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More on spin orientation in MnO at 0 K and zero applied field : theoretical predictions

R. Bidaux, R. Conte and J. A. Nasser

DPh-G/PSRM, CEN Saclay, B.P. N° 2, 91190 Gif sur Yvette, France

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Résumé. — A haute température ($T > T_N \sim 110$ K), MnO est un cristal c.f.c., le groupe ponctuel des ions Mn^{++} étant $m\bar{3}m$. A T_N ce composé présente un ordre antiferromagnétique dont certaines propriétés sont assez bien décrites au moyen des interactions d'échange et dipolaires et du couplage magnéto-élastique provenant des interactions d'échange. On peut ainsi montrer que : 1) les interactions d'échange créent une structure antiferromagnétique à deux sous-réseaux, 2) les interactions dipolaires imposent aux spins (les termes spectraux de Mn^{++} sont $L = 0$ et $S = \frac{5}{2}$) d'être orthogonaux aux directions $[111]$, et 3) que le couplage magnéto-élastique provenant de l'échange crée un tenseur de déformation de symétrie $3m$.

Dans cet article nous étudions le rôle à 0 K du couplage magnéto-élastique provenant des interactions dipolaires. Bien qu'il soit petit, ce couplage provoque des effets qualitativement remarquables. Ainsi lève-t-il partiellement la dégénérescence planaire d'orientation des spins. On montre l'existence de douze domaines antiferromagnétiques, la direction d'alignement des spins d'un domaine étant contenue dans le plan défini par les axes $[111]$ et $[100]$ de la structure c.f.c. et étant presque orthogonale à l'axe $[111]$. Par ailleurs, cette interaction crée dans chaque domaine un tenseur de déformation de symétrie inférieure à $3m$. Ces résultats sont en accord avec ceux que l'on déduit des études expérimentales des propriétés magnétiques de MnO en champ nul et sans contrainte mécanique. En présence d'une pression hydrostatique ou d'une contrainte uniaxiale suivant la direction $[111]$, les résultats que nous obtenons ne sont pas qualitativement différents des précédents. En revanche, notre étude laisse prévoir la possibilité de favoriser un seul des douze domaines antiferromagnétiques en appliquant une contrainte uniaxiale ayant des composantes non nulles sur les deux axes $[111]$ et $[100]$ de la maille c.f.c. Un tel résultat serait très intéressant pour l'étude des propriétés magnétiques de MnO.

Abstract. — Above its Néel temperature T_N (~ 110 K), MnO has the f.c.c. crystalline structure, the point group of Mn^{++} ions being $m\bar{3}m$. At T_N an antiferromagnetic order takes place. Some properties of that order are well enough described by taking into account the exchange and dipolar interactions and the magneto-elastic coupling from exchange interactions. So : 1) exchange interactions are responsible for the two-sublattice antiferromagnetic structure, 2) dipolar interactions set the spins (for Mn^{++} $L = 0$ and $S = \frac{5}{2}$) parallel to the (111) planes of the f.c.c. structure and 3) magneto-elastic coupling due to exchange creates a $3m$ symmetry strain tensor.

In this article we study the effects of the magneto-elastic coupling coming from dipolar interactions. Although they are small, these effects are qualitatively important. So, the dipolar magneto-elastic coupling removes partially the spin orientation degeneracy. We show there are twelve antiferromagnetic domains in MnO, the spin direction of a domain being contained in the plane defined by the $[111]$ and $[100]$ axes of the f.c.c. structure and being nearly orthogonal to the $[111]$ axis. Furthermore, this interaction creates in each domain a strain tensor of symmetry lower than $3m$. All the results agree with those one can deduce from the experimental studies of MnO in a null magnetic field and null stress tensor.

The above results are not qualitatively changed by application of a hydrostatic pressure or an uniaxial stress along the $[111]$ axis. But we show that one could favour one among the twelve antiferromagnetic domains by applying a stress with components on both the $[111]$ and $[100]$ axes. Such a result would be very interesting for the magnetic study of MnO.

1. **Introduction.** — One may describe the onset of a magnetic order in a crystalline compound in the following way :

1) Magnetic order is supposed to take place in presence of a well defined regular lattice, with nuclei fixed in their mean position.

2) Since magnetic interactions depend on relative positions of nuclei, a coupling is induced between the preexisting lattice structure and the magnetic order above described ; therefore both original structures can be perturbed.

Thus, to each type of magnetic interaction, one

associates a Hamiltonian which can be considered as the sum of $\mathcal{H}_m + \mathcal{H}_{me}$; $\mathcal{H}_m$ is purely magnetic and defined in terms of 1); $\mathcal{H}_{me}$ describes the magneto-elastic coupling and will be dealt with as a perturbation of $\mathcal{H}_m$. In the case of a ferromagnetic or antiferromagnetic order, $\mathcal{H}_m$ and $\mathcal{H}_{me}$ can be written respectively $A\mathcal{O}$ and $A'\varepsilon\mathcal{O}'$; $\mathcal{O}$ and $\mathcal{O}'$ are operators defined in the field of the eigenstates of $\mathcal{H}_m$; ε is the tensor of homogeneous strains of the crystal; A and A' are tensors which characterize the respective magnitude of $\mathcal{H}_m$ and $\mathcal{H}_{me}$. $\mathcal{H}_{me}$ is obtained by expanding magnetic interactions to first order as a function of nuclei displacements from their mean positions, as defined in the original crystal; these displacements in turn are expressed as linear functions of the strain tensor.

When $A\mathcal{O}$ and $A'\varepsilon\mathcal{O}'$ commute, $\mathcal{H}_{me}$ does not perturb the eigenstates of $\mathcal{H}_m$, and therefore the magnetic structure will be that ruled by $\mathcal{H}_m$, although the strains are not null. Such is the case, for example, when magnetic interactions are limited to isotropic exchange. On the other hand, when $A\mathcal{O}$ and $A'\varepsilon\mathcal{O}'$ do not commute, the effect of $\mathcal{H}_{me}$ results in both crystal strains and a possible modification of the magnetic structure. Thus, this modification requires as a necessary condition the non commutation of $A\mathcal{O}$ and $A'\varepsilon\mathcal{O}'$. This situation is met with a crystalline field or dipolar interactions.

For example, it has been recently showed that in FeCl_2 — a uniaxial antiferromagnetic compound — the behaviours of the two sublattices in presence of an applied field orthogonal to the anisotropy axis [1] are made inequivalent by the magneto-elastic coupling due to the crystalline field. We should want to show here that the magnetoelastic coupling due to dipolar interactions partially removes the spin orientation degeneracy in MnO .

MnO is an insulator compound which exhibits a magnetically ordered phase at low temperature ($T < T_N \simeq 110$ K). In the paramagnetic phase, Mn^{++} ions belong to a f.c.c. lattice and the point group of their crystallographic locations is $m\bar{3}m$.

From the neutron diffraction studies of Shull

et al. [2], Roth [3, 4] proposes for MnO the following antiferromagnetic collinear structure (type II structure), represented on figure 1 : the moments of the ions Mn^{++} which belong to a same plane (111) are equivalent, thus defining a ferromagnetic sheet. Moments of two adjacent ferromagnetic sheets are opposite. Moreover, Roth assumes the moments to be parallel to the sheets.

Such a structure can be explained by antiferromagnetic isotropic exchange interactions between first neighbours and between second neighbours. Nevertheless, since these interactions do not yield the absolute direction of spins and since the fundamental state of the free Mn^{++} ion has for spectral terms $L = 0$ and $S = \frac{5}{2}$, we are induced to explain the observed planar anisotropy by dipolar interactions [5].

Finally, several works [6, 7] show that the directions of easy spin alignment do not generate a plane but are discrete directions associated with a very weak anisotropy energy. But neither these directions nor the origin of that anisotropy are yet known [3, 8-15].

In this article, we show that the magneto-elastic coupling due to the dipolar interactions partially removes the degeneracy of planar orientations which remains when only the previously mentioned interactions are taken into account. We thus prove the appearance of easy spin alignment directions which are found to be very slightly outside (111) planes. We also show how the spin alignment directions can be modified under certain mechanical stresses.

In the second part, we present the interactions taken into account and the method we use. In the third part we give the results at 0 K in absence of stresses. In the last part we give the results at 0 K when different mechanical stresses are applied.

2. Interactions and variational energy of the system.

— Our hypotheses are the following :

- 1) the magnetic interactions between Mn^{++} ions are the isotropic antiferromagnetic exchanges between first neighbours and between second neighbours;
- 2) the magnetic structure below T_N is the antiferromagnetic structure with two sublattices α and β which is described in the introduction and represented on figure 1. What we are looking for is the spin alignment direction.

Let us call $\mathbf{S}^i$ the spin of the Mn^{++} ion located at site i , S_j^i , $j = 1, 2, 3$, its components on the axes $\mathbf{Ox}_j$, represented on figure 1, which are parallel to the cube edges in the f.c.c. cell, and ε_{lm} , $l, m = 1, 2, 3$ the components in those axes of the crystal homogeneous strain tensor ε .

The hamiltonian $\mathcal{H}_s$ of the system of spins is :

$$\mathcal{H}_s = \mathcal{H}_m + \mathcal{H}_{me}$$

Fig. 1. — Antiferromagnetic order of MnO at 0 K and $H = 0$; ● ions of α sublattice, ○ ions of β sublattice.

with

$$\begin{aligned}\mathcal{H}_m &= \sum_{\langle i,j \rangle} [-2 J^{ij} \mathbf{S}^i \cdot \mathbf{S}^j - 2 \tilde{J}^{ij} \mathbf{S}^i \mathbf{S}^j + (g\mu_B)^2 S_p^i T_{pq}^{ij} S_q^j] \\ \mathcal{H}_{me} &= \sum_{\substack{\langle i,j \rangle \\ p,q,l,m=1,2,3}} \varepsilon_{lm} [(E_{lm}^{ij} + \tilde{E}_{lm}^{ij}) \mathbf{S}^i \cdot \mathbf{S}^j + (g\mu_B)^2 D_{lm,pq}^{ij} S_p^i S_q^j].\end{aligned}$$

In these expressions g equals 2, μ_B is the Bohr magneton and the summation is extended over all different couples of Mn^{++} ions located at sites i and j . Tensors $-2 J^{ij}$ and $-2 \tilde{J}^{ij}$ correspond to the isotropic antiferromagnetic exchange between two Mn^{++} ions which are respectively first (there are 12 such ions) and second (they are $\tilde{n} = 6$) neighbours, T_{pq}^{ij} represents the dipolar interactions, E_{lm}^{ij} and $\tilde{E}_{lm}^{ij}$ correspond to the magneto-elastic coupling coming from the exchange interactions between two Mn^{++} ions respectively first and second neighbours, and $D_{lm,pq}^{ij}$ represents the magneto-elastic coupling due to dipolar interactions.

Let us call S_u the projection of a spin on unit vector $\mathbf{u}$ and $|M_S, \mathbf{u}\rangle$ its eigenvectors ($M_S = -\frac{5}{2}, \dots, \frac{5}{2}$). We call S_p the projection of the same spin on $\mathbf{OX}_p$ and we define

$$X_p = \langle M_S, \mathbf{u} | S_p | M_S, \mathbf{u} \rangle = - \langle M_S, -\mathbf{u} | S_p | M_S, -\mathbf{u} \rangle \quad (1)$$

with $X_p X_p = \frac{25}{4}$ for $M_S = \pm \frac{5}{2}$.

In order to find the fundamental state of $\mathcal{H}_s$ we choose a trial function such that the Mn^{++} ions belonging to sublattice α be in the state $|\frac{5}{2}, \mathbf{u}\rangle$ and those belonging to sublattice β in the state

$$|\frac{5}{2}, -\mathbf{u}\rangle = |-\frac{5}{2}, \mathbf{u}\rangle.$$

With that trial function the mean value E_s of $\mathcal{H}_s$ per unit volume is :

$$\begin{aligned}E_s(X, \varepsilon) &= \frac{N}{2} X_p X_q \{ [2 \tilde{J}^{ij} + \varepsilon_{lm} (E_{lm} + \tilde{E}_{lm})] \delta_{pq} + \\ &+ (g\mu_B)^2 (T_{pq} + \varepsilon_{lm} D_{lm,pq}) \}. \quad (2)\end{aligned}$$

In this expression N is the number of Mn^{++} ions per unit volume and δ_{pq} the Kronecker tensor. Tensors appearing in E_s describe the interaction between a Mn^{++} ion and all other Mn^{++} ions. These tensors have at least the symmetry 3m of the magnetic sublattices, the ternary axis being the principal diagonal of the cube of the f.c.c. cell, and the mirrors being the planes defined by that ternary axis and the three cube edges.

In (2), the three tensors $E_{lm} \delta_{pq}$, $\tilde{E}_{lm} \delta_{pq}$ and $D_{lm,pq}$ have at least the symmetries of a tensor τ_{ijkl} of symmetry 3m separately symmetrical with respect to the first two indices and the last two indices; in the axes $\mathbf{OX}_j$, it has eight different components :

$$\tau_{ijkl} = \begin{cases} \tau_{11} & i=j, k=l & j=k \\ \tau_{12} & i=j, k=l & j \neq k \\ \tau_{14} & i=j, k \neq l & i+l+k=6 \\ \tau_{15} & i=j, k \neq l & i+l+k \neq 6 \\ \tau_{41} & i \neq j, k=l & i+j+k=6 \\ \tau_{51} & i \neq j, k=l & i+j+k \neq 6 \\ \tau_{44} & i \neq j, k \neq l & i+j=k+l \\ \tau_{45} & i \neq j, k \neq l & i+j \neq k+l. \end{cases}$$

In the case of a symmetrical two-index tensor with 3m symmetry, its components τ_{ij} on axes $\mathbf{OX}_p$ are in number 2 :

$$\tau_{ij} = \begin{cases} \tau_{11} & i=j \\ \tau_{12} & i \neq j. \end{cases}$$

Using their definition and symmetry arguments, we can write down the components on axes $\mathbf{OX}_j$ of tensors appearing in (2) :

- $2 \tilde{J}^{ij}$, the exchange interaction between a Mn^{++} ion and its $\tilde{n}$ second neighbours, is a scalar ;
- T_{pq} , the dipolar interaction, is $T(1 - \delta_{pq})$;
- E_{lm} and $\tilde{E}_{lm}$, the magneto-elastic coupling coming from the exchange interactions between respectively first and second neighbours, can also be written

$$E_{lm} = E(1 - \delta_{lm}), \quad \tilde{E}_{lm} = \tilde{E} \delta_{lm}.$$

- $D_{lm,pq}$, the magneto-elastic coupling coming from dipolar interactions, has the symmetries of the above described tensor τ_{ijkl} ; moreover since it is obtained by derivation of a dipolar interaction tensor, its components verify the following supplementary relations :

$$\begin{aligned}D_{11} + 2 D_{12} &= 0, \quad D_{41} + 2 D_{51} = 0, \\ D_{14} + D_{41} &= 2 D_{45}, \quad D_{12} = D_{44}.\end{aligned}$$

The total energy E_t of the crystal per unit volume is the sum of the spin energy E_s and the elastic energy :

$$E_t(X, \varepsilon) = E_s(X, \varepsilon) + \frac{1}{2} c_{lm,pq} \varepsilon_{lm} \varepsilon_{pq}. \quad (3)$$

The elastic stiffness tensor $c_{lm,pq}$ can be split into a non-magnetic contribution and a magnetic contribution. The non-magnetic contribution has the symmetry m3m of the paramagnetic phase. The magnetic contribution comes from the second derivatives of the crystal magnetic interactions with respect to

the deformations; it is non-zero in the magnetically ordered phase and has the 3m symmetry of the magnetic sublattices. Therefore, the elasticity tensor has the 3m symmetry at 0 K.

In order to find the spin orientation and the crystal lattice deformations at equilibrium at 0 K, we must minimize E_s with respect to the X_p 's under the constraint (1) and then the total energy with respect to the deformation tensor ε_{lm} . These two operations may be done in any order.

The minimization of $E_t(X, \varepsilon)$ with respect to the ε_{lm} 's yields

$$\varepsilon_{pq} = s_{pqlm} \pi_{lm} \quad (4)$$

with

$$\pi_{lm} = -\frac{N}{2} [(E_{lm} + \tilde{E}_{lm}) \delta_{uv} + (g\mu_B)^2 D_{lmuv}] X_u X_v \quad (4a)$$

and

$$s_{ijkl} c_{klmn} = c_{ijkl} s_{klmn} = \frac{1}{2} (\delta_{im} \delta_{jn} + \delta_{in} \delta_{jm}) \quad (4b)$$

The symmetries of tensors s_{ijkl} and c_{ijkl} are the same.

After substituting in $E_t(X, \varepsilon)$ the ε_{lm} 's by their expression (4) depending on the X_p 's, we obtain :

$$E_t(X) = \frac{N}{2} \left[2 \tilde{J}^{ij} \delta_{pq} + (g\mu_B)^2 T_{pq} - \frac{1}{2} \frac{25}{4} \frac{N}{2} Q \delta_{pq} \right] X_p X_q - \left(\frac{N}{2} \right)^2 (g\mu_B)^2 [A_{pq} \delta_{uv} + \frac{1}{2} (g\mu_B)^2 B_{pquv}] X_p X_q X_u X_v \quad (5)$$

with

$$\begin{aligned} Q &= (E_{lm} + \tilde{E}_{lm}) s_{lm pq} (E_{pq} + \tilde{E}_{pq}) \\ A_{pq} &= (E_{lm} + \tilde{E}_{lm}) s_{lm uv} D_{uv pq} \\ B_{pquv} &= D_{lm pq} s_{lm nr} D_{nr uv} \end{aligned}$$

Tensors A_{pq} and B_{pquv} have at least the 3m symmetry. A_{pq} is symmetric and B_{pquv} has the symmetries of an elasticity tensor (invariance under the exchange of p, q , of u, v and of $(pq), (uv)$).

When minimizing $E_t(X)$ under constraint (1), we obtain the spins orientation at equilibrium at 0 K. Using relation (4), we deduce the equilibrium deformations at 0 K. This minimization is easier to perform if we use symmetry adapted variables.

Let $Oy_1 y_2 y_3$ be the direct frame of reference, represented on figure 1, such that its axes have respectively the orientations $[1 - 1 0]$, $[1 1 - 2]$, $[111]$. We define φ and w as the cylindrical coordinates of the unit vector $\mathbf{u} = \frac{2}{3} \mathbf{M}$, where $\mathbf{M}$ is a spin of sublattice α at equilibrium : w is the projection of $\mathbf{u}$ on Oy_3 and φ is the angle between Oy_1 and the projection of $\mathbf{u}$ on the plane $Oy_1 y_2$. In these new variables, the total energy simply becomes :

$$E_t(\varphi, w) = E_0 + E_2 w^2 + E_4 w^4 + E_3 w(1 - w^2)^{3/2} \sin 3\varphi \quad (6)$$

with $-1 \leq w \leq 1$.

This expression is clearly seen to be invariant under the point group 3m. Energies E_2, E_4, E_3 are defined by

$$E_2 = K \left[3 T - \frac{N}{2} \frac{25}{4} 3 A_{12} - \frac{K}{2} (-a - 2b + 6 B_{1113}) \right]$$

$$E_4 = -\frac{1}{12} K^2 (7a + 20b)$$

$$E_3 = -\frac{\sqrt{2}}{3} K^2 (-a + b)$$

with the notations

$$K = \frac{N}{2} \frac{25}{4} (g\mu_B)^2$$

$$a = B_{1122} - B_{1111} + 2 B_{2323} = B_{12} - B_{11} + 2 B_{44}$$

$$b = B_{1123} - B_{1113} + 2 B_{2313} = B_{14} - B_{15} + 2 B_{45}$$

The extrema of E_t are to be found among the edges $w = \pm 1$ of the w domain and the solutions of the variational equations in the interval $-1 < w < 1$:

$$\frac{\partial E_t}{\partial \varphi} = 3 E_3 w(1 - w^2)^{3/2} \cos 3\varphi = 0 \quad (7a)$$

$$\begin{aligned} \frac{\partial E_t}{\partial w} &= 2 E_2 w + 4 E_4 w^3 + \\ &+ E_3 (1 - w^2)^{1/2} (1 - 4 w^2) \sin 3\varphi = 0 \end{aligned} \quad (7b)$$

3. Results. — **3.1 SPIN ORIENTATION.** — If we neglect the magneto-elastic coupling coming from dipolar interactions, the total energy depends no longer on φ :

$$E_t' = E_0' + E_2' w^2.$$

In this expression E_0' and E_2' are constants, $E_2' = 3 KT$. If we assume T positive, E_t' is minimum for $w = 0$, i.e. when spins are parallel to the crystallographic plane (111).

When we do not neglect the magneto-elastic coupling, the extrema of E_t are given by :

- $w = \pm 1$, $E_t = E_0 + E_2 + E_4$
- $w = 0$, $\sin 3\varphi = 0$, $E_t = E_0$
- $\cos 3\varphi = 0$, $w = \pm w_0$, $\sin 3\varphi = \pm 1$

$$E_t = E_0 - \frac{w_0^2}{1 - 4w_0^2} [E_2 + (2E_2 + 3E_4)w_0^2],$$

where w_0^2 is a root of a third degree equation.

If we consider $\mathcal{K}_{me}$ as a perturbation to $\mathcal{K}_m$, the dominant term in E_2 is E'_2 and E_2 is therefore positive. It is reasonable to admit that $|E_4|$ and $|E_3|$ are very small compared with E_2 . Under these conditions, the solution with the lowest energy is :

$$\cos 3\varphi = 0, \quad w = \pm w_0, \quad \sin 3\varphi = \mp 1,$$

$$w_0 \simeq \frac{E_3}{2E_2}, \quad E_t \simeq E_0 - \frac{1}{4} \frac{E_3^2}{E_2}.$$

There seems to be six independent orientations of the sublattice α spins but, since the orientations of the spins of the two magnetic sublattices are opposite and cannot be distinguished from one another, there remain only three spin alignment directions at equilibrium at 0 K. These directions are parallel to the three mirrors of the point group 3m, i.e. those defined by the ternary axis [111] and the three edges [100] of the cube of structure f.c.c. and their angle with the plane (111) is $|w_0| \ll 1$. Thus these directions are roughly parallel to the $[11\bar{2}]$ directions. In a previous article [16] we had considered only the solutions $w = 0$, $\sin 3\varphi = 0$, because we were looking for solutions located only in the plane (111).

Thus, from our model, to a given ternary axis of the f.c.c. structure are associated three spin alignment directions, i.e. three antiferromagnetic domains, and two directions differ by a rotation of $2\pi/3$ around the ternary axis. Since there are four equivalent ternary axes, our model predicts in a MnO crystal the presence of twelve different antiferromagnetic domains.

3.2 ANISOTROPY ENERGIES. — Several authors have defined macroscopic anisotropy energies, which we shall now try and recover in our model.

3.2.1 Anisotropy with respect to the (111) plane. — Kaplan [5] and Keffer and O'Sullivan [6] showed that the macroscopic dipolar energy of a cubic crystal may be written, in the MnO case, as $K_1 \cos^2 \theta$, θ being the angle of the ternary axis [111] with the spin direction. This can be recovered using relation (6) if we define $w = \cos \theta$ and assume $E_4 = E_3 = 0$. The latter relation means that we neglect the magneto-elastic coupling coming from dipolar interactions. We then have $K_1 = 3KT$.

3.2.2 Anisotropy around the ternary axis. — By assuming the spins to be in the (111) plane, several authors [6, 7] propose an expression of the type $-\frac{1}{2} K_2 \cos 6\eta$ for the anisotropy energy relative

to the rotation around the [111] directions, where η is the angle between the spins direction and the spins easy alignment direction. Yamada [7] shows that, according to the sign of K_2 , the spins easy alignment direction in the plane is either a $[1\bar{1}0]$ type direction if K_2 is negative, or a $[11\bar{2}]$ type direction otherwise.

We can retrieve in our model a similar expression by partially minimizing $E_t(\varphi, w)$ with respect to w . Solving equation (7b) after linearization in w and substituting that solution in (6), we get, if we keep only the w^2 term :

$$\tilde{E}_t(\varphi) \simeq E_0 - \frac{E_3^2}{4E_2} \sin^2 3\varphi.$$

If we define

$$\eta = \varphi - \frac{\pi}{6} \quad \text{and} \quad K_2 = \frac{E_3^2}{8E_2} > 0,$$

we retrieve the results of Yamada provided we consider η as the angle between the projections on the (111) plane of the spins direction and the spins easy alignment direction.

3.3 DEFORMATIONS AT EQUILIBRIUM. — Using relations (4) and (4a) we can decompose the crystal deformation ε_{pq} into two parts :

$$\varepsilon_{pq} = \varepsilon_{pq}^e + \varepsilon_{pq}^d \quad (8)$$

$$\varepsilon_{pq}^e = -\frac{N}{2} s_{pqlm} (E_{lm} + \tilde{E}_{lm}) \delta_{uv} X_u X_v \quad (8a)$$

$$\varepsilon_{pq}^d = -\frac{N}{2} (g\mu_B)^2 s_{pqlm} D_{lmuv} X_u X_v \quad (8b)$$

where ε_{pq}^e and ε_{pq}^d are the deformations respectively created by the exchange interactions and the dipolar interactions.

3.3.1 Deformations due to the exchange interactions. — These deformations do not depend on the spins orientations at equilibrium and they have the 3m symmetry :

$$\varepsilon_{ij}^e = \varepsilon_{11}^e \quad \text{for } i = j \quad \text{and} \quad \varepsilon_{12}^e \quad \text{for } i \neq j.$$

3.3.2 Deformations coming from dipolar interactions. — These deformations depend on the spins orientation at equilibrium and therefore on the antiferromagnetic domain under consideration. We shall study in more detail the domain defined by a spin orientation $\varphi = \pi/2$, $w = w_0$.

The tensor $M_{pquv} = s_{pqlm} D_{lmuv}$ is invariant under the permutation of indices p, q and the permutation of indices u, v , and it has the 3m symmetry. Moreover, the relations between the D_{lmuv} 's imply :

$$2M_{1122} + M_{1111} = 2M_{1311} + M_{2311} = 0.$$

The antiferromagnetic domain under consideration is such that the spins are contained in the plane

defined by the ternary axis and the edge $\mathbf{OX}_1$ of the cube. It follows that $X_2 = X_3$. Using the latter relation, relation (1) and the symmetries of $M_{pq\mu}$, we can show that

$$\varepsilon_{22}^d = \varepsilon_{33}^d \quad \text{and} \quad \varepsilon_{12}^d = \varepsilon_{13}^d.$$

Thus, deformation coming from dipolar interactions depend on spins orientation at equilibrium and, for a given antiferromagnetic domain, have monoclinic symmetry and then are not invariant under the 3m group. However, the sum of the deformation tensors associated with the three antiferromagnetic domains which are deduced from one of them by a rotation of $2\pi/3$ around the ternary axis [111] does have the 3m symmetry.

3.4 DISCUSSION OF THE RESULTS. — In order to estimate the numerical values of the effects predicted by our model (deformations and spins orientation), we must know the values of the elastic stiffness coefficients for MnO at 0 K, the value of the dipolar interaction tensor elements and that of the different magneto-elastic coupling parameters.

We have computed by the method of Bidaux *et al.* [17] the non zero components of T_{pq} and $D_{lm\mu}$. We thus have found :

$$\begin{aligned} a_0^3 T &= 14.46, \quad a_0^3 D_{11} = -11.9, \quad a_0^3 D_{14} = 5.56, \\ a_0^3 D_{15} &= -24.48, \quad a_0^3 D_{41} = 20.04 \end{aligned}$$

where a_0 is the value that the cube's edge would have at 0 K in the absence of the magneto-elastic coupling. A good approximation for a_0 is the length of the edge just before the magnetic transition, i.e. 4.437 Å according to the expansion measurements of Morosin [18]. We deduce that the energies $(g\mu_B)^2 T$, $(g\mu_B)^2 D_{11}$, $(g\mu_B)^2 D_{14}$, $(g\mu_B)^2 D_{15}$ and $(g\mu_B)^2 D_{41}$ evaluate respectively to 0.287, -0.236, 0.11, -0.485 and 0.397 cm⁻¹, with $g = 2$.

Computations cannot yield E and $\tilde{E}$, the magneto-elastic coupling parameters due to exchange interactions. Moreover we do not know the value at 0 K of MnO elastic coefficients, since these quantities have only been measured in the paramagnetic cubic phase [19] to [22].

Given the available experimental results, the value at 298 K of the elastic stiffness coefficients measured by Oliver [20] and the value of the deformation tensor for MnO between T_N and 4 K measured by Morosin [18] will enable us to deduce the value of E and $\tilde{E}$ and the spins orientation at 0 K. We chose the Oliver values in order to be able to compare our results with those of Pépy [23]. These values are for c_{11} , c_{12} and c_{14} respectively 2.23 ± 0.06 , 1.2 ± 0.1 and $0.79 \pm 0.02 \times 10^{11}$ N/m².

For the antiferromagnetic domain defined by $\varphi = \pi/2$ and $w = w_0$ we have computed the value at 0 K of the deformations ε_{ij}^d due to dipolar interactions. For so doing we have taken $X_2 = X_3$,

$X_1 \simeq -2X_2$, $N = 4.58 \times 10^{28}$ atoms/m³. We have found for ε_{11}^d , ε_{22}^d , ε_{13}^d and ε_{23}^d respectively -9.4×10^{-6} , 2.4×10^{-6} , 1.2×10^{-5} and 3.5×10^{-6} . Since the deformations measured by Morosin are

$$\varepsilon_{ij}^{\text{exp}} = -1.2 \times 10^{-3}$$

for $i = j$ and -5.5×10^{-3} for $i \neq j$, we conclude that deformations of crystalline structure of MnO which are observed between T_N and 4 K cannot be attributed to a magneto-elastic coupling due to dipolar interactions. If we attribute them to the coupling due to exchange interactions, we find for the energies E and $\tilde{E}$ values respectively next to 306 cm⁻¹ and 196 cm⁻¹. We must point out that in these computations we supposed $|w_0| \ll 1$ and we neglected elastic stiffness coefficients c_{14} , c_{15} and c_{45} which are no more necessarily zero in the antiferromagnetic phase.

Pépy [23] measured the spin waves dispersion curves in MnO at different temperatures by neutron spectroscopy. To explain his results he defines a model involving the dipolar interactions, two exchange integrals J_1 and J_2 between respectively first and second neighbours and two parameters ε_1 , ε_2 describing the variations of these exchange integrals with the distance. He considers the crystal deformations between T_N and 4 K as coming only from magneto-elastic coupling of exchange origin and neglects the variations of dipolar interactions with deformations. Comparing our model with that of Pépy, we establish that :

$$\begin{aligned} -2J^{ij} &= J_1, \quad E \equiv -2(r^{ij})_0 \frac{\partial}{\partial r^{ij}} (-2J^{ij}) = 4\varepsilon_1 J_1 \\ -2\tilde{J}^{ij} &= J_2, \quad \tilde{E} \equiv -2(\tilde{r}^{ij})_0 \frac{\partial}{\partial \tilde{r}^{ij}} (-2\tilde{J}^{ij}) = 4\varepsilon_2 J_2. \end{aligned}$$

To prove these relations we used the fact that J^{ij} and $\tilde{J}^{ij}$ depend only on the distance between ions, $(r^{ij})_0$ and $(\tilde{r}^{ij})_0$ being the distance between two Mn⁺⁺ ions respectively first and second neighbours in a f.c.c. lattice whose edge is a_0 . Using the values of J_1 , J_2 , ε_1 , ε_2 proposed by Pépy, we find for the energies E and $\tilde{E}$ respectively 305 and 187 cm⁻¹. These values agree well with those obtained in our model.

To find the value of the anisotropy constants K_1 and K_2 and that of the angle w_0 , we must compute the values of the macroscopic energies E_2 and E_3 .

The expression for E_2 is made of three terms : the first one, $3KT$, corresponds to a dipolar interaction energy computed for one antiferromagnetic domain, assuming the crystalline structure is f.c.c. with an edge a_0 ; the second term corresponds to the variation of that energy with the deformations ε_{ij}^e due to exchange; the third term, like E_4 and E_3 , depends only on the variation of dipolar interactions

with the deformations ε_{ij}^d . Given the values of tensors ε_{ij}^e and ε_{ij}^d , one can predict that the second term in E_2 is roughly $10^{-3} KT$ and the third term of E_2 roughly $10^{-6} KT$. We separately computed the three terms of E_2 and found respectively 2.44×10^6 , -5.0×10^4 and -14.2 J/m^3 , which yields

$$E_2 \simeq 2.39 \times 10^6 \text{ J/m}^3 \simeq 3 \text{ KT}.$$

For E_4 and E_3 we found respectively -26 and 6.5 J/m^3 .

These results show that the hypotheses of our model, i.e. $T > 0$, $E_2 \gg |E_3|$ and $E_2 \gg |E_4|$, are well verified. w_0 is found to be equal to 1.4×10^{-6} and the anisotropy constants K_1 and K_2 to 2.44×10^6 and $2.2 \times 10^{-6} \text{ J/m}^3$. Our value of K_1 is in excellent agreement with that proposed in references [5] and [6], i.e. $1.64 \times \frac{3}{2} \times 10^6 \text{ J/m}^3$.

The numerical values of w_0 , ε_{ij}^d and K_2 lead us to conclude that the magneto-elastic coupling of dipolar origin : 1) removes the planar orientation degeneracy by orienting the spins along directions contained in the planes defined by the cube edges and the ternary axis; the spins which make a very small angle with the (111) plane are roughly parallel to the $[11\bar{2}]$ directions; 2) creates deformations of order 10^{-6} , described by a tensor which for a given antiferromagnetic domain have the monoclinic symmetry, although the sum of the deformation tensors corresponding to the three domains associated with a given ternary axis does possess the 3m symmetry and therefore cannot be qualitatively distinguished from the deformations of exchange origin; 3) implies the existence of a very small anisotropy of rotation of the spins around the ternary axis.

These results are in agreement with those deduced by Pépy [23] from the experimental study of magnons in MnO, i.e. : 1) there exist in the (111) plane three easy alignment directions of spins which are deduced from one of them by a rotation of $2\pi/3$ around every $[111]$ axis; 2) dispersion curves can be interpreted if one neglects the anisotropy energy in a spins rotation around $[111]$ axis. Furthermore, our results agree with the fact that deformations due to the existence of easy spin alignment directions have not been observed.

It should be outlined that a few papers [15, 6, 24], following some studies in presence of an applied magnetic field, attribute to K_2 values much larger than ours but very divergent from each other. Our opinion is that the presence of an external magnetic field may strongly modify the Hamiltonian of the system if the Zeeman energy is larger than the dipolar energy or the dipolar magneto-elastic coupling energy.

We shall now show how the application of mechanical stresses can modify the preceding results.

4. Application of mechanical stresses. — In presence of a mechanical stress described by the tensor with components σ_{ij} in axes $\mathbf{OX}_j$, we obtain the strains and the spins orientation at equilibrium at 0 K by minimizing the free energy $F_t(\mathbf{X}, \varepsilon)$ with respect to the components X_i and ε_{ij} of respectively the mean value of sublattice α spin and crystal strains. The free energy is

$$F_t(\mathbf{X}, \varepsilon) = E_t(\mathbf{X}, \varepsilon) - \sigma_{ij} \varepsilon_{ij},$$

$$E_t(\mathbf{X}, \varepsilon) \text{ being defined by relation (3).}$$

Minimizing with respect to ε_{lm} yields :

$$\varepsilon_{lm} = s_{lmij}(\pi_{ij} + \sigma_{ij}),$$

π_{ij} being defined by (4a). ε_{lm} may be considered as the sum of three terms : ε_{lm}^e , ε_{lm}^d previously defined by (8a) and (8b), and $\varepsilon_{lm}^e = s_{lmij} \sigma_{ij}$. The ε_{lm}^e 's will take the same values than without stress, while the ε_{lm}^d 's will vary if the stresses cause the spins orientation at equilibrium to vary.

Substituting the ε_{lm} 's by their values, we obtain a free energy $F_t(\mathbf{X})$ which depends only on the spins orientation :

$$F_t(\mathbf{X}) = E_t(\mathbf{X}) - s_{lmij} \pi_{lm} \sigma_{ij} - \frac{1}{2} s_{lmij} \sigma_{lm} \sigma_{ij},$$

$E_t(\mathbf{X})$ being defined by relation (5). The last term in $F_t(\mathbf{X})$ does not depend on $\mathbf{X}$. The second one corresponds to the variation of the exchange and dipolar energies with the strains ε_{lm}^e created by the external stresses. Only the variation of dipolar energy with ε_{lm}^e depends on the orientation of the mean value of the spins.

In order to find the spins alignment direction, we must minimize $F_t(\mathbf{X})$ with respect to $\mathbf{X}$. We shall study the cases where one applies a hydrostatic pressure, a uniaxial stress along a $[111]$ axis and a uniaxial stress along a $[100]$ axis. We neglect the elastic stiffness coefficients c_{14} , c_{15} and c_{45} which may appear in the rhombohedral phase; for evaluating orders of magnitude we shall use the values of c_{11} , c_{12} and c_{44} proposed by Oliver.

4.1 HYDROSTATIC PRESSURE AND UNIAXIAL STRESS ALONG $[111]$. — In these two cases, the stress tensors are respectively, in the axes $\mathbf{OX}_j$: $\sigma_{ij} = -\sigma \delta_{ij}$ and $\sigma_{ij} = -\sigma/3$ (with σ positive for a compression). The strain tensors created by these stresses are :

$$(\varepsilon_{ij}^e)_H = -(s_{11} + 2s_{12}) \sigma \delta_{ij} \text{ for a hydrostatic pressure,}$$

$$(\varepsilon_{ij}^e)_{111} = -\frac{1}{3}[(s_{11} + 2s_{12}) \delta_{ij} + 2s_{44}(1 - \delta_{ij})] \sigma \text{ for the second case.}$$

For both cases the free energy becomes :

$$F_i(w, \varphi) = F_0 + (E_2 + F_2) w^2 + E_4 w^4 + E_3 w(1 - w^2)^{3/2} \sin 3\varphi$$

where F_0 is a constant and E_2, E_4, E_6 are defined by (6). F_2 comes from the variation of the dipolar interactions with the strains ε_{ij}^c :

$$(F_2)_H = 3 K(\varepsilon_{11}^c)_H (D_{14} + 2 D_{15})$$

$$(F_2)_{111} = 3 K[(\varepsilon_{11}^c)_{111} (D_{14} + 2 D_{15}) + 2(\varepsilon_{12}^c)_{111} (D_{44} + 2 D_{45})]$$

The results of the minimization of F_i will therefore be the same as those of the minimization of E_i ; only the numerical values of the angle w_0 and the anisotropy constants K_1, K_2 will change.

Using the values proposed by Oliver for the elastic stiffness coefficients, we obtain (σ is expressed in N/m^2) :

$$(\varepsilon_{ij}^c)_H = -0.219 \times 10^{-11} \sigma \delta_{ij},$$

$$(F_2)_H = 1.6 \times 10^{-5} \sigma \text{ J/m}^3$$

$$(\varepsilon_{ij}^c)_{111} = -[0.073 \delta_{ij} + 0.21(1 - \delta_{ij})] \cdot 10^{-11} \sigma,$$

$$(F_2)_{111} = -1.7 \times 10^{-5} \sigma \text{ J/m}^3.$$

For a stress of order 1 kbar ($\sigma \simeq 10^8 \text{ N/m}^2$), we find respectively for $(\varepsilon_{11}^c)_H, (\varepsilon_{11}^c)_{111}$ and $(\varepsilon_{12}^c)_{111}$: $-2.19 \times 10^{-4}, -7.3 \times 10^{-5}$ and -2.1×10^{-4} ; and, for $(F_2)_H$ and $(F_2)_{111}$ respectively : $1.6 \times 10^3 \text{ J/m}^3$ and $-1.7 \times 10^3 \text{ J/m}^3$. The relative variation F_2/E_2 is of order 10^{-3} , and so are the relative variations of w_0, K_1 and K_2 .

Thus, a hydrostatic pressure and a uniaxial stress along a [111] axis yield qualitatively similar results at the level of an antiferromagnetic domain. It is however well known that a uniaxial pressure along a [111] axis favours the three domains associated with this ternary axis, with respect to the other antiferromagnetic domains.

4.2 STRESS ALONG A [100] AXIS. — The stress tensor is $\sigma_{ij} = -\sigma \delta_{i1} \delta_{j1}$ (with σ positive for a compression). The non zero associated strains are :

$$(\varepsilon_{11}^c)_{100} = -s_{11} \sigma, \quad (\varepsilon_{22}^c)_{100} = (\varepsilon_{33}^c)_{100} = -s_{12} \sigma.$$

The free energy in variables w, φ writes :

$$F_i(w, \varphi) = F_0 + (E_2 + (F_2)_{100}) w^2 +$$

$$+ F'_1 w(1 - w^2)^{1/2} \sin \varphi + F'_2(1 - w^2) \cos 2\varphi + E_4 w^4 + E_3 w(1 - w^2)^{3/2} \sin 3\varphi$$

F_0 is a constant different of the preceding one, F_2, F'_1 and F'_2 come from the variation of dipolar interactions with the ε_{im}^c strain; by applying a strong enough stress, we may make these parameters much larger than E_4 and E_3 . The spin orientation will therefore be found with a good approximation by minimizing the expression

$$F_0 + (E_2 + (F_2)_{100}) w^2 + F'_1 w(1 - w^2)^{1/2} \sin \varphi + F'_2(1 - w^2) \cos 2\varphi.$$

When F'_2 is positive, which is the case in our model, the minimum is obtained for $\cos \varphi = 0, \sin \varphi = \pm 1, w = \mp w_0$; if we assume $E_2 + (F_2)_{100}$ to be much greater than the other terms, w_0 simply evaluates to $F'_2/2(E_2 + (F_2)_{100})$.

Using the values proposed by Oliver for the elastic stiffness coefficients, we get :

$$(\varepsilon_{11}^c)_{100} = -0.719 \times 10^{-11} \sigma,$$

$$(\varepsilon_{22}^c)_{100} = 0.25 \times 10^{-11} \sigma$$

$$F_2 = 6.38 \times 10^{-6} \sigma \text{ J/m}^3,$$

$$F'_2 = 7.68 \times 10^{-6} \sigma \text{ J/m}^3$$

$$F'_1 = -2.46 \times 10^{-5} \sigma \text{ J/m}^3.$$

For a compression of one kbar ($\sigma \simeq 10^8 \text{ N/m}^2$), this yields

$$(\varepsilon_{11}^c)_{100} = -7.2 \times 10^{-4},$$

$$(\varepsilon_{22}^c)_{100} = 2.5 \times 10^{-4},$$

$$F_2 = 6.4 \times 10^2 \text{ J/m}^3,$$

$$F'_1 = -2.5 \times 10^3 \text{ J/m}^3,$$

$$F'_2 = 7.7 \times 10^2 \text{ J/m}^3.$$

From that, we deduce for w_0 a value next to -5.2×10^{-4} .

We see that a compression along a [100] axis contributes to moving the spin alignment direction away from the (111) plane. This phenomenon could still be observed with stresses of order 10 kbar. Moreover, such stresses suppress two of the three antiferromagnetic domains associated, in the absence of stresses, with every ternary axis. It should be possible to experimentally observe this phenomenon. Last by applying a stress with components on both the [111] and [100] axes, one should create in MnO a unique antiferromagnetic domain. Such a result would be very interesting for the magnetic study of this compound.

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