Twinning structure and orientational magnetic transition in antiferromagnetic $RbMnCl_3$

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In a crystal of $RbMnCl_3$ in the antiferromagnetic phase, a significant interrelationship has been detected between the directions of crystal strain, both spontaneous and produced by external interactions, and the sublattice magnetizations. It is demonstrated for the first time that an external magnetic field can untwin the crystal and change the configuration of the crystalline domains. It is concluded that exact knowledge of the twinning structure is necessary in investigation of the magnetic properties of a crystal.

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INTRODUCTION

Recently a large number of problems have been solved that are related to the effect of the spin system of magnetic materials on their optical properties. The achievements in this area are now being used with increasing frequency for study of the magnetic characteristics of crystals by spectroscopic methods. The present paper is one of the results of the development of investigations in this direction.

In recent publications,[1,2] a detailed description has been given of a structural phase transition in the crystal $RbMnCl_3$ ($T_c = 272$ K) and of its magnetic properties in the antiferromagnetic phase ($T_N = 94.6$ K). In this article, we report on a significant interdependence that we have discovered between the twinning and magnetic configurations in $RbMnCl_3$. The essence of this phenomenon consists of the following. When $T < T_N$, the action on an external magnetic field on a specimen free from mechanical stresses leads to an irreversible rearrangement of the crystalline domains (CD) by displacement of the domain boundaries. On the other hand, in the absence of a magnetic field a transformation of the twinning structure is accompanied by spin reorientation. In each CD, the antiferromagnetism vector **l** follows after the direction of the strain.

EXPERIMENTAL METHOD

The measurements were made on a low-temperature magnetooptical setup whose construction made it possible to act on the specimen with a uniaxial pressure and a magnetic field, to measure its temperature from room temperature to 4.2 K, to make observations on the twinning structure, and to monitor the absorption spectrum. The $RbMnCl_3$ specimens were cut in the form of plates of thickness 1.5–2 mm, perpendicular to the original sixfold axis, and were mounted in an optical cryostat. The direction of propagation of the light through the specimen coincided, with accuracy up to 5°, with the axis c_6. The magnetic field of a superconducting solenoid, of Helmholtz construction, was applied in the direction perpendicular to the specimen. The absolute and relative errors of measurement of the intensity were 0.1 and 0.05 kOe respectively. The experiments in a magnetic field were done at temperature 4.2 K.

The construction of the cryostat provided for a possibility of rotating the specimen about the direction of propagation of the light, through an arbitrary angle, during the measurements. The presence of visual monitoring made it possible to read the angle of rotation with an accuracy within 1°. Uniaxial pressure was applied to the specimen by two methods. It was either clamped in a prescribed direction in a movable ring, or mounted in a special press with a regulated load. In the first case the value of the pressure was not monitored, but the possibility of rotating the specimen in the magnetic field remained. In the second, a monitored pressure was applied to an immovable specimen. Under the necessity of studying unstressed crystals, the specimens were mounted in the cryostat freely, without clamps or adhesives.

The observation on the twinning structure of the crystal was done visually in crossed polarizers. In each specific case, the positions of the polarizer and analyzer were chosen for best contrast of the CD picture.

The determination of the direction of the vector **l** was based on the polarization property of the exciton absorption line at frequency 23509.5 cm^{-1} (the optical transition ${}^6A_{1g} \rightarrow {}^4A_{1g}, {}^4E_g$ in the Mn^{2+} ion). The intensity of this line depends on the relative directions of the vector **l** and of the polarization of the light **E**: it is a maximum when $\mathbf{E} \parallel \mathbf{l}$ and a minimum when $\mathbf{E} \perp \mathbf{l}$. The absorption coefficients are respectively 8 and 1 cm^{-1}.[3 1)] Thus by determining the direction of **E** that corresponded to the maximum absorption, we found the orientation of **l** in the crystal. In the case of a single-domain structure, this operation was performed for each of the CD separately. The monitoring of the absorption spectrum was done with a DFS-13 spectrometer (linear dispersion 4 Å/mm) with a photoelectric recording system. The latter operated in one of two modes: scanning, with output of the section of the spectrum under investigation to an oscillograph, or fixed wavelength, with registry on a recorder of the intensity of light at the frequency of the absorption line.

EXPERIMENTAL RESULTS

Crystalline domains in a magnetic field. A structural phase transition in $RbMnCl_3$, with change of symmetry $D_{6h}^4 \rightarrow C_{2h}^2$, occurs at $T_c = 272$ K. In the low-tem-

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perature phase, three equally privileged directions, perpendicular to the original axis c_6, are allowed for the second-order axis, and the crystal becomes twinned. The value of the double refraction Δn_c increases greatly with lowering of the temperature,[1] and the contrast of the CD picture in crossed polarizers is heightened. We recorded the existence of different CD with certainty for $T \lesssim 200$ K. The dimensions of the regions of equal darkening were of the order of 1 mm. During the transition through $T_N = 94.6$ K, no clear changes were observed (from the point of view of the conclusions of our work, this result requires careful verification in a special experiment).

In a specimen free from mechanical stresses, cooled to 4.2 K, the CD picture was found to be strongly subject to the influence of the magnetic field. Smooth rotation of the specimen in a field of the order of 10 kOe was accompanied by continuous changes of dimensions, shape, and number of CD. An analogous phenomenon was observed also on an immovable specimen when a magnetic field was turned on. The origination of each new domain occurred only at the boundaries of the crystal. Transformation of the structure occurred by displacement of the domain boundaries, which could be either smooth or by jumps. Furthermore, the mobility of the boundaries was determined by the complexity of their configuration. Solitary straight boundaries behaved with the greatest mobility in a field.

Figure 1 gives some examples of the transformation of the CD structure in an unstressed specimen. The arrows denote the vectors **l** (correct except perhaps for sign), which coincide within each CD with the axis of the indicatrix section. The direction of the field H is horizontal in the plane of the figure. The specimen was diaphragmed, and the edge regions are not visible. The diameter of the visible part of the specimen is 5 mm. Each of the sequences of photographs *a*, *b*, and *c* corresponds to a smooth variation of the magnetic field. The positions of the specimen in the solenoid differ: in Figs. 1a and 1b by 30°, in Figs. 1a and 1c by 60°.

The changes of the domain structure shown in Fig. 1a began at $H = 8.6$ kOe and continued smoothly at this intensity for several minutes (the photographs were taken at intervals of 2 min). In the situation of Fig. 1b, the growth of the white (in the figure) domain has already begun at a field of about 4 kOe. At intensity 5.2 kOe the field was fixed, but the boundary continued to move smoothly, after a few seconds traversed the greater part of the specimen, and stopped. At $H = 7.3$ kOe, a single CD remained within the field of view. Finally, in the case shown in Fig. 1c the boundary stopped at field 6.2 kOe and did not move until 7.2 kOe, after which the new domain discontinuously occupied the whole visible part of the specimen. A characteristic feature of the process is its irreversibility: on decrease of the magnetic field to zero, the crystal remained practically a single domain (Fig. 1a, b, c).

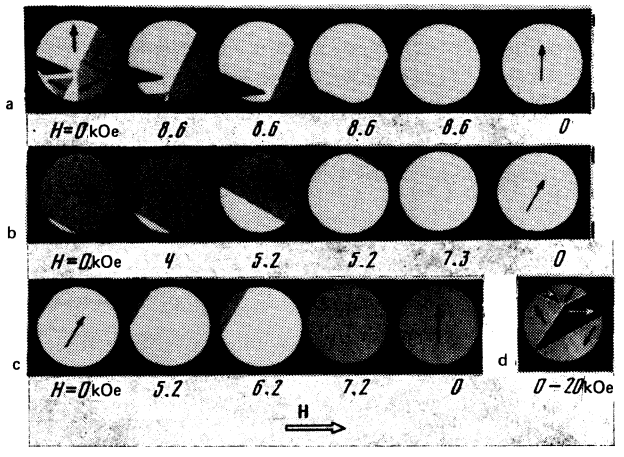

FIG. 1. Change of twin structure of $RbMnCl_3$ in a magnetic field.

Figure 1d shows an example of a "rigid" configuration of linked boundaries. Such a structure, for a fixed position of the specimen, was found to be insensitive to the magnetic field; it could be destroyed only by rotation at $H \gtrsim 7$ kOe.

In order to elucidate the relation between the observed transformation of CD and the behavior of the spin system in a magnetic field, the following experiments were set up.

Effect of pressure on the direction of the vector **l**. Figure 2 shows a typical variation of the light intensity at the frequency of the absorption line, 23509.5 cm^{-1}, with the angle between the directions of the vectors of polarization (**E**) and of uniaxial pressure on the crystal (**P**). (We note that in this experiment the whole specimen was projected on to the input to the spectrometer, and its twin structure was not monitored.) Starting from the polarization property of the indicated line (see above), we interpret this result as follows. At $P = 0$ there existed in the specimen some preferred direction of orientation of the vector **l**. An applied pressure, beginning approximately with 10 kgf/cm^2, set **l** perpendicular to **P**. Subsequent increase of the pressure to 100 kgf/cm^2 did not change the direction of **l**. It remained unchanged also after removal of the pressure; that is, the process of reorientation of **l** was irreversible.

Thus this experiment established that the directions of the strain (compression) of the crystal lattice and of the orientation of **l** in $RbMnCl_3$ are mutually perpendicular.

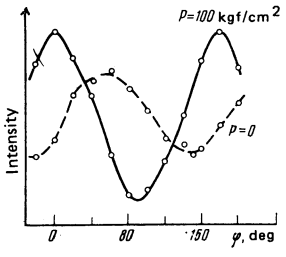

FIG. 2. Effect of uniaxial pressure on polarization of the absorption line. φ is the angle between the electric vector **E** of the light and the pressure **P**.

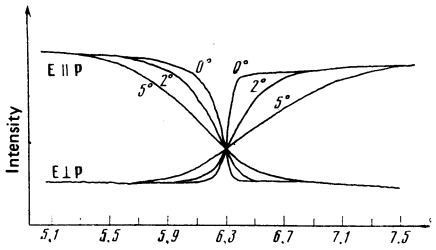

FIG. 3. Variation of light intensity at frequency 23 509.5 cm^{-1} with magnetic field (kOe) for various polarizations and angles between H and l.

Orientational magnetic transition. According to the data of Ref. 2, a magnetic field $H \geq H_c = 6.3$ kOe, applied in the basal plane, will orient the vector l in a direction $l \perp H$. We observed spin reorientation by a spectroscopic method in crystals with a fixed twin structure. The reason for this was that in the case of free boundaries, the conditions of passage of polarized light through the crystal change in a magnetic field (see Fig. 1), and this complicates the interpretation of the results. Achieving the necessary structure was possible by two methods: either by producing in the free specimen a "rigid" configuration of the type of Fig. 1d, or by use of pressure. The second method permitted great freedom of manipulation of the specimen in a field without changes of the CD structure. Choice of the optimum direction of the pressure was made by taking into account the possible spontaneous orientations of the vector l in the crystal (Fig. 1) and the relation between l and P (Fig. 2).

The results of the experiment with use of the method of uniaxial pressure are given in Fig. 3. Exact determination of the original orientation of l was accomplished experimentally on the basis of the rate of change of the transition. Both series of curves, $E \perp P$ and $E \parallel P$, reflect well the turning of the vector l through 90° at the critical field. The transition is reversible: on decrease of the field intensity, the light intensity changes along the same curves in the reverse direction. One notices the sharp dependence of the steepness of the reorientation process on the angle between H and l: this is characteristic of spin-flip transitions (see, for example, Refs. 5–7).

In this experiment, visual monitoring of the state of the crystal was done simultaneously with observation of the absorption line. It is important to note that over the whole range of magnetic fields, the form of the specimen in crossed polarizers remained unchanged. The last remark applies also to the case in which spin reorientation was observed in a free specimen with the twin structure shown in Fig. 1d.

DISCUSSION OF RESULTS

Thus in a crystal of $RbMnCl_3$ in the antiferromagnetic phase, a significant interrelation is observed between the directions of strain and of the sublattice magnetizations. The crystalline domains are simultaneously antiferromagnetic domains. Within each CD, the vector l is perpendicular to the direction of the spontaneous compression of the lattice and coincides with a twofold axis. In neighboring CD they are turned with respect to one another through 120 (60) degrees. An external pressure, causing the crystal to become single-domain, will orient $l \perp P$. In a specimen with fixed domain structure, spin reorientation in a magnetic field is reversible with respect to the intensity of H.

In rotation of the vector l by the field, the value of the magnetostriction is less than the spontaneous strain in the CD. In a free crystal the magnetoelastic energy exceeds the energy exceeds the energy of displacement of domain boundaries; therefore the magnetic transition is accompanied by reorganization of the twin structure. After completion of the transition, the specimen becomes single-domain; the direction of the symmetry axis is that one of the three possible ones that is closest to a direction perpendicular to the field (see Figs. 1b and c). Thus the magnetic orientational transition in this case is accompanied by a change of the direction of the "easy" axis. A graphic example of this mutual influence of the spin and lattice systems is the situation shown in Fig. 1b: starting under the influence of the magnetic field, a reorganization of the CD and, obviously, a rotation of l continue at intensities 5.2 kOe $< H_c = 6.3$ kOe.

It should be noted that the possibility of untwinning of a crystal by a magnetic field must be taken into account in purely magnetic measurements. As is now clear, the mechanical and thermal past history of the specimen, and also its attachment to the experimental apparatus, may show up in three ways. 1) The specimen permits free motion of domain boundaries. Spin reorientation is subject to the influence of reorganization of the twin structure. 2) The twinning configuration is fixed. Here there is no reason to expect equal relative volumes of different CD, and exact knowledge of them is necessary for interpretation of the results. 3) The crystal is in a fixed single-domain state. Apparently this situation is best for investigation of such characteristics as the susceptibility, the exchange and anisotropy fields, etc.

Finally, the phenomenon of reorganization of the twin structure of a crystal by a magnetic field, first observed here as far as is known to us, is also of independent interest. Here there is a possibility of nonlocal action upon twin boundaries, and this is undoubtedly important for study of their nature and of the mechanisms of their motion.

In closing, we express our sincere gratitude to V. V. Eremenko for constant interest in and support of the research, and to B. V. Beznosikov for kindly providing the single crystals.

[1)]We reported earlier[4] an analogous effect of polarization of electric-dipole absorption in the hexagonal antiferromagnet $CsMnF_3$. We intend to consider a possible mechanism of this phenomenon in one of our subsequent publications.

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Dynamics of interphase boundaries in antiferromagnets

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The nonlinear dynamics of an antiferromagnet in the intermediate state, in which a collinear and a flopped phase of the antiferromagnet can coexist, is investigated theoretically. The motion of an individual interphase boundary is considered with account taken of the magnetic dipole interaction. The scattering of two domain walls and their bound states are investigated. The steady-state motion of a domain wall is studied with account taken of relaxation under the influence of an external magnetic field. The possibility of observing this phenomenon in experiment is discussed.

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It is known that in antiferromagnets (AFM) there can exist 180° domain walls (DW) that have many features in common with the domain walls of ferromagnets. The most substantial difference between the DW in these magnets is that the 180° DW in AFM are not topological DW and are therefore metastable.

However, as shown by V. Bar'yakhtar *et al.*,[1] near first-order transition points there can exist in AFM a thermodynamically stable domain structure with 90° DW (intermediate state of AFM). These DW are of great interest primarily because they are separate essentially different phases of the AFM (the collinear phase $\Phi_{\parallel}$ and the flopped phase $\Phi_{\perp}$). Since the susceptibilities of these phases differ substantially, when the external magnetic field (or some other external parameter) is changed, the phase-equilibrium condition is violated and the 90° DW goes into motion. Thus, the question arises of experimental and theoretical study of the dynamics of the interphase boundaries in AFM under the influence of an external magnetic field.

Investigations of this kind are being quite intensively pursued in recent years in connection with the study of the motion of DW in ferromagnets and in ferrites. However, the motion of DW in AFM has a number of fundamental differences. It was shown in Ref. 2 that the limiting velocity of a 180° DW in an AFM is of exchange order of magnitude ($\sim Ja/\hbar$. where J is the exchange integral) and exceeds considerably the limiting velocity of DW in ferromagnets. At the same time, in the simplest model of a purely uniaxial AFM without allowance for the magnetic dipole interaction, considered by I. Bar'yakhtar and the author,[2] a 90° interphase DW cannot move at all, i.e., its limiting velocity is zero.

This result is understandable, since a 90° DW separates the phases $\Phi_{\parallel}$ and $\Phi_{\perp}$, in which we have respectively $\mathbf{m}=0$ and $\mathbf{m}=m_0\mathbf{n}\neq 0$, where $\mathbf{n}$ is a unit vector along the chosen axis of the crystal (the z axis). Consequently, when the DW moves a change should take place in the value of the z-projection of the total magnetization $I_z=\int m_z dx$, namely $I_z=VM_0$.[3] At the same time, I_z commutes with the Hamiltonian of the uniaxial AFM without allowance for the magnetic dipole interaction or the anisotropy in the basal plane [see (3)], i.e., $I_z=0$, which leads to the condition $V=0$ for a 90° DW. Thus, to study the motion of a 90° DW it is necessary to go outside the framework of the model of Ref. 2.

In the present paper we consider the dynamics of a 90° DW in a uniaxial AFM with account taken of the magnetic dipole interaction. We obtain a solution that describes a moving DW, and an expression for the limiting velocity of the DW. We investigate the study-state motion of the DW under the influence of an external magnetic field with account taken of the relaxation. We obtain more complicated solutions, which describe the interaction of the 90° DW, and also dynamic antiferromagnetic solitons.

1. EFFECTIVE EQUATIONS OF THE MAGNETIZATION DYNAMICS

We consider an AFM with two equivalent sublattices whose magnetizations will be designated $\mathbf{M}_1$ and $\mathbf{M}_2$, $|\mathbf{M}_1|=|\mathbf{M}_2|=M_0$. It is convenient to describe the nonlinear synamics of the AFM in terms of the normalized vectors of antiferromagnetism l and magnetization m:

$$\mathbf{l}=(\mathbf{M}_1-\mathbf{M}_2)/2M_0, \quad \mathbf{m}=(\mathbf{M}_1+\mathbf{M}_2)/2M_0, \tag{1}$$

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