The temperature dependence of the magnetocrystalline anisotropy of chromium: a curve with two extrema

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(Submitted 8 August 1988)
Zh. Eksp. Teor. Fiz. **95**, 1784–1795 (May 1989)

The response of the incommensurate modulated magnetic structure in the AF_1 phase of chromium to changes in temperature and to other probes has been investigated using various methods, including neutron diffraction; the results of these investigations are analyzed and compared. It is shown that anomalies in the temperature dependences of various properties of this system are due to the paradoxical behavior of temperature-induced rearrangements of the magnetic structure's domain texture. These various rearrangement processes are investigated one by one in terms of the crystal's-free energy decrease due to the surmounting of potential barriers associated with magnetocrystalline anisotropy (MCA) forces by forces due to elastic stresses in the crystal lattice. A generalization of the MCA concept is proposed for modulated structures of the static wave type: the MCA energy of these structures is described to first approximation as a sum of three parts, corresponding to a part which fixes the spin directions, one which fixes the direction of the spin-modulation vector, and one which fixes the mutual orientation of these two vectors in the lattice. It is suggested that the curve which describes the temperature dependence of the MCA energy has two extrema; using this hypothesis, it is possible to explain all the anomalous effects.

The incommensurate ordering of atomic magnetic moments in chromium, which was discovered in 1958 (Refs. 1, 2), further lowers the symmetry of chromium's antiferromagnetic state from that of the body-centered cubic paramagnetic phase; based on this fact, one might anticipate the appearance of magnetostrictive distortions of the crystal lattice: rhombohedral in the phase AF_1, and tetragonal in the AF_2 phase. The phase AF_1 of chromium, which lies between the Néel point T_N and the spin-flip point T_F, has three different mutually perpendicular "special" directions which coincide with the principal axes of the crystal lattice. One of these is the polarization axis of the sublattices with atomic spins $+\mathbf{S}$ and $-\mathbf{S}$ (the vertices and centers of the unit cells). The second is an "incommensurability" axis; the wave vector $\mathbf{Q}$ of the sinusoidal spin modulation is oriented along this axis. The third direction is the "free" axis. It is normal to the first two directions, and we denote it by the unit vector $\mathbf{n}$. Because of this "specialization" of the principal axis, the AF_1 phase can contain domains of six types, based on the number of possible combinations of $\mathbf{S}_i$ and Q_j, for $i, j = x,y,z$ but $i \neq j$. The AF_2 phase has only one "special" axis, since the spins are parallel to the incommensurability axis below T_F. Correspondingly, the number of different domains in this phase is reduced to three, where now $i = j = x,y,z$.

The domains in chromium have been observed directly in experiments. Repeated attempts were made to observe the magnetostrictive lattice distortions associated with these domains, but they were not seen until 1969, when Steinitz's group, using hyperfine-structure methods, carried out sufficiently complete and reliable measurements to seen them.[3] These authors confirmed the assertions made above, and also added several fundamentally new assertions of their own. The most informative result reported in Ref. 3 from the standpoint of this article is clearly the curve $\epsilon_Q(T)$ which describes the temperature dependence of the difference between the lattice parameters along the incommensurability and free axes. We note here only two of its distinctive features: the curve in the AF_1 phase interval passes through a maximum and through a point where it changes sign. The fact that these points of the curve coincide with special points at which the behavior of the physical properties of chromium becomes anomalous—a behavior which has not been explained for twenty years—has motivated the formulation of a novel hypothesis: that the temperature dependence of the magnetocrystalline anisotropy energy has two extrema.

This article is devoted to an analysis of the causes of the anomalous temperature effects in chromium. It will be shown here that all the anomalies of the AF_1 phase acquire a regular character when viewed in the light of the hypothesis described above. This is clearly rather convincing evidence that the double-extremum character of the temperature dependence of the magnetocrystalline anisotropy (MCA) energy is a real and not just hypothetical phenomenon.

Crystals with modulated magnetic structure (called either "off-register" or "incommensurate," because in the majority of these systems the magnetic superperiod varies smoothly with temperature and passes through a great many values which are not integer multiples of the period of the crystal lattice), and the distinctive physical properties characteristic of such crystals, have attracted the unwavering interest of many researchers. Theoretical progress in this area, both at the phenomenological and the quantum-mechanical levels, has been reviewed both by Izyumov[4] and Kulikov and Tugeshev[5]; it is clear from these reviews how much attention both experimenters and theorists have devoted to chromium. As a result, the distinctive features of chromium—the appearance of a spin modulation and its secondary effects, i.e., higher harmonic modulations, strain waves in the lattice, charge-density modulation, etc.—have all been explained. With regard to the "normal" properties, the situation in the case of chromium is less satisfactory. To describe these properties, a primarily phenomenological approach is used, and as a rule only the narrow temperature region close to the Néel point is analyzed because the features of the temperature variations of the properties far from the Néel point can be interpreted only with difficulty.

Of course, within the phenomenological approach there

0038-5646/89/051029-08$04.00

is always the possibility of explaining the experimental temperature curve entirely in terms of temperature-dependent coefficients of the free-energy expansion coefficients connected with the property under discussion. However, without identifying the microscopic causes of the temperature variation of these coefficients, such an explanation can hardly be considered persuasive. This type of explanation is encountered in one of the fundamental works on chromium by Shimizu,[6] in which he discussed the function $\epsilon_Q(T)$ described in Ref. 3 among other things; although he did not propose to interpret the whole of this function on the basis of fitting with a suitably-chosen value of the coefficient C_1, he did link the change in the value of $\epsilon_Q(T)$ to a change of the sign of C_1. It is difficult, however, to agree even with this, all the more since in the Appendix to Ref. 6, in a microscopic interpretation of the coefficients in the free energy expansion he assumed, obtained the equation $C_1 = -4J_0a^{-1}$, where J_0 is the exchange integral for the undistorted crystal lattice and a is the BCC lattice constant. Now, it is natural to expect here a sign change of C_1 (and therefore of J_0) to lead to the replacement of the antiferromagnetic order of chromium by ferromagnetic order, or to other fundamental changes in the spin structure. However, no sign changes in the latter are seen in experiments at the temperature in question, except for the possibility of a change in the domain texture of the spin structure, about which more will be said in this article.

1. MCA ENERGIES IN INCOMMENSURATE MAGNETIC STRUCTURES

Let us recall that the appearance of spontaneous magnetization in crystals is caused by the Coulomb exchange interaction; however, this interaction implies an equiprobable orientation of the magnetism vector along any of the crystallographic axes,[7] which in practice is never observed. The concept of MCA energy was introduced in order to explain the preferred orientation of the spontaneous magnetization which exists in crystals. Experimentally, the MCA energy is determined by measuring the amount of the work which must be expended to rotate a single-crystal sample in a saturating magnetic field from the easiest magnetization axis to most "difficult" axis.[8]

In the case of incommensurate magnetic structures, we add to the usual vector element of the magnetic symmetry, i.e., to the spin direction, a second element—the direction of its incommensurate ordering. The orientation of this second element for any type of incommensurate structure also has a "preference" to become aligned with a definite crystallographic axis. Experiment shows that this alignment is stronger than that of the spin direction, i.e., it is usually more difficult to vary the orientation $\mathbf{Q}$ than to change $\mathbf{S}$. This fact naturally makes it necessary to broaden the concept of the MCA energy in order to treat incommensurate structures.

For structures of the static-wave type such as are encountered in chromium, the MCA energy can be represented to first approximation as a sum of three terms: one term serves to fix the spin orientation in the crystal lattice (MCA1), another fixes the orientation of $\mathbf{Q}$ (MCA2), and the third fixes their mutual orientation (MCA3). Based on their analysis of the results of various experiments, the authors of Ref. 9 made certain qualitative estimates of the energy barriers for MCA1, MCA2, and MCA3 in chromium. From these estimates it follows that the MCA1 barrier in its pure form can be observed only in the plane perpendicular to $\mathbf{Q}$, where its profile possesses fourfold symmetry with a smooth transition from a value of zero along the principal axes to a maximum along the diagonals. It is likely that the process of spin rotation is reversible: to the accuracy of neutron-diffraction experiments no dissipation of energy is observed.

At room temperature, the MCA2 barrier is higher than MCA1. It has a profile of rectangular type with symmetry which is also fourfold in cross sections passing through the principal axes. Therefore the direction $\mathbf{Q}$ is rigidly fixed along the [100], [010] or [001] directions. The rotational phase-slip of the vector $\mathbf{Q}$ is clearly an irreversible process, associated with markedly hysteretic phenomena indicating dissipation of energy.

The profile of barrier MCA3 possesses twofold symmetry; however, in the AF_1 phase, after a small jump from a value of zero it increases smoothly to a maximum as the angle between $\mathbf{S}$ and $\mathbf{Q}$ decreases, while in AF_2 it has a rectangular form. The height of the MCA3 barrier in both cases exceeds that of MCA2.

Whereas the MCA1 energy is easily identified with the usual energy of the natural magnetic anisotropy of the undeformed crystal, which in turn is a consequence of spin-spin, spin-orbit, and orbit-lattice interactions, it is not possible to have even a qualitatively complete understanding of the nature of the other two terms either at the microscopic or the phenomenological level; this applies especially to the MCA2 energy. We can only assume, by analogy with the usual total MCA energy, that terms corresponding to elastic strain energy of the lattice in the absence of magnetic order, and to magnetostructive strain energy in the crystal latice, must be present in the second and third MCA components as well.

The above-mentioned analogy is taken into account in all the phenomenological variants of the free energy of chromium known to us; therefore the majority of the terms in these variants are components of the MCA energy. The number and specific form of the terms in each variant is determined by the model used by the author, and by the range of problems to be solved. However, analysis shows that none of these variants admit the possibility of a change in orientation of $\mathbf{Q}$ from one coordinate axis to the other; correspondingly, none of them contain terms which could describe the MCA2 energy.

However, the latest and most accurate phenomenological theory of chromium proposed by Walker[10] does admit variations of the orientation of $\mathbf{Q}$ in the original expression of the free energy, although the allowed variations are completely insignificant. Just as Shimizu[6] had done in the final analysis, Walker[10] used the vector $\boldsymbol{\delta} = (1/2)\mathbf{G}_0 - \mathbf{Q}$, where $\mathbf{G}_0$ is the smallest vector of the reciprocal lattice in the $\mathbf{Q}$ direction, as a small parameter. However, Zhimizu[6] originally took the orientation of $\boldsymbol{\delta}$ to be parallel to $\mathbf{G}_0$ in keeping with the neutron-diffraction experiments (the magnitude of $\boldsymbol{\delta}$ also changes with temperature and on the average equals $(1/25)|\mathbf{Q}|$). Walker,[10] however, using his original expression of the free energy, initially finds equilibrium expressions for the projections $\boldsymbol{\delta}$ on all the coordinate axes, and then shows that terms containing the projection $\boldsymbol{\delta}$ perpendicular to $\mathbf{G}_0$ can be neglected within the approximations he uses. Hence, Walker was able formally to arrive at an important conclusion which agrees with the results of experiment.

However, Walker's theory is valid only for $|\boldsymbol{\delta}| \ll |\mathbf{Q}|$, and therefore it allows no estimate to be made of the order of magnitude of the energy barrier which fixes the direction of $\mathbf{Q}$, nor does it allow inferences about the nature of this barrier. In order to make such estimates and inferences, the theory, descriptively speaking, must allow virtual values of $\boldsymbol{\delta}$ no smaller than $(3/4)|\mathbf{Q}|$; to be precise, no smaller than $2|\mathbf{Q}|\sin(\pi/8)$.

2. ANOMALIES IN THE PROPERTIES OF CHROMIUM AS A CONSEQUENCE OF DOMAIN-REARRANGEMENT ANOMALIES

Experimentally, the anomalous behavior of the physical properties of chromium alluded to above are manifested in their non-monotonic temperature dependence above 230 K. An especially striking and detailed anomaly of this kind is observed in the intensity curves for magnetic neutron diffraction scattering. For this reason, neutron diffraction experiments on this system are found to be much richer in information than experiments which measure the characteristics of other properties, e.g., elasticity, internal friction,[11] electrical resistivity,[12] etc.

Since neutron scattering is a method which is uniquely sensitive to the magnetic structure of materials, it may be assumed that the anomalies are related to changes in this structure. From this same fact there follows the comparably low information content of the other properties, which are less sensitive to the structure.

In order to clarify the nature of the change in magnetic structure leading to the anomalous temperature behavior of the neutron reflection intensities, a very laborious "closed" experiment was performed.[13] Usually, in neutron-diffraction experiments with cooled or heated samples, measurements are limited to those reflections which do not require readjustment of the crystal under study. In Ref. 13, all the magnetic "satellites" of the neutron reflections were measured (these occupy in the reciprocal space of the crystal positions which are shifted relative to the main line by the vector $\pm\mathbf{Q}$) near the type (100) in which nuclear reflections of the neutrons with half-wave supersites were used as "monitor" reflections. Special measures were taken to eliminate hysteresis and other distortions of the results in order to ensure that a fully adequate diffraction pattern was reconstructed at each temperature point for which each reflection corresponded to one and the same magnetic state. Among other things, this leads to a more than threefold increase in the volume of measurements due to repeated reproduction of the full temperature curves for each reflection.

In Ref. 13 it was shown that the overall total intensity of magnetic neutron scattering, and consequently the magnetization of the spin sublattices, grows monotonically and without anomalies with temperature. Only half the local intensities of the spatial distribution of magnetic scattering undergo anomalous variation. The results of Ref. 14 show that these variations are related to changes in the domain texture of the magnetic structure of chromium, i.e., to the spontaneous redistribution of the total sample volume between domains of different polarizations and modulations ($\mathbf{S}_i$ and $\mathbf{Q}_j$), above all between domains with different modulations $\mathbf{Q}_j$. In this case, numerical analysis of the data says that the fraction of the sample volume occupied by domains of various modulations apparently varies in the normal way, despite the clearly anomalous behavior of half of all the reflections of the diffraction pattern. The volume V_x occupied by domains with modulation $\mathbf{Q}_x$, which originally (i.e., in the creation of the ordered structure at the Néel point) amounted to the largest fraction—48.5%—of the overall volume of the crystal, grows monotonically as the temperature falls, while the volumes V_y and V_z occupied by the modulations $\mathbf{Q}_y$ and $\mathbf{Q}_z$ decrease monotonically. The anomalous behavior is manifest only in the rate of domain rearrangement.

Nonmonotonic changes in these rates are more clearly revealed by decreasing volumes; however, as a consequence an increasing volume behaves this way as well—the change in its growth rate is less clear, but is also nonmonotonic. In the samples we investigated all the original volume fractions were found to be significantly different: whereas the original fraction of the growing volume exceeded by roughly a factor of two the total decreasing volume, the original decreasing fractions also differed from one another by roughly a factor of two. The most evident nonmonotonicity in the domain rearrangement was manifest in the rates of change in the volume V_z occupied by domains of modulation $\mathbf{Q}_z$ which originally received the smallest fraction of the crystal below.

We can convince ourselves of this latter fact after comparing the temperature curves for the magnetic reflections and the temperature evolutions of the total intensity of the satellites of the jth family s (see Fig. 4 and Table I in Ref. 13). Using these data it is easy to calculate curves for the evolution of the volume $V_z(T)$ and based on these curves to compute the temperature dependence of the rate of domain rearrangement. In Fig. 1 we present a curve constructed according to the results of such a calculation, which shows the relative decrease in the volume V_z as the temperature varies from T_N to T_F. It is apparent that below room temperature a very steep monotonic growth in the rates of spontaneous rearrangement is observed. After 270 K a less steep but still monotonic falloff begins. Near the lower boundary for the existence of the AF_1 phase the rearrangement ceases, although roughly three quarters of the original value $V_z(T_N)$ remains. In this section of the curve, along with the fact that the rearrangement rate does not increase monotonically with increasing magnetization, there also appears an anomalously high rate of rearrangement over a rather short portion of the temperature range (around 60 degrees) compared to the overall interval of rearrangement (317–110 K). Anoma-

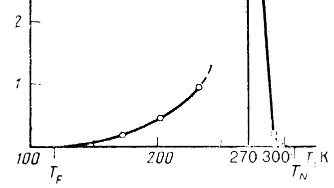

FIG. 1. Variation of the rate of spontaneous rearrangement of an anisotropic domain structure in chromium as the temperature of the crystal decreases through T_N. The points are calculated using the data of Table I of Ref. 13.

lous also is the fact that this short portion accounts for roughly 80% of the overall rearranged volume.

Still more striking anomalies are observed in the domain rearrangement of chromium when the rearrangement occurs not spontaneously, but under the action of external causes, e.g., it is stimulated by application of a strong magnetic field. For fixed temperature, even magnetic fields with the largest attainable intensities (up to hundreds of kOe) applied to single-crystal chromium produce very little change in the ratio of domains of different modulations in the structure of the AF_1 phase. In order for the rearrangement to occur actively, as in the case of spontaneous rearrangement, it is necessary for the temperature to change, i.e., the sample must be cooled; this produces an element of "spontaneous" rearrangement along with the "forced" rearrangement. However, in contrast to the spontaneous rearrangement, the application of an external magnetic field allows us to carry out a rearrangement of directions which is not partial but total. Thus, according to Ref. 15, a field in the range 22–32 kOe applied to the sample parallel to one of the modulation wave vectors $\mathbf{Q}_j$ converts this modulation into a growing one which is independent of the volume fraction, which its domains occupy in the sample at room temperature. Therefore as the sample is cooled to T_F its volume is found to be almost completely (up to 93%) occupied by domains of this modulation. This single-modulation (i.e., single $\mathbf{Q}$) state of the magnetic structure is, as a rule, stably preserved in the crystal after it is raised to room temperature.

In Ref. 15 a more detailed neutron-diffraction investigation of the rearrangement process was also carried out: the authors found a dependence of the degree of rearrangement on the amount of cooling of the sample. It turned out that whereas the purely spontaneous rearrangement continued up to T_F, in the presence of the field it took place much earlier—at 230 K. Further cooling of the sample does not change the degree of rearrangement at 230 K.

This latter work establishes the existence of a lower limit for the range of temperatures which affect rearrangement in a field. Subsequently, Ref. 16 showed that this range is also bounded from above. In order to find the bound, the authors of Ref. 16 did not turn on the magnetic field until the sample was cooled or heated to a definite temperature T_H, i.e., to a "switch-on" temperature for that field. Further cooling to liquid-nitrogen temperature was then carried out in the presence of the magnetic field, after which the degree of rearrangement was determined by neutron diffraction at room temperature. These authors carried out 10 full steps and two abbreviated ones (from $T_H = 330$ K to 308 and 303 K). For each of the 10 value of T_H the degree of rearrangement was also determined for ten values of field—from 3 to 32 kOe.

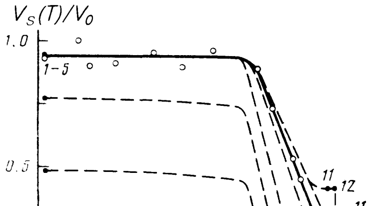

FIG. 2. Dependence of the dynamics of formation of a single-$\mathbf{Q}$ state in chromium in a field $\mathbf{H}\|\mathbf{Q}$ on the temperature T_H, at which the magnetic field $H = 22$ kOe is switched on as the temperature of the crystal is lowered through T_N: 1—$T_H = 295$ K; 2, 11, 12—330 K; 3—303 K; 4—286 K; 5—268 K; 6—258 K; 7—245 K; 8—231 K; 9—213 K; 10—179 K. The points ○ are from Ref. 15, ● from Ref. 16.

Figure 2 shows an extension of the principal results of Refs. 15 and 16 on the boundaries of the smallest range of rearrangement temperatures in a field. One plot traces the results obtained for a field value of 22 kOe which is equally effective in both investigations. The temperature dependence of the degree of rearrangement is given for all T_H in the range from 330 K ($T_N = 309$ K) to 179 K. The degree of rearrangement is given as a ratio of the volume $V_s(T)$ in the sample occupied by domains in the selected modulation to the total sample volume V_0. The original values of $V_s(T_H)$ for all the curves are reduced for clarity to a common level. The solid curve from Ref. 15, and the dashed curve 1 constructed from points given in Ref. 16, coincide with one another throughout. Curves 2–5 also coincide with the solid curve, but only at temperatures below 230 K. For each of the dashed curves in the set 1–12, only two reference points are experimental—the initial and final points[16]; however, the overall course of these curves was constructed based on natural conclusions which follow from the mutual positions of all the reference points and the course of the solid line, which is drawn entirely through experimental points.

Taken as a whole, the curves in Fig. 2 demonstrate strikingly that decreasing the value of T_H down to 268 K does not affect the degree of rearrangement attained in a field of 22 kOe, and that a further decrease in T_H sharply decreases it—already it has falled by 25% at $T = 258$ K. Based on this, we are confident in assuming that 270 K is the upper bound of the smallest range of temperatures which affect rearrangment in a field, from which we conclude that this range is 40 degrees in all, i.e., from 270 to 230 K. Nevertheless, temperatures within this short interval are found to be capable of inducing such high rearrangement rates that the reconstruction of the domain structure in a field attains its maximum possible value, i.e., is virtually complete.

3. ENERGETIC BASIS FOR REARRANGEMENT AND STABILITY OF CHROMIUM DOMAIN STRUCTURES. REARRANGEMENT PARADOXES

The analysis of the causes of spontaneous rearrangement of the chromium domain structure presented in Ref. 17 shows that its source is magnetostrictive distortion of the crystal lattice, and the motive force is the variation of the degree of distortion with temperature. In that paper the authors investigated two models of the domain structure: a regular chainlike structure and a regular bulk structure. For real and hypothetical variants of the chain model and for the regular bulk model expressions were obtained for the tensor of the elastic strains which appear in the crystal lattice as a consequence of magnetostriction, and for the tensor corresponding to the overall case of a real domain structure based on these expressions.

Since we do not include strains in the lattice of a multidomain crystal other than those caused by magnetostriction, the principal axes of the strain tensor coincide with the principal axes of the lattice throughout the crystal; the strain is uniformly distributed within the boundaries of each domain, and has the same value within the domains of a given modulation. Therefore the strain tensor of the crystal can be described in the form of a table, each of whose rows is made up of the three principal value of the tensor for a single-modulation domain[17]:

$$\varepsilon_{11}=-\frac{\epsilon_1(L_{22}L_{32}+L_{23}L_{33})}{L_{22}L_{32}+L_{23}L_{33}+L_{21}L_{31}},\quad \varepsilon_{21}=\frac{\epsilon_2 L_{12}L_{32}}{L_{11}L_{31}+L_{12}L_{32}+L_{13}L_{33}},$$

$$\varepsilon_{31}=\frac{\epsilon_3 L_{13}L_{23}}{L_{11}L_{21}+L_{12}L_{22}+L_{13}L_{23}};$$

$$\varepsilon_{12}=\frac{\epsilon_1 L_{21}L_{31}}{L_{22}L_{32}+L_{23}L_{33}+L_{21}L_{31}},\quad \varepsilon_{22}=-\frac{\epsilon_2(L_{11}L_{31}+L_{13}L_{33})}{L_{11}L_{31}+L_{12}L_{32}+L_{13}L_{33}},$$

$$\varepsilon_{32}=\frac{\epsilon_3 L_{13}L_{23}}{L_{11}L_{21}+L_{12}L_{22}+L_{13}L_{23}};$$

$$\varepsilon_{13}=\frac{\epsilon_1 L_{21}L_{31}}{L_{22}L_{32}+L_{23}L_{33}+L_{21}L_{31}},\quad \varepsilon_{23}=\frac{\epsilon_2 L_{12}L_{32}}{L_{11}L_{31}+L_{12}L_{32}+L_{13}L_{33}},$$

$$\varepsilon_{33}=-\frac{\epsilon_3(L_{11}L_{21}+L_{12}L_{22})}{L_{11}L_{21}+L_{12}L_{22}+L_{13}L_{23}}.$$

Here the quantities L_{ij} are the average values of the dimensions in the ith direction of the sum of domains of the jth modulation. Assuming that the magnetostriction along **S** and the Poisson coefficient μ both equal to zero, we can set $\epsilon_1=\epsilon_2=\epsilon_3=\epsilon_Q$. Taking into account the Poisson compression, we obtain

$$\epsilon_1=\epsilon_Q-\mu(\varepsilon_{21}+\varepsilon_{31});\quad \epsilon_2=\epsilon_Q-\mu(\varepsilon_{12}+\varepsilon_{32});$$

$$\epsilon_3=\epsilon_Q-\mu(\varepsilon_{13}+\varepsilon_{23}).$$

This somewhat lowers the advantage in elastic energy of a domain rearrangement while not changing the rearrangement process qualitatively. Including the magneostriction along **S** also does not affect the qualitative course of the change in elastic energy as the L_{ij} vary; however, it makes the expression for ε_{ij} even more cumbersome.

It has been shown that the elastic energy of magnetostrictive distortions of the chromium lattice reaches a maximum when the volume of the crystal is distributed in equal portions among domains with all three modulations, i.e., for the case of an isotropic domains structure. As the domain anisotropy grows, the elastic energy decreases monotonically, decreasing by 25% for structures with domains of two modulations and going to zero for the single-**Q** state with one polarization.

Hence, it has been shown that the maximum anisotropy of the domain structure corresponds to a minimum in the crystal elastic energy. Nevertheless, in practice the spontaneous appearance of single-**Q** domain structures has never been observed in a chromium crystal. The reason is that even very perfect crystals do not have an ideal lattice; it is distorted by a certain quantity of defects. Domains of various modulations also nucleate on variously oriented distortions; these domains correct the lattice defects with their rhombic (at first close to tetragonal) distortions and thereby decrease the elastic energy of the original strains. However, the fields of the defects are local and rather rapidly relaxing, while the intensity of magnetostriction, as shown in Ref. 17, is uniformly distributed within the limits of each domain, and therefore encompasses the volume of the crystal. Because of this, as the temperature decreases, along with the growth in magnetostriction the elastic energy of a sufficiently perfect crystal, after passing through a minimum not far from the Néel point, begins to fall. For crystals with an isotropic and uniform distribution of defects, which ensures an isotropic domain structure, this growth, as we see, is a maximum. Nevertheless, the isotropic structure does not undergo a spontaneous rearrangement. The stability of this structure, which is in equilibrium, but is in principle unstable, is due to potential barriers associated with the MCA forces. Being at the beginning vanishingly small, they nevertheless ensure the structure's stability because overcoming them in the neighborhood of the maximum even for significant variations in the domain structure would lead to only a vanishingly small energy advantage which is insufficient to compensate for the irreversible loss of energy in rearrangement.

In the case of an originally anisotropic domain structure the situation with regard to energy balance changes significantly. Its elastic energy also grows proportional to the squared magnetostriction, although more slowly than in the isotropic case, because it is not initially at a maximum. However, the latter circumstance ensures a finite energy advantage for some finite change in the domain structure corresponding to the value of the irreversible energy loss upon rearrangement (with the additional condition that the level of elastic energy exceeds the MCA barrier).

As we see, in order to understand the observed features of the dynamics of spontaneous rearrangement of the domain structure of chromium, it is important to know the temperature variation of the magnetostriction and MCA energy. The change in magnetostriction for chromium, as was noted above, has been accurately measured,[3] while the MCA energy in incommensurate structures has not been investigated at all. If we approach this from the theoretical viewpoints marshalled to describe the usual MCA energy, then its value should increase monotonically from zero at the Néel point to an upper limit at 0 K together with spontaneous magnetization of the spin sublattices (proportional to third or higher power of the magnetization).[7] However, in this case the spontaneous-rearrangement dynamics presented in Fig. 1 can be explained to some degree only at small distances from the Néel point, where, according to the curve of Steinitz *et al.*,[3] the change in the primary components of the magnetostriction, i.e., those along the direction of the modulation, can still be considered linear.

Below room temperature the rearrangement process becomes anomalous. It follows from Fig. 1 that the rate of rearrangement below 290 K grows rapidly, and by 230 K the rearrangement turns out to be 80% complete. However, at just these temperatures the MCA energy which hinders the rearrangement increases sharply, while the elastic strain energy which brings about the rearrangement slows its growth, which for 270 K completely ceases. Later the elastic energy begins to decrease, since the magnetostriction along **Q** passes through a maximum at 270 K, while at 230 K it changes sign. On the face of it, a paradox—forces which cause rearrangement decrease, while the latter is enhanced!

The situation in the case of "strong" rearrangement appears at first glance to be better—its admissibility at any temperature causes no doubt. First of all, when a sufficiently high magnetic field is applied, it always can, in principle, impart to the crystal a rearrangement energy which exceeds the MCA barrier, no matter how increases in the temperature and fields of original ("defect") stresses cause the barrier to rise. Secondly, the active component of the magnetostriction for strong rearrangement is found to be lattice distortion not along **Q** but along **S**. Because in antiferromagnets the susceptibility χ_n in directions normal to the spin is always larger than that parallel to the spin χ_S, in domains with unfavorable spin orientations the field forces a change these orientations to orthogonal directions. Consequently, strong stresses in the crystal can only be avoided when its whole volume is occupied by domains with a selected modulation, i.e., parallel to the field. Therefore the basic rearrangement dynamics in the strong case is also governed by the decrease in elastic energy which accompanies the rearrangement. However, here an additional contribution to the forces causing the rearrangement is made here by the gain in the purely magnetic energy: as shown by Steinitz *et al.*,[18] the rhombic phase AF_1 of chromium possesses a magnetic susceptibility of just this symmetry (not only $\chi_n > \chi_S$, but also $\chi_Q > \chi_n$).

A moderate but stable linear increase[3] in the absolute value of the magnetostriction along **S** as the temperature varies from T_N to T_F, giving rise to the steady growth of elastic strains necessary for rearrangement, as well as a monotonic growth of the MCA energy, exclude the appearance of any peculiarities in the dynamics of strong rearrangement. Under these conditions we find no explanation either for the unvarying completion of the total rearrangement at 230 K, nor for the existence of an upper bound of the interval of full rearrangement at 270 K (see Fig. 2). Both these points are also perceived as paradoxical.

4. PARADOXES IN THE REARRANGEMENT OF DOMAINS AS A CONSEQUENCE OF TWO TEMPERATURE EXTREMA IN THE MCA ENERGY

All the phenomena listed above were for us fundamental in overcoming the inertia of the classical viewpoint, in which the temperature dependence of the MCA energy is treated as monotonic. In this case, an important role was played by a generalization of the MCA energy to include incommensurate magnetic structures and the identification of the part especially responsible for the rearrangement dynamics. This part—MCA2, which fixes the direction of the spin modulation in the crystal—was taken to be proportional to the square of the magnetostriction of the lattice along the direction of the modulation, i.e., to the elastic strain energy caused by this component of the magnetostriction. This assumption caused the appearance of two extrema on the MCA2 energy curve in the energy curve in the interval of the AF_1 phase: a minimum at 230 K and a maximum at 270 K. Both of them, by virtue of additivity, extend also to the overall temperature dependence of the total MCA energy of chromium.

The dash-dot curve in Fig. 3, which coincides below 230 K and above 295 K with the solid curve, was calculated using a constant proportionality coefficient 0.3 according to the curve from the work of Steinitz *et al.*[3] for magnetostriction along the incommensurate axis. The solid curve corresponds to the temperature dependence of the MCA2 energy barrier. It was calculated in the same manner as the dash-dot curve, but with a proportionality coefficient 0.25 on the interval 230–295 K. The dashed curve shows the temperature variation of the usual MCA energy.1)

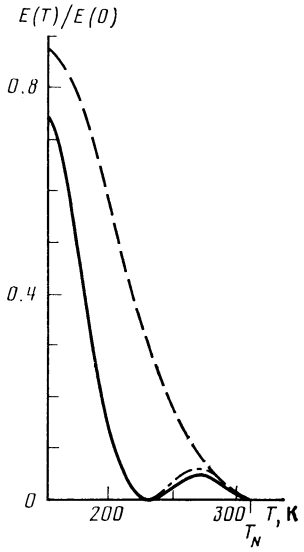

FIG. 3. Temperature dependence of the energy of magnetostrictive lattice strains along **Q** (dash-dot curve), the MCA2 energy (solid curve) and the usual MCA energy (dashed curve).

In light of the energy dependence assumed for MCA2, the experimentally observed features of the rearrangement dynamics of the domain structure cease to be anomalous.

First of all, the spontaneous rearrangement of the structure is now perceived to be a normal phenomenon not only near the Néel point but for any temperature, because the energy barrier now nowhere exceeds the energy of the elastic strains which cause the rearrangement.

Secondly, we now have an explanation for the increase in activity of the spontaneous rearrangement on the portion 295–230 K. The value of the energy barrier on this portion is a smaller fraction of the magnetostriction energy than for other temperatures. This also resolves the paradox: although the forces ensuring rearrangement slow their growth and then decrease, the forces which resist rearrangement slow their growth and decrease even faster.

Thirdly, it becomes clear why the rearrangement of the structure in a magnetic field is complete at 230 K. The point is that the magnetic-field energy received by the crystal (which is proportional to the spontaneous magnetization) attains large values in this region of temperatures, while the energy barrier resisting it, in contrast becomes minimal. At 230 K it simply disappears. Therefore it is natural to expect the maximum possible degree of rearrangment of the structure just before this point is reached, especially as any additional energy received by the crystal increases more slowly than the rise of the MCA2 barrier.

Finally, in order to understand the fourth feature, i.e., the existence of an upper limit of 270 K for the interval of full rearrangement, it is necessary to recall that strong rearrangement is ineffective at fixed temperature: it takes temperature-induced changes in the field of elastic strains of the crystal to activate it. It is also necessary to take it into account that the increase in the interaction energy of the magnetic structure of chromium with the field is proportional to the square of H; the degree of rearrangement grows correspondingly, only in this case the total gain in free energy of the crystal due to rearrangement has a limit to which it tends with increasing H apparently asymptotically as the degree of rearrangement goes to unity with increasing H. In this case a given value of H produces a given degree of rearrangement. From this we can draw the following conclusion: by dropping the upper boundary of the working temperature range below 270 K, we cancel out the most active part of this range, i.e., the part in which the decrease in the MCA2 barrier occurs. At the same time, we cancel out the "dynamic reserve", i.e., the total change in the elastic-stress field; this in turn prevents the crystal from responding to the largest attainable dose of conversion energy, which consequently prevents it from undergoing the largest possible degree of rearrangment for a given value of H.

CONCLUSION

While we were preparing this article to send to the editors of JETP, there appeared in the March issue of the journal a paper by Fawcett *et al.*,[19] in which the authors reported that they tried to create a single-**Q** state in single-crystal chromium by the method used in Refs. 15 and 16 (i.e., by cooling the sample in a magnetic field at liquid-nitrogen temperatures) and obtained a negative result. The authors of Ref. 19 also concluded that one should not expect any other result, because the method was fundamentally incorrect from the point of view of thermodynamics. However, most likely their lack of success was related to the fact that the crystal under study had a large number of impurity defects, because it was obtained by arc melting. Prolonged annealing only produces a rather uniform distribution of defects; the small anisotropy of the domain structure attests to this.

The crystals of chromium iodide investigated in Refs. 15 and 16 were of very high quality; however, as we showed above, it is just this high level of perfection which allows the observed peculiarities of the domain structure to appear, whereas the rearrangement of the domains in less perfect crystals having the same energetics should somehow differ in the dynamics of the rearrangement processes. The investigation of these processes requires additional experiments and an independent analysis; however, certain estimates can be made immediately. In order to make these estimates, it is necessary to take into acount the fact that the value of the relative strain of the lattice around the defects can amount to several tenths of a percent or even several percent, while the magnetostriction is three to four orders of magnitude smaller ($4\cdot 10^{-4}\%$) even near 270 K. Therefore, although the former does decrease with distance while the latter does not change, as the number of defects grows in the crystal a point is inevitably reached when the elastic energy of the defect-induced distortions becomes comparable to the magnetostriction energy not only near T_N, but also near 270 K, and subsequently greatly exceeds it. In such crystals the dymamics of the spontaneous rearrangement has no directivity and is fully subordinate to the original distortion of the lattice; its action can only decrease the crystal's elastic energy, and moreover may even be blocked by the MCA barriers in the case of an unfavorable distribution of impurities.

The same thing applies to rearrangement in a magnetic field. Under these conditions the most efficient way to create a single-**Q** state is by cooling the sample through T_N (see the jump in curves 11 and 12 in Fig. 2), because magnetostriction is practically absent near T_N, while at T_N the value of the magnetization immediately acquires a finite value (i.e., a first-order phase transition occurs). Therefore, the decisive factor in forming a domain structure in a field turns out to be the domains' magnetic energy, i.e., the relation $\chi_{\rm Q} > \chi_{\rm n} > \chi_{\rm S}$. If the value of H is insufficient to give rise to a completely single-**Q** state, a further drop in temperature of the crystal accompanied by the formation of a dominant domain structure should improve the onset of the single-**Q** state when this leads to a gain in free energy.

The considered cases of creation of a single-**Q** state in insufficiently perfect crystals (including total blocking of the rearrangement in crystals cooled in a magnetic field which is turned out at room temperature) was observed by Golovkin *et al.*[20] Somewhat similar results were apparently obtained obtained by Fawcett *et al.*[19]: a field of 120 kOe was sufficient while 25 kOe was insufficient to create a single-**Q** state. Several differences in the results of experiments with a field of 25 kOe could have been a consequence of fluctuations in the anisotropy of the domain structure; such effects were observed by Werner *et al.*[21] and Golovkin *et al.*[22] even without the action of the field, simply as a result of the thermal cycling.

As for destruction of a single-**Q** state below T_F by a field because of the fact that $\chi_{\rm n} > \chi_{{\rm S}\|{\rm Q}}$, this was shown to be impossible in the paper by Steinitz *et al.*,[3] which the authors of Ref. 19 cited. In fact, even a partially single-**Q** state can be destroyed below T_F only by a field $\mathbf{H}\|\mathbf{Q}$ which far exceeds the field $\mathbf{H}_c$ which causes this state. Thus, after the action of a field $H_c = 35$ kOe, which produces an 80% single-**Q** state, it was necessary to apply $H = 45$ kOe before the first signs of a change appeared in the single-**Q** state at 100 K, and only at $H = 100$ kOe did the single-**Q** fraction decrease to 54%; however, even in this field the crystal did not pass into the alternative two-**Q** state which is favored in terms of magnetic-field energy. From this we can conclude that a field of the same magnitude as that in which the single-**Q** state is created does not provide below T_F enough magnetic-energy gain to surmount the MCA barrier and compensate for the irreversible energy loss upon rearrangement.

1) We associate thus the height of the MCA2 barrier (which, like the MCA1 barrier, can be observed in its pure form only in the plane perpendicular to **S**) directly with the magnitude of the magnetostrictive distortions of the crystal-lattice parameter in the direction of **Q**. For the case $\mathbf{Q} \neq \mathbf{G}_0/2$, in analogy with $\mathbf{Q} = \mathbf{G}_0/2$, we associate its rectangular profile—i.e., the strict orientation of **Q** parallel to the principal axes—with the symmetry (including translational) of the direct and reciprocal spaces of a crystal that has a lattice corresponding to the cases under discussion.

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Translated by Frank J. Crowne