = \frac{\partial p}{\partial \epsilon_{11}} \epsilon_{11} + \frac{\partial p}{\partial \epsilon_{22}} \epsilon_{22} + \frac{\partial p}{\partial \epsilon_{33}} \epsilon_{33}$$

with

$$\frac{\partial p}{\partial \epsilon_{11}} = \frac{\partial p}{\partial \epsilon_{22}}.$$

The hamiltonian so obtained commutes with the Ôno model hamiltonian. Also, the fundamental states of the Fe⁺² ions are still the kets $|\pm 1\rangle$ defined in Appendix I but by considering the quantities c, d and e to be functions of ϵ_{11} and $\epsilon_{11} + \epsilon_{22}$.

In the Ôno model the quantities c, d and e depend only on a , the ratio between the trigonal part of crystalline field (see Ôno's paper [14]) and the spin orbit coupling of the free Fe⁺² ion. This ratio varies with strain.

Let us call g one of the three quantities c, d or e , and g_0 the value of the quantity in the Ôno model. In presence of strains, g varies by δg with

$$\delta g = \frac{dg}{da} \delta a = \frac{dg}{da} \left(\frac{\partial a}{\partial \epsilon_{11}} \epsilon_{11} + \frac{\partial a}{\partial \epsilon_{22}} \epsilon_{22} + \frac{\partial a}{\partial \epsilon_{33}} \epsilon_{33} \right)$$

with

$$\frac{\partial a}{\partial \epsilon_{11}} = \frac{\partial a}{\partial \epsilon_{22}}.$$

We have calculated the values of the quantities c, d and e for the following values of a : 0, 1, 1.25, 1.5 [13]. The values of $\partial a / \partial \epsilon_{11}$ and $\partial a / \partial \epsilon_{33}$ can be obtained from experimental studies of Narath *et al.* [18], Vettier [12] and Nasser [13].

1. (A.F.) PHASE. — $\delta m_3^{(1)}$ is obtained by calculating the variation of the mean value, in the state $|1\rangle$, of $-\mu_B(L_3 + 2S_3)$, the component relative to Ox_3 of the magnetic moment operator of one Fe⁺² ion, when the quantities c, d and e vary by $\delta c, \delta d$ and δe . We obtain:

$$\chi_{311} = -\mu_B q \frac{\partial a}{\partial \epsilon_{11}}$$

$$\chi_{333} = -\mu_B q \frac{\partial a}{\partial \epsilon_{33}}$$

with

$$q = 10 e_0 \frac{\delta e}{\delta a} + 4 d_0 \frac{\delta d}{\delta a} - 2 c_0 \frac{\delta c}{\delta a}.$$

2. (S.P.) PHASE. — One can obtain $\delta \tilde{m}_3$ by calculating the variation of the mean value in the state $|-1\rangle$, of $-\mu_B(L_3 + 2S_3)$, when the quantities c, d and e vary by $\delta c, \delta d$ and δe . We obtain:

$$\tilde{\chi}_{311} = -\chi_{311}$$

$$\tilde{\chi}_{333} = -\chi_{333}.$$

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