SOVIET PHYSICS – SOLID STATE VOL. 12, NO. 1 JULY, 1970

MAGNETIC SUBLATTICE INVERSION IN UNIAXIALLY COMPRESSED MANGANESE FLUORIDE

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Translated from Fizika Tverdogo Tela, Vol. 12, No. 1,
pp. 83-88, January, 1970
Original article submitted July 21, 1969

A study has been made of the effect of uniaxial compression on the critical field for inversion of the magnetic sublattice of antiferromagnetic MnF_2 at T = 4.2°K. The method used allows an external pulsed magnetic field to be established along the symmetry axis of the crystal with an accuracy of $\vartheta \leq 5'$, and comparative measurements to be made of H_c for compressed and free specimens. The minimum width of the transition region close to $H_c = 91.7$ kOe is ~300 Oe and is doubled when $\vartheta \sim 20'$. When uniaxial compression is applied along the four-fold axis the transition region becomes wider, with H_c growing almost linearly with pressure, such that $(1/H_c)(dH_c/dp) = 2.9 \cdot 10^{-12}$ cm²/dyn. The magnitude of the effect is in agreement with the size of the magnetostriction jump in the critical field, measured earlier. An analysis of the contributions of magnetic-dipole coupling and classical magnetostriction shows that part of the effect is due to the dependence of the exchange integrals J_{12} between ions of opposite sublattices on the interatomic distances, where $-(1/\chi_\perp)(d\chi_\perp/dp) = (1/J_{12})(dJ_{12}/dp) = 1.9 \cdot 10^{-12}$ cm²/dyn.

Molecular field theory has established [1] a simple link between the magnetic susceptibility $\chi_\perp$ of an antiferromagnet measured in sufficiently strong fields at low temperatures, and the intersublattice exchange integral J_{12}. From this it follows that the relationship between J_{12} and the interatomic distances can be reliably determined by measuring how $\chi_\perp$ is influenced by the external pressure. However, attempts at measurement come up against the difficulty of detecting small increments $\chi_\perp(p)$ against a background of large $\chi_\perp$ values, which is scarcely feasible at the accuracy level of traditional methods. The problem can be resolved, however, through a study of the pressure dependence of the critical field for inversion of the magnetic sublattice H_c, since the field strength can be measured quite accurately. In fact at low temperatures H_c can be written in the form

$$H_c = \sqrt{\frac{K}{\chi_\perp}}, \qquad (1)$$

where K is the anisotropy constant. Since it is often possible to calculate the relationship between K and the pressure, by making an experimental study of H_c we can also determine the sought-for $\chi_\perp(p)$ relationship.

1. MEASUREMENT OF THE CRITICAL FIELD

The inversion field of the MnF_2 magnetic sublattice is equal to $H_c \simeq 95$ kOe [2]. A magnetic field of this value can be obtained without difficulty under pulsed conditions. In the present investigation a pulsed field was developed in a cooled multiturn solenoid (internal diameter 25 mm, length 100 mm), which provided a high, uniform field within the specimen. When the capacitor bank was discharged through the solenoid, with a total charge energy of up to 75 kJ, a magnetic field pulse (H) was created in the solenoid which reached its maximum value (up to 300 kOe) in $7.5 \cdot 10^{-3}$ sec. A field

proportional to H was used to deflect the beam of a S1-16 electron oscillograph. By increasing the sensitivity of the oscillograph X channel and simultaneously applying an initial deflection to it, it was possible to examine individual sectors of the field pattern on the screen at scales up to 300 Oe/cm.

The MnF_2 specimen, oriented by x-ray methods to an accuracy of 2°, was mounted in the "finger" of a metal cryostat lying within the solenoid and cooled to the boiling point of liquid helium. When the strength of the external magnetic field reached the value H_c, there was a rapid inversion of the magnetic sublattice of the specimen in the plane perpendicular to the field. At this point there was an abrupt jump in the crystal magnetization [2], and the differential susceptibility $\chi_d = dM/dH$ attained a sharp maximum. Changes in the magnetization of the specimen were recorded by the induction method [3]. In the induction coil [(2), Fig. 1] surrounding the specimen 1 a sharp emf splash arises, proportional to χ_d and reaching a maximum value of about 2 V. This emf was fed to the Y channel of the oscillograph. This results in a sharp blip appearing on the extended field scale, corresponding to inversion of the sublattice (Fig. 2, first splash).

The width of this splash determines the accuracy in finding H_c, and is extremely critical with regard to the orientation of the external field relative to the axis of ordering. The solenoid was secured with micrometer screws, which make it possible to tilt the solenoid to adjust the field direction. The correctness of alignment of the field along the specimen axis was judged from the half-width and amplitude of the differential susceptibility splash. The field could thus be lined up to an accuracy of not less than 5 min of arc.

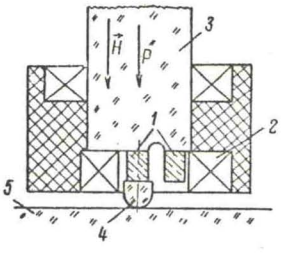

Fig. 1. Apparatus for exerting uniaxial pressure on a specimen.

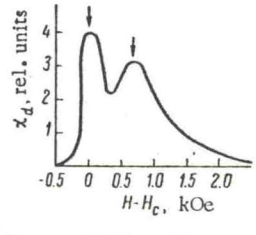

Fig. 2. Differential susceptibility of free (p = 0) and deformed (p = 2.5 katm) MnF_2 specimens, plotted against magnetic field strength close to $H = H_c$. T = 4.2°K.

2. METHOD OF CREATING UNIAXIAL PRESSURE

The method of imposing uniaxial pressure on the specimen is clear from Fig. 1. A reliable relative reading of the shift H_c under pressure was obtained by simultaneously observing signals on the oscillograph screen from deformed and free specimens. The orientations of these specimens could not differ more than 5-10'. The required coaxiality was achieved in the following way. On the quartz rod 3 transmitting the external pressure one wide specimen was glued and then cut into two parts (Fig. 1). On one part was glued a quartz hemisphere 4, resting on a quartz bearing surface 5. The other part of the specimen was left free. Fused quartz was chosen as the construction material because of its elastic properties and its coefficient of thermal expansion, close to the corresponding value for MnF_2 in the basal plane. This meant that nonuniform stresses in the specimen could be avoided during cooling. Pressure was transmitted to the quartz rod and the specimen through an argentan tube, loaded outside the cryostat with weights up to 30 kg. Specimens were held at pressures up to 3 katm without breakdown. The hermetic seal where the tube emerged from the cryostat had a dry frictional threshold not greater than 0.2 kg. The cross-sectional dimensions of the specimen were found using a traveling microscope, to an accuracy of ±5 %.

3. THE TRANSITION WIDTH

If we consider the inversion of a magnetic sublattice as a phase transition of the first kind, then as the magnetic field gets closer in direction to the axis of ordering the differential suceptibility χ_d at $H = H_c$ must increase, attaining an infinite value when the directions coincide exactly ($\vartheta = 0$). It has been noted experimentally that χ_d reaches a definite maximum, the value of which falls by a half when the magnetic field moves off the axis by $\vartheta \sim 20'$. The high uniformity of the magnetic field in the position of the specimen rules out a trivial explanation of the finite transition width. (The minimum width, reckoned as the magnetic field region within which χ_d exceeds half its maximum value, is

~300 Oe.) It must be pointed out that in certain cases [4, 5] magnetic sublattice inversion is not a phase transition of the first kind, and is spread over a field-strength region of the same order as that observed experimentally. An investigation of the mechanisms considered in [4, 5] is of independent interest. The methods we have used to attain exact angular alignment make it hopeful that such an investigation will be completed soon. The transition width found in the present study cannot be unambiguously interpreted, due to the masking effect of the demagnetizing fields of the specimen. The experimental conditions did not permit this to have the shape of an ellipsoid, so the demagnetizing fields in the specimen were nonuniform with a strength estimated at ~400 Oe, which fully explains the observed transition region width.

A relatively small transition width may be quite sensitive toward the state of a crystal. In our experiments the effects of structure defects on transition widths were not investigated. From the reproducibility of the results and the absence of any traces of block structure under the polarizing microscope as on x-ray photographs, it may be expected that their contribution would have been negligible. When pressure was applied the transition width increased, doubling at $p \sim 2.5$ katm. We suggest that this broadening was due to deviation of the pressure axis direction away from the ordering axis by ±5° and to pressure nonuniformity in the bulk of the specimen.

4. EXPERIMENTAL RESULTS

Figure 2 shows the differential susceptibilities χ_d of both specimens plotted against the magnetic field strength, with a uniaxial pressure of 2.5 katm applied to one specimen. The absolute value of the critical field for a free specimen (first χ_d splash) at 4.2°K was 91.7 ± 1 kOe, according to our measurements. The critical field of the compressed specimen, determined from the position of the second splash, was displaced by 700 Oe. The error in the relative scales of the magnetic field on the oscillogram is estimated at 5%.

Imposition of uniaxial pressure on a specimen leads not only to a shift in H_c but also to a broadening of the transition. This makes measuring the shift more difficult, particularly at low pressures, when the individual splashes are poorly resolved. By stabilizing the supply circuits to the apparatus it was later possible to get quite good reproducibility in the position of the χ_d maximum on the scale (not poorer than 50 Oe), which allowed the free-specimen peak to be distinguished. This made it possible to determine the H_c shift for lower pressures.

Figure 3 is a graph of the relationship $\Delta H_c(p)$. Within experimental error limits it can be described as a straight line of slope $(1/H_c)(dH_c/dp) = 2.9 \cdot 10^{-12}$ cm²/dyn.

These results were reproduced with three specimens, and the relationship quoted refers to a specimen in the shape of a cylinder (D = 0.76 mm, l = 1.2 mm).

5. DISCUSSION

Returning to the problem expressed at the beginning of the article, we will consider the individual mechanisms contributing to the measured values of $H_c(p)$. These include: 1) classical magnetostriction; 2) change in the anisotropy constant with pressure; and 3) the sought-for quantity $d\chi_\perp/dp$. The first two contributions can be calculated, and the last one found by experiment. Classical magnetostriction is a phenomenon in which a magnetic substance in an external field is subject to forces causing its elongation along the magnetic field direction and its compression perpendicular to this direction. With the specimen form factor taken into account, evaluations in [6] give $\Delta U_c = 0.24 \cdot 10^{-5}$ and hence $[(1/H_c)(dH_c/dp)]_\perp = 0.27 \cdot 10^{-12}$ cm²/dyn. Possible inaccuracies in determining the specimen's effective form factor are not of particular importance in view of the small size of the contribution.

The anisotropy energy of the crystals considered is almost entirely of magnetic-dipole origin [7], with the anisotropic part of the magnetic dipole field equal to

$$\mathbf{H}_a = \sum_i \frac{3(\boldsymbol{\mu}\mathbf{r})\,\mathbf{r}}{r_i^5}, \qquad (2)$$

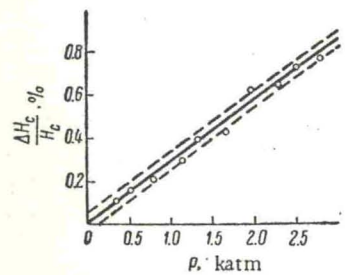

Fig. 3. Relative changes in the critical field values of a MnF_2 speciment plotted against uniaxial pressure applied along the fourfold axis. The broken lines enclose the average scatter values of the experimental points.

where μ is the magnetic moment of the ion creating the field and $\mathbf{r}$ is its radius vector relative to the point considered. The total field is obtained by summing over all the magnetic ions of the crystal. In calculating how the anisotropy energy varies with the deformation, we will use an approximate method. We represent K in the form

$$K \sim \frac{\gamma}{v}. \tag{3}$$

This relationship between K and the volume of the unit cell ($v = a^2c$) is tied up with the fact that the field due to magnetic dipoles decreases as the cube of the interatomic distances [see (2)]. The quantity $\gamma = (a - c)/(a + c)$ is a dimensionless parameter characterizing the extent to which the magnetic lattice deviates from a body-centered cubic lattice. The anisotropy field of the latter equals zero, and (3) can be considered as the first term in the series of an expansion of K in terms of γ. Direct differentiation of (3) with respect to the relative changes in the unit cell constants along the different crystallographic directions gives

$$\frac{1}{K}\frac{\partial K}{\partial U_a}\Big|_c = 0.53, \quad \frac{1}{K}\frac{\partial K}{\partial U_c}\Big|_a = -3.6 \text{ and } \frac{1}{K}\frac{dK}{dp} = \frac{1}{K}\frac{\partial K}{\partial U_c}S_{33} + \frac{1}{K}\frac{\partial K}{\partial U_a}S_{13} = -3.4 \cdot 10^{-12} \text{ cm}^2/\text{dyn}. \tag{4}$$

Here $S_{33} = 0.90 \cdot 10^{-12}$ cm^2/dyn and $S_{13} = 0.36 \cdot 10^{-12}$ cm^2/dyn are the elastic compliance constants of the crystal, calculated from ultrasonic wave velocities at low temperatures [8] and data in [9]. The error in this calculation is because γ is not small. However, direct calculation of the quantity in (4) by summing the contributions of the sixteen nearest neighbors gives the same value.

The quantity $\chi_\perp(p)$ in the measured quantity can be determined from the expression

$$\frac{1}{H_c}\frac{dH_c}{dp} = \frac{1}{2}\left(\frac{1}{K}\frac{dK}{dp} - \frac{1}{\chi_\perp}\frac{d\chi_\perp}{dp}\right), \tag{5}$$

derived from (1) by differentiation, where the term on the left represents the experimental result after subtraction of the classical effect. Using the relationship $\chi_\perp \sim 1/J_{12}$ [1] we find

$$-\frac{1}{\chi_\perp}\frac{d\chi_\perp}{dp} = \frac{1}{J_{12}}\frac{dJ_{12}}{dp} = 1.9 \cdot 10^{-12} \text{ cm}^2/\text{dyn}.$$

This relationship, which indicates that the bulk interaction between the sublattices grows when the crystal is uniaxially compressed, gives only part of the information on the relationship between J_{12} and the interatomic distances.

In fact the expression

$$\frac{1}{J_{12}}\frac{dJ_{12}}{dp} = \frac{1}{J_{12}}\frac{\partial J_{12}}{\partial U_c}\Big|_a S_{33} + \frac{1}{J_{12}}\frac{\partial J_{12}}{\partial U_a}\Big|_c S_{13} \tag{6}$$

is an equation with two unknowns. A similar equation relating to H_c measurements under hydrostatic pressure conditions, also retaining the initial symmetry of the crystal, can be used in conjunction with (6) to determine both quantities of interest; the relevant experiments are being carried out at the present time. However, even now it is possible to compare our results with those from other experiments. There is qualitative agreement between our results and those of Gibbons [10], who observed marked contraction of a MnF_2 crystal along the symmetry axis during antiferromagnetic ordering, and also with the results obtained by Astrov, Novikova, and Orlova [11] and by Benedek and Kushida [12], who observed an increase in the temperature of ordering (T_N) with homogeneous compression. Quantitative agreement with the cited results should not be expected, since the latter include the relationship between the interatomic distances and the sublattice exchange integral J_{11}, the contribution of which cannot be neglected.

Finally, if we examine the inversion of magnetic sublattices as a phase change of the first kind, we can write the equation linking the change in magnetic field with the uniaxial pressure as a phase equilibrium curve [5]:

$$\frac{dH_c}{dp} = -\frac{\Delta U_c}{\Delta m}, \tag{7}$$

where ΔU_c and Δm are respectively the jumps in the relative crystal deformation along the C_4 axis and in the magnetic moment. This expression, which is analogous to a Clapeyron-Clausius equation, allows data on critical field displacements with pressure to be used to calculate the corresponding jumps in crystal dimensions at the critical point. The result $\Delta U_c = 0.26 \cdot 10^{-4}$ is in agreement with direct magnetostriction measurements on MnF_2 in a strong field [13].

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