

Domain walls in Jahn-Teller crystals with competing structural and magnetic ordering

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(Submitted 4 March 1986)

Zh. Eksp. Teor. Fiz. **91**, 1444–1453 (October 1986)

When ordering processes compete in the bulk of a crystal, the one with the lowest energy predominates. Attention is drawn to the fact that if a stronger ordering of the domain structure type is inhomogeneous, a weaker ordering can appear within the domain walls. In particular, in a structural transition in a Jahn-Teller crystal a magnetic ordering may exist in the domain walls that separate regions with opposite displacements. When an allowance is made for the dipole-dipole interaction, it is found that the effective dimensionality of the space for fluctuations near the point of appearance of ferromagnetism localized within a wall is 2.5, as for any other planar ferromagnet with an easy axis in the plane of the sample.

1. INTRODUCTION

Crystals with a strong spin-orbit interaction exhibit a significant correlation of magnetic and elastic properties. This correlation (magnetoelastic interaction) is particularly strong in compounds of rare-earth ions in which the spin-orbit coupling is strong because of the proximity of the f shell to the nucleus, whereas the interaction of electrons with the environment is weak. For this reason a $(2J + 1)$ -fold degenerate ground multiplet of a free ion with the momentum J splits weakly in crystal fields, so that the ground state remains frequently degenerate or pseudodegenerate. In these situations it has been found that structural phase transitions with temperatures T_c of the order of $10 K$ occur because of the cooperative Jahn-Teller (JT) effect.¹⁻³ In the case of nonconducting compounds of rare-earth ions the exchange effects are slight (they are comparable with the direct magnetic dipole effects) because of the weak covalence, so that the temperatures of magnetic transitions T_m may be of the order of T_c . The proximity of T_c and T_m , and the strong magnetoelastic coupling results in an effective interaction between the JT structural and magnetic ordering mechanisms: they either enhance or weaken one another. In the case of competition, the coexistence of these two types of ordering is generally impossible and only one ordering is realized in the bulk of a crystal: it is that which has the lowest free energy. However, the proximity of the system to the second "suppressed" phase transition is manifested by a series of characteristic physical properties. For example, experimental investigations have shown that when a structural transition takes place, its temperature is lowered by a magnetic field, the magnetic susceptibility of a structurally ordered is independent of temperature, etc.; the behavior of such homogeneously ordered systems has been investigated in considerable detail.^{1,2}

We would like to draw attention to a situation in which a stronger ordering is inhomogeneous and produces a domain structure. In the case of a stronger magnetic transition in a fairly large sample such a state corresponds to the minimum of the free energy which includes the energy of the

stray field. If the structural transition is stronger, then a domain structure is usually still present in a sample because of stresses in the system or because of other factors. In either case the main ordering process is less effective in domain walls where a second weaker ordering may appear.¹⁾ In other words, a phase transition in domain walls may result in a weaker ordering of the type which is suppressed in the rest of the crystal. Clearly, the most interesting case from the experimental point of view is a magnetic transition between structural domains. The modern technique of magnetic measurements should be sufficient to detect such wall-localized ferromagnetism.

2. COMPETITION BETWEEN MAGNETIC AND STRUCTURAL ORDERING IN A CRYSTAL WITH JAHN-TELLER IONS

We shall consider the interaction of the magnetic ordering with the ordering of JT distortions in the case of a crystal (such as a tetragonal crystal of $TmVO_4$) in which the ground state of ions in the rare-earth sublattice is a non-Kramers doublet separated by a large gap (of the order of 100 cm^{-1}) from excited states. Low-temperature properties of compounds with rare-earth ions of this type are determined by the structure of the lowest electronic levels of these ions in the crystal electric field (in the case under discussion there are two such levels). A structure of this type can be described conveniently by the pseudospin operator \hat{S}_m of an ion located at a site with the coordinate m . It is introduced in such a way that $(2S + 1)$ is the number of electronic states allowed for in the calculations. The electron operators included in the Hamiltonian are then expressed in terms of the components $\hat{S}_{m\alpha}$ ($\alpha = x, y, z$), so that the effective Hamiltonian has the usual form of the spin Hamiltonian. It contains the interactions of spins with one another (of the $\hat{S}_{n\alpha} \hat{S}_{m\alpha}$ type), with the magnetic field $H_\alpha \hat{S}_{m\alpha}$, initial splitting ($\hat{S}_{m\alpha}^2$), etc. It is important to note that, in contrast to the true spin, the different pseudospin components may (and usually do) correspond to quantities of very different physical nature. Thus, it is easy to show that the basis of functions of a non-Kramers doublet ($S = 1/2$) of the group D_4 , which we

shall discuss as an example, has matrix elements J_x and J_y of the total momentum \mathbf{J} which vanish. Therefore, the interaction with a magnetic field, usually written in the form $g\mu_B H \mathbf{J}$ (for the complete basis of the spin-orbit multiplet) is $g'\mu_B H_z \hat{S}_z$ for the basis of functions of a non-Kramers doublet truncated by the crystal field. The eigenstates of \hat{S}_z are therefore states with a specific value of the magnetic moment along the z axis selected by the crystal field. The eigenfunctions \hat{S}_x and \hat{S}_y correspond to states with a specific value of the electron quadrupole moment, and the components \hat{S}_x and \hat{S}_y occur in the operator of the interaction of a non-Kramers doublet with the low-symmetry electrostatic fields of the lattice which appear as a result of displacement of ions in the crystal. Therefore, the electron energy in the field of displacements (which can be, for example, of the B_1 symmetry for the D_4 group of the unit cell of a crystal of the type under discussion) is

$$H_{e,pn} = \sum_{m,\kappa} V_{m\kappa} \delta_{m\kappa} u_\kappa + 1/2 \sum_{\kappa} \lambda_\kappa u_\kappa^2, \quad (1)$$

where u_κ are the displacements of ions and $V_{m\kappa}$ are the parameters of the interaction; the index κ labels displacements corresponding to phonons κ . Eliminating u_κ from the Hamiltonian (1), we obtain the spin part of the electron Hamiltonian describing the intersite interaction³:

$$- \sum_{m,n} B_{mn} \delta_{m\kappa} \delta_{n\kappa}, \quad B_{mn} = \sum_{\kappa} (V_{m\kappa} V_{n\kappa} / \lambda_\kappa).$$

The complete spin Hamiltonian which allows both for the magnetic interactions (of the $\hat{S}_{mz} \hat{S}_{nz}$ type) and for the JT distortion interactions (of the $\hat{S}_{my} \hat{S}_{ny}$ type) becomes

$$H = - \sum_{m,n} (I_{mn} \delta_{mz} \delta_{nz} + B_{mn} \delta_{my} \delta_{ny}). \quad (2)$$

In Eq. (2) we have to separate the long-range contributions. The magnetic dipole interaction makes the following contribution to I_{mn} :

$$I_{mn}^{(d)} = (g'\mu_B)^2 [3(m_z - n_z)^2 - (m - n)^2] / |\mathbf{m} - \mathbf{n}|^5, \quad (3)$$

which is of the same order of magnitude as the contribution to the exchange interaction in compounds of the kind under discussion. This contribution is important when dealing with the inhomogeneous magnetic structures and magnetic fluctuations. The interaction of "spins" with a homogeneous deformation makes a contribution to B_{mn} which is independent of \mathbf{m} and \mathbf{n} (and inversely proportional to the total number of sites in the system), whereas the interaction of spins with displacements corresponding to acoustic vibrations makes a contribution to B_{mn} similar to the magnetic-dipole contribution.¹ All other interactions in Eq. (3) are of the short-range nature with a characteristic scale of the interaction of the order of the interatomic distance. The presence of long-range contributions to Eq. (3) has the effect that fluctuations of the order parameter ($\langle \hat{S}_{my} \rangle$ or $\langle \hat{S}_{mz} \rangle$) are suppressed, which is manifested not by a power-law but by a weaker logarithmic divergence of thermodynamic functions near the critical points.^{4,5} Therefore, three-dimensional or-

dering in this system can be described with high accuracy using the self-consistent field approximation.

We shall now consider the specific case of ordering of the ferromagnetic type on the assumption that $2I = \sum_n I_{mn} > 0$ and $2B = \sum_n B_{mn} > 0$. We shall write down the functional of the free energy of the system as a function of the average values of the spin at a site $S_m = \langle \hat{S}_m \rangle$ (angular brackets denote statistical and quantum-mechanical averaging). This functional is

$$\mathcal{F}\{S_m\} = - \sum_{m,n} (I_{mn} S_{mz} S_{nz} + B_{mn} S_{my} S_{ny}) - T \sum_m \sigma(S_m), \quad (4)$$

where

$$S_m^2 = S_{my}^2 + S_{mz}^2,$$

$$\sigma(x) = - \int_0^x b_{1/2}(y) dy = - \left[\left(\frac{1}{2} + x \right) \ln \left(\frac{1}{2} + x \right) + \left(\frac{1}{2} - x \right) \ln \left(\frac{1}{2} - x \right) \right],$$

and $b_{1/2}(y)$ is the reciprocal of the Brillouin function for the spin 1/2. Expansion of Eq. (4) as a series in terms of the order parameter S_m in the region, where $S_m \ll 1$, gives

$$\mathcal{F}\{S_m\} = - \sum_{m,n} (I_{mn} S_{mz} S_{nz} + B_{mn} S_{my} S_{ny}) + 2T \sum_m \left(S_{mz}^2 + S_{my}^2 + \frac{2}{3} S_{mz}^4 + \frac{2}{3} S_{my}^4 + \frac{4}{3} S_{mz}^2 S_{nz}^2 \right). \quad (5)$$

It is clear from Eq. (5) that the two types of ordering—magnetic ($S_{mz} \neq 0$) and structural ($S_{my} \neq 0$)—compete with one another since the energy of their interaction $4/3TS_{mz}^2 S_{my}^2$ is positive. The competition, described by the free-energy functional of Eqs. (4) or (5), excludes the coexistence of the two types of ordering in the bulk of a crystal, since the homogeneous solution with $S_{mz} \neq 0$ and $S_{my} \neq 0$ has a higher energy than the purely magnetic solution $S_{mz} \neq 0$, $S_{my} = 0$ or the solution with the structural ordering $S_{mz} = 0$, $S_{my} \neq 0$ (the magnetic solution is preferable for energy reasons when $I > B$ and the structural solution when $B > I$). As already pointed out, the coexistent solutions are possible in the case of a domain structure with the stronger type of ordering predominating in the interior of a crystal. We shall assume that the structural transition is stronger.

3. WEAKER ORDERING IN A DOMAIN WALL

We shall describe the properties of an inhomogeneous ordered state inside a domain wall at temperatures close to T_c by expanding the density of the free-energy functional as a series in powers of the order parameter and its gradients. This can be done by transforming Eq. (5) from the site representation S_m to a continuous representation $S(\mathbf{r})$. We shall also replace $S_z(\mathbf{r})$ with $m(\mathbf{r})$, representing the average magnetic moment of an ion at a point \mathbf{r} (in units of $2g'\mu_B$), and express $S_y(\mathbf{r})$ in terms of $u(\mathbf{r})$, which is the displacement of an ion at a point \mathbf{r} [in units of $2u(T=0)$], where $u(T=0)$ is the displacement at $T=0$. Then, the free-ener-

gy functional (per ion) becomes

$$\mathcal{F} = \frac{4}{V} \int d\mathbf{r} F\{u(\mathbf{r}), m(\mathbf{r})\}, \quad (6)$$

where

$$F\{u, m\} = \frac{1}{2} \kappa_1 (\nabla u)^2 + \frac{1}{2} \kappa_2 (\nabla m)^2 - \frac{1}{2} A (u^2 + m^2) + \frac{1}{2} K m^2 + \frac{1}{4} D (a^2 + m^2)^2,$$

$$A = T_c - T, \quad T_c = B, \quad K = B - I, \quad D = \frac{4}{3} T,$$

and V is the volume of the system. The density of the free-energy functional F written in this form is identical with that given in Ref. 6, where an analysis is made of the structure of a domain wall in a ferromagnet with the easy-axis anisotropy, and u and m are regarded as the components of the real spin; the quantity u then plays the role of the projection of the spin along the easy magnetization axis and m is the projection perpendicular to this axis. Then $u(\mathbf{r})$ is the displacement which appears at the temperature T_c and $m(\mathbf{r})$ is the magnetization which would have appeared at the temperature $T_m = I = T_c - K$ in the absence of a structural transition (i.e., when $B < I$).

Equation (6) ignores the terms describing the long-range parts of the magnetic dipole interaction and of the interaction of pseudospins at different sites via acoustic vibrations. In the case of inhomogeneous spatial structures the corresponding energies depend on the angles between the axis along which u and m vary and the direction of the magnetization vector (z axis) or the vector representing the polarization of the displacements.¹ The minimum energy corresponds to transverse structures. We shall therefore consider only transverse inhomogeneous structures on the assumption that u and m depend only on x , that the magnetization vector is directed along the z axis, and that the displacement polarization vector lies in the yz plane. For these transverse structures the free-energy functional including the dipole-dipole interaction has a minimum and assumes the form described by Eq. (6).

Within the framework of the functional (6) the homogeneous solutions with $u = \pm u_0$, where $u_0 = (A/D)^{1/2}$, appear below the temperature T_c and in this case there is no magnetization ($m = 0$). Let us now consider the solution for u and m in the case when a domain wall separating regions with opposite directions of the displacements ($-u_0$ and u_0) is present in a crystal. The behavior of the order parameters u and m inside this wall is described by the system of equations

$$\begin{aligned} \kappa_1 \ddot{u} &= -Au + Du(u^2 + m^2), \\ \kappa_2 \ddot{m} &= -(A-K)m + Dm(u^2 + m^2) \end{aligned} \quad (7)$$

with the boundary conditions $u = \pm u_0$, $m = 0$ in the limit $x \rightarrow \pm \infty$ ($\ddot{u} = d^2u/dx^2$). The system of equations (7) is obtained from the condition for the minimum of the functional of Eq. (6). Like this functional, these conditions are valid if

the range of variation of u and m is much greater than the interatomic distance.

The system (7) has two types of solution. Below T_c we have the solution

$$u = u_0 \operatorname{th}[x(A/2\kappa_1)^{1/2}], \quad m = 0. \quad (8)$$

It corresponds to the usual variation of the main (bulk) order parameter (deformation) in a wall and there is no additional (magnetic) ordering. Below the temperature T_{mw} , which will be defined below, a solution with $m \neq 0$ appears in the wall (at temperatures $T < T_{mw}$ this solution corresponds to a lower free energy). The temperature T_{mw} is defined at the point of appearance of a nonzero solution of the equation

$$\kappa_2 \ddot{m} + (A-K)m + m \operatorname{th}^2[x(A/2\kappa_1)^{1/2}] = 0, \quad (9)$$

which is obtained in the second equation in the system (7) by substituting in it the solution for u from Eq. (8) and by neglecting terms of the order of m^3 . Equation (9) has the form of the Schrödinger equation for motion in a potential of the $\cosh^{-2}x$ type, and it can be rewritten in the form

$$\ddot{m} + m \frac{A}{\kappa_2} \operatorname{ch}^{-2}[x(A/2\kappa_1)^{1/2}] = \frac{K}{\kappa_2} m = -Em. \quad (9a)$$

The eigenvalues E_n of Eq. (9a) are given by the formula (see Ref. 7)

$$E_n = -\frac{A}{8\kappa_1} \left[-(1+2n) + \left(1 + \frac{8\kappa_1}{\kappa_2}\right)^{1/2} \right]^2, \quad n = 0 \text{ or } 1. \quad (10)$$

Equating the ground-state energy E_0 to the right-hand side of Eq. (9a), we obtain

$$T_{mw} = T_c - \alpha(T_c - T_m), \quad \alpha = 8\kappa_1\kappa_2^{-1} [(1+8\kappa_1/\kappa_2)^{1/2} - 1]^{-2}. \quad (11)$$

It is clear from Eq. (11) that the magnetic moment appears in a structural wall only when the magnetic interactions are not too weak, because the value of T_{mw} is positive only if $T_m > (1 - \alpha^{-1})T_c$. In fact, the transition to the continuous representation is justified only for $T_c - T_{mw} \ll T_c$, i.e., when $\alpha(T_c - T_m) \ll T_c$. It is clear from Eq. (11) that the conditions for the appearance of the magnetization in a structural wall become easier on increase in the ratio κ_1/κ_2 , but in a typical situation the value of κ_1/κ_2 does not differ too much from T_c/T_m (we can expect $\kappa_1 \propto T_c a^2$ and $\kappa_2 \propto T_m a^2$, where a is the interatomic distance). We then have $\alpha \approx 1$ and the temperature T_{mw} is close to T_m , but the results obtained are valid only if $T_c - T_m \ll T_c$.

We can determine the form of $m(x)$ below T_{mw} by solving the system (7) and retaining terms of higher orders in m . It is shown in Ref. 6 that this system of equations admits an analytic solution if $\kappa_1 = \kappa_2$, which is of the form

$$\begin{aligned} u(x) &= u_0 \operatorname{th}[x(K/\kappa_1)^{1/2}], \\ m(x) &= u_0 (1 - 2K/A)^{1/2} \operatorname{ch}^{-1}[x(K/\kappa_1)^{1/2}]. \end{aligned} \quad (12)$$

It follows from this solution that if $T < T_{mw}$ then the magnetization appears in a domain wall of a ferroelastic and its appearance alters considerably the temperature dependence of the wall thickness L . If $T > T_{mw}$, it follows from Eq. (8) that the wall thickness is $L \approx (2\kappa_1/A)^{1/2} \propto (T_c - T)^{-1/2}$

and that it decreases as a result of cooling. It follows from Eq. (12) that if $T < T_{mw}$, the wall thickness is $L \approx (2\kappa_1/A)^{1/2} \propto (T_c - T)^{-1/2}$ and that it decreases as a result of cooling. It follows from Eq. (12) that if $T < T_{mw}$, the wall thickness is $L_m \approx [\kappa_1/(T_c - T_m)]^{1/2}$ (if $\kappa_1 = \kappa_2$, we find that $T_{mw} = 2T_m - T_c$). Therefore, below T_{mw} the wall thickness ceases to depend on temperature. An analytic solution of Eq. (12) can be obtained only for $\kappa_1 = \kappa_2$, but it should provide a qualitatively correct description of the behavior of the system for all values of α of the order of unity.

The results given above are valid only in the range of temperatures near T_c . Further cooling to $T = 0$ makes the wall thickness at temperatures $K \ll T_c$ equal approximately to L_m , but the nature of the solution changes somewhat. It follows from the free-energy functional (4) that in the limit $T \rightarrow 0$ the quantity $(S_{my}^2 + S_{mz}^2)^{1/2} = (u^2 + m^2)^{1/2}$ tends to $1/2$. It follows that in a domain wall the average pseudospin can only rotate, but there should be no change in its absolute value. This corresponds to the usual Bloch solution in a domain wall in weakly anisotropic ferromagnets at sufficiently low temperatures.

At very low values of T_m the wall thickness for rotational and purely structural types of solution is close to the interatomic distance. The question which of these solutions has the lower energy can be determined only for the specific behavior of the functions B_{mn} and I_{mn} as a dependence on the coordinates of the sites \mathbf{m} and \mathbf{n} .

4. THERMODYNAMICS OF A MAGNETIC TRANSITION IN A DOMAIN WALL

Using the solution (12), we can calculate the temperature dependence of the magnetization M inside a wall in that area of a wall which is occupied by one ion. Integrating $m(x)$ with respect to x , we obtain

$$M(T) = \int_{-\infty}^{+\infty} m(x, T) dx = \pi [\kappa_1 (T_{mw} - T) / D (T_c - T_m)]^{1/2}. \quad (13)$$

The moment lies in the plane of the wall and on the appropriate surface of a sample it creates a magnetic fringing field, which can be detected with a sensitive magnetometer. For example, if the wall length is 1 mm and we have $a \approx 10^{-7}$ cm, then for moments with the concentration $n \approx 10^{22}$ cm $^{-3}$ the magnetic flux is $\Phi \approx 10^{-6}$ Oe·cm 2 , which can easily be detected with a SQUID device.

We can calculate the specific heat discontinuity at the point T_{mw} if we know the surface energy of a wall for the solutions represented by Eqs. (8) and (12). Simple steps yield the specific heat discontinuity:

$$\Delta c = \frac{2T_{mw}}{D} \left(\frac{2\kappa_1}{T_c - T_m} \right)^{1/2}. \quad (14)$$

The ratio of this discontinuity at the point T_{mw} to the one at the structural transition point T_c is a/d , where d is the average distance between domain walls. For $a \approx 10^{-7}$ cm and $d \approx 10^{-4}$ cm, the discontinuity at the point T_{mw} is of the same order as the discontinuity of the specific heat at a superconducting transition and it can be estimated quite simply by an experimental method.

The expressions (13) and (14) are obtained in the self-consistent field approximation and their application to the magnetization localized within a wall requires separate justification. We have mentioned above that the dipole-dipole interaction makes fluctuations of the three-dimensional ordering effectively the same as in a four-dimensional space. This remains true in the case of the localized magnetization as long as the correlation radius $(\kappa_2/T_m \tau)^{1/2}$ does not exceed the magnetic layer thickness L_m . Therefore, the Ginzburg-Levanyuk parameter governing the range of strong thermodynamic fluctuations $T_{mw} = (1 \pm \tau_G)$ is in our case

$$\tau_G \approx \kappa_2 / T_m L_m^2 \approx \kappa_2 (T_c - T_m) / \kappa_1 T_m.$$

The parameter τ_G is small under conditions such that we can use a continuous representation of the magnetic ordering in a wall, which justifies the use of the mean-field theory in the description of the localized magnetic ordering in a wall.

In the range of reduced temperatures $\tau = |T - T_{mw}| / T_{mw} < \tau_G$ the divergence of fluctuations is no longer logarithmic with respect to τ , but as $\tau^{-3/4}$, i.e., exactly as in a system without the dipole-dipole interaction but with the dimensionality of space $d = 5/2$. We shall now prove this. In the k representation of a uniaxial magnet under consideration the free energy of the dipole-dipole interaction is

$$\mathcal{F}_{d-d} = \sum_{\mathbf{k}} \frac{k_z^2}{k_x^2 + k_y^2 + k_z^2} m_{\mathbf{k}} m_{-\mathbf{k}} \theta_{em}, \quad \theta_{em} = 8\pi n g'^2 \mu_B^2, \quad (15)$$

where $g' \mu_B$ is the nominal magnetic moment of an ion and n is the concentration of magnetic ions; it is assumed that θ_{em} and T_m are approximately of the same order of magnitude. In the range $\tau < \tau_G$ the important fluctuations are those with the projections of the moment k_z and k_y along the layer. Integrating Eq. (15) with respect to the distribution of k_x , typical of a localized solution for a wall of thickness L_m , we obtain (see Ref. 8)

$$\mathcal{F}_{d-d} = \sum_{k_y, k_z} L_m k_z^2 (k_y^2 + k_z^2)^{-1/2} m(k_y, k_z) m(-k_y, -k_z) \theta_{em}. \quad (16)$$

Using Eq. (16), we find that fluctuations of the specific heat above T_{mw} are described by the expression

$$c_{fl}(\tau) \approx \kappa_1^{3/4} \int dk_y dk_z \left[\frac{\kappa_1}{T_m} (k_y^2 + k_z^2) + \tau + \frac{k_z^2}{(k_y^2 + k_z^2)^{1/2}} \frac{L_m \theta_{em}}{T_m} \right]^{-2} \approx \left(\frac{\kappa_1}{\tau} \right)^{3/4} (T_m L_m^2)^{-1/4}. \quad (17)$$

Therefore, on transition through τ via τ_G the fluctuation dimensionality of space changes from $d = 4$ to $d = 2.5$.

5. STRUCTURAL JAHN-TELLER WALLS IN A MAGNETIC FIELD

We have shown earlier that cooling below T_{mw} gives rise to a magnetic ordering in structural domain walls. However, above the point T_{mw} (as in the case when there is no point T_{mw}) the presence of structural JT walls in a system also has a significant influence on its magnetic properties. In

turn, an external magnetic field alters the wall structure.

When the term $(-2g'\mu_B mH)$, describing the interaction with a magnetic field, is added to Eq. (16), we readily obtain an expression for the magnetic susceptibility (per one magnetic ion)

$$\chi_m = (1/2g'\mu_B)^2 (K - A + Du^2)^{-1}. \quad (18)$$

In the homogeneous case (or inside a domain) we find that below T_c we have $u^2 = u_0^2 = A/D$ and it is then clear from Eq. (18) that χ_m is independent of temperature below T_c , whereas above T_c it obeys the Curie-Weiss law: $\chi_m \propto (T - T_m)^{-1}$. We can also easily show that below T_c the magnetic moment is proportional to a magnetic field even when its intensity is high, right up to that corresponding to splitting of a non-Kramers doublet because of displacements u (it is proportional to u). On the other hand, above T_c an increase in the field causes saturation of the moment. These conclusions are in agreement with the experimental results and they have been known for a long time.¹

When domain walls are present in a crystal, the susceptibility begins to depend on temperature below T_c and the magnetic moment increases nonlinearly on increase in the applied magnetic field (both these effects are naturally proportional to the number of domain walls).

The contribution of a wall to the magnetic susceptibility of a sample can be found from the equation

$$\kappa_1 \ddot{m} + (A - K)m - Dmu_0^2 \text{th}^2 [x(A/2\kappa_1)^{1/2}] + g'\mu_B H = 0. \quad (19)$$

Near T_c at temperatures within the interval $T_c - T \ll T_c - T_m$ we find from Eq. (19) that the magnetic susceptibility of a wall (calculated per area occupied by one ion) is given by

$$\Delta\chi_{mw} = \chi_0 [2\kappa_1 (T_c - T)]^{1/2} / (T_c - T_m),$$

$$\chi_0 = (1/2g'\mu_B)^2 (T_c - T_m)^{-1}. \quad (20)$$

The solution in a wider temperature range (but still subject to the condition that $T_c - T_m \ll T_c$) is possible if $\kappa_1 = 3\kappa_2$. We then obtain

$$\Delta\chi_{mw} = \chi_0 [2\kappa_1 (T_c - T)]^{1/2} / [T_c - T_m + 2/3(T_c - T)], \quad (21)$$

which shows that the contribution of a wall increases as a result of cooling right down to T_{mw} , as demonstrated in Fig. 1, case *a* [if $\kappa_1 = 3\kappa_2$, we find that $T_c - T_{mw} = 3(T_c - T_m)^2$ and Eq. (21) has its maximum at $T = T_{mw}$]. At low values of T_m , when the point T_{mw} does not exist, the contribution of domain walls to the magnetic susceptibility reaches its maximum and then disappears in the limit $T \rightarrow 0$ (case *b* in Fig. 1). This is due to the fact that at low temperatures the change in the absolute value of u is undesirable from the point of the free energy and a structural domain wall is converted in the limit $T \rightarrow 0$ into a discontinuity of the direction of u between the neighboring lattice centers. Transformation of a 180° wall of the type described by Eq. (8) as a result of cooling was investigated in Ref. 9. It follows from Eqs. (20) and (21) that below T_c cooling has the effect that, in addition to the background with a constant

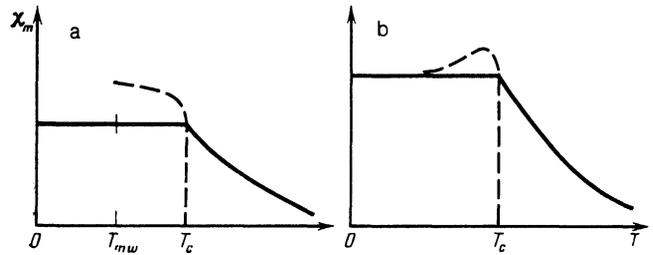


FIG. 1. Bulk contribution to the susceptibility (continuous curve) and the contribution of domain walls (dashed curve). The contribution of domain walls depends on their concentration: a) $T_{mw} > 0$, when a spontaneous moment appears in a wall below T_{mw} ; b) point T_{mw} does not exist.

value of χ_m , there should be a rise of the susceptibility of the bulk of the crystal and this rise should be proportional to $(T_c - T)^{1/2}$ because of the contribution of domain walls. Compared with the bulk contribution, that of the domain walls represents $[2\kappa_1 (T_c - T)]^{1/2} / d(T_c - T_m)$ and its is of the order of $a[(T_c - T)/T_c]^{1/2} / d$.

We have considered above the influence of domain walls on the magnetic susceptibility. In strong magnetic fields the structure of a wall itself changes and the magnetic moment of the wall increases nonlinearly on increase in the field. We shall find the first nonlinear correction to the moment of a wall near the temperature T_c , where $T_c - T \ll T_c - T_m$. It then follows from Eq. (19) that $m = m_0 = g'\mu_B H / K$ and the equation for u is

$$\kappa_1 \ddot{u} + Au - Du^3 - Dum_0^2 = 0, \quad (22)$$

which shows that the solution is of the type given by Eq. (8), but with A now dependent on H :

$$A(H, T) = T_c - T - Dm_0^2(H), \quad m_0(H) = g'\mu_B H / K. \quad (23)$$

The amplitude u_0 falls on increase in H since the magnetic field reduces the structural transition temperature. The wall width then increases for the same reason. Consequently, the magnetic moment of a wall is given by

$$M(H, T) = \chi_0 H \frac{\{2\kappa_1 [T_c(H) - T]\}^{1/2}}{T_c - T_m}$$

$$= \chi_0 H \frac{[2\kappa_1 (T_c - T)]^{1/2}}{T_c - T_m} \left[1 - \frac{Dm_0^2(H)}{2(T_c - T)} \right]. \quad (24)$$

The nonlinearity appears in fields $g'\mu_B H$ which are of the order of $(T_c - T_m)[T_c - T_m]/T_c]^{1/2}$.

6. CONCLUSIONS

It is shown above that if the temperature of a latent magnetic transition T_m lies slightly above the temperature of a structural transition T_c , then a localized magnetism (of the ferromagnetic or antiferromagnetic type) may appear inside a structural domain wall.

The point T_m is the point of a potential magnetic transition, which in fact does not occur because a stronger structural transition prevents it. However, the magnetic susceptibility χ_m obeys the Curie-Weiss law $\chi_m \propto (T - T_m)^{-1}$ above T_c and it demonstrates the existence of a temperature

of a latent magnetic transition T_m . This susceptibility result is very important, because it makes it possible to find compounds with similar parameters T_m and T_c . Such crystals exhibit a localized magnetism within structural domain walls. When structural domain walls are set in motion (for example, by the application of a stress), we can record the motion of the ferromagnetic moment localized in a wall.

Crystals with JT ions are most interesting for the observation of the magnetism localized in structural walls, since the magnetoelastic coupling in such crystals is maximal.

By way of example of compounds which are close in properties to the situation of interest to us, we shall consider thulium vanadate $TmVO_4$ (Ref. 10). The Tm ion contributes a non-Kramers doublet and the JT cooperative transition occurs at $T_c = 2.1$ K. The thermodynamics of this compound is described well by the simplest molecular field model. Above T_c this susceptibility obeys the Curie-Weiss law with the antiferromagnetic constant $\Theta = -0.51$ K; the g factor along the c axis is $g_c = 10.1$, whereas for other directions it is $g_a = g_a' = 0$. Therefore, in this compound a structural transition suppresses an antiferromagnetic transition which would have occurred at $T_m = 0.51$ K. The effects of interest to us will be manifested more clearly in compounds with somewhat closer values of T_m and T_c , and preferably with the ferromagnetic Curie-Weiss constant Θ .

Our analysis applies to insulating crystals with the exchange and dipole-dipole interactions of magnetic moments. In this case the coexistence of two types of ordering in the bulk is impossible. In metal systems the RKKY interaction

of magnetic ions may facilitate the appearance of inhomogeneous magnetic structures. Then, the coexistence of inhomogeneous magnetic and structural ordering in the bulk is in principle possible.

The authors are grateful to A. P. Levanyuk and D. I. Khomskii for valuable discussions and to V. L. Ginzburg and E. G. Maksimov for reviewing the paper in its manuscript form and critical comments.

¹For some types of structural transitions the domains are called twins. We are then speaking of a transition layer between twins which we shall call a domain wall.

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Translated by A. Tybulewicz