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## PIEZOMAGNETISM AND DOMAINS IN MnF2

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Abstract. – Neutron topographic experiments show that the simultaneous application of an uniaxial stress and a magnetic field when cooling a  $MnF_2$  crystal through  $T_N$  favours one type of 180° antiferromagnetic domain. This appears to be related to the piezomagnetism of  $MnF_2$ , in spite of the weakness of this effect, which was measured in our case by a SQUID.

 ${\rm MnF_2}$  is antiferromagnetic below  $T_{\rm N}\simeq 67~{\rm K}$ . Magnetic moments of opposite sign, directed along the *c*-axis, are respectively located at the corners and the center of the tetragonal unit cell. These two sites, although equivalent, are not related by a lattice translation (the fluorine environment being rotated by 90° around c) leading to two possible 180° antiferromagnetic domains.

The puzzling behaviour of these domains under an applied magnetic field was pointed out more than 25 years ago [1], and more fully investigated when polarized neutron diffraction topography, the only technique available for their observation, was developed [2]: the domain structure remaining in virtually zero field is nearly reproduced when the crystal is cooled from the paramagnetic to the antiferromagnetic phase under a small field (typically  $10^{-2}$  T), the domains changing sign and mostly retaining the wall locations if the field is reversed. The 210 reflection was used to investigate the domains because, at about 20 K, the absolute values of the nuclear and magnetic structure factors are almost exactly equal. This implies, for neutrons polarized along [001], that only one type of domain contributes to diffraction. The flipping ratio R is hence almost proportional to the ratio of the volumes occupied by the two kinds of domains. Figure 1 shows the R obtained by cooling the sample under the simultaneous application of a magnetic field (0.01 T)along [001] and an uniaxial stress along [110], and a schematic drawing of the corresponding topographs : it shows that one type of domain is favoured with respect to the other. The uniaxial stress was not in this case completely uniform, and the single domain state was not reached.

The transition metal fluorides exhibit piezomagnetism, i.e. a magnetic moment results from an applied uniaxial stress, its magnitude being proportional to the stress. Theoretical treatments of this effect were



Fig. 1. – Flipping ratio R as a function of the stress applied during the cooling of the MnF<sub>2</sub> crystal  $(4 \times 4 \times 2.5 \text{ mm}^3)$  through  $T_N$  in a 0.01 T magnetic field, and schematic drawing of the topographs recorded after removing the field, at 20 K, with neutrons polarized along [001].

worked out [3, 4] and measurements of the piezomagnetic coefficients were performed by Borovik-Romanov and co-workers, mainly on  $\text{CoF}_2$  [5, 6] but also on  $\text{MnF}_2$  [5], the measured coefficients being then at the limit of the possibilities of the instrumental set-up [7]. The experiment described in figure 1, as well as the previous ones [1, 2], appears to be in qualitative agreement with a piezomagnetism related field-domain interaction mechanism, as was found for  $\text{CoF}_2$  [8]. The sign of the piezomagnetic coefficients is different for the two domains, and consequently the sign of the resulting magnetic moment. A magnetic field can thus interact with the domains.

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It appeared interesting to check this kind of explanation by measuring one of the piezomagnetic coefficients again in a different way. ( $\Lambda_{xyz}$  following the notation of [6]). We used for this purpose a SQUID magnetometer. The crystal was mounted in a device designed to apply uniaxial stresses but to give only a weak magnetic signal. It was checked that the applied stress remains practically constant when the temperature is lowered. We limited the applied stress to  $6 \times 10^6$  Nm<sup>-2</sup> to avoid the occurence of (101) cracks. The measurement is only possible if the crystal is in a practically single domain state. The experimental procedure to favour one type of domain consisted in applying at room temperature the required uniaxial stress, then cooling the sample in a field of 0.6 T directed along the c axis, and finally performing the measurement of the sample magnetization, along c, in a nominally zero field. The residual field in which the measurements were carried out was constant for a given run (for a given applied stress) and we measured it, at the end of each run, by using a well known paramagnetic substance : it appeared always to be within a range of  $\pm 3 \times 10^{-5}$  T. The measurements were then corrected for the magnetization corresponding to the AF susceptibility of MnF2, and induced by this residual field, in order to obtain the actual zero field value.

Figure 2 shows the results obtained for the magnetization as a function of the temperature and the applied uniaxial stress. It indicates clearly that no piezomagnetic moment is present in zero applied stress, at any temperature, or above the Néel temperature, for any stress. A magnetization, which increases linearly with the stress, occurs below the Néel temperature. These results were obtained by subtracting the cell signal,



Fig. 2. – Magnetic moment of a  $MnF_2$  crystal  $(8.5 \times 7 \times 2 \text{ mm}^3)$ , measured in zero field after cooling the sample simultaneaously under an applied stress and a field of 0.6 T, as a function of this stress and the temperature.

measured independently : it was nearly the whole signal above  $T_{\rm N}$ , and most of it ( $\simeq 90$  %) below. This limits the accuracy, and although our experiment leads to a fair measurement of the piezomagnetic coefficient at about 60 K ( $\Lambda_{xyz} = 7 \times 10^{-8}$  AN<sup>-1</sup>m in good agreement with [5]) it does not appear possible to go further and to extract its temperature dependence in the ordered phase, without reducing the cell signal.

The weakness of the piezomagnetism of  $MnF_2$  is such that at about 65 K, 1) the piezomagnetic moment induced by the maximum stress we applied corresponds to the magnetization induced in  $MnF_2$  by a field of the order of  $10^{-4}$  T, and 2) the Zeeman energy is of the order of kT, under a field of 0.01 T, for regions of about 1  $\mu$ m in diameter.

Preliminary computer simulations seem nevertheless to indicate that the explanation for the field-domain interaction could reside in the dynamics of the paraantiferromagnetic transition combined with the piezomagnetism [9]. In these two dimensional simulations the sample is subdivided in squares whose "volume" is assumed to be such that their piezomagnetic energy is much larger that the thermal fluctuations. Local conditions of "neighbourhood" (the sign which predominates within the neighbouring cells is favoured for the one considered : this corresponds physically to reducing the wall surface) are in competition with "piezomagnetic" conditions. Very weak piezomagnetic terms can determine, within this frame, the sign of the domain in a given region because the first criterion is often inoperative in the first steps, a cell being surrounded by as many 'plus' than 'minus' neighbours.

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