Circulation lines and motion of antiphase boundaries in an improper ferroelectric

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Circulation lines are known to exist in an improper ferroelectric with a two-component order parameter, gadolinium molybdate. These lines represent linear intrinsic defects. Following a path around such a defect causes a point in the order parameter plane to traverse a closed contour an integral number of times. Such lines appear at contacts between domain walls and also at walls rotated 180° out of phase, where they represent subdomain boundaries with different directions of rotation in the order parameter plane. Shear deformation associated with the induced Lifshitz invariant in the free energy creates a force which displaces a circulation line. A study is reported of the motion of a contact between 90° polarization and 180° antiphase boundaries as a result of the action of such a force together with the coercive force. An analysis is made of the phase diagram of an antiphase wall in the case when the bulk transition in an improper ferroelectric is a second-order phase transition far from the tricritical point. This phase diagram includes a region where a stable antiphase rotation wall exists in which circulation lines can appear.

1. INTRODUCTION

The domain structure of improper ferroelectrics is currently the subject of active theoretical and experimental studies (see Refs. 1–7 and the bibliography cited there). We shall consider the specific features of the structure of domain walls in a classical improper ferroelectric and ferroelastic, of gadolinium molybdate (GMO). A bulk phase transition in this crystal takes place from the D_{2d}^{3} symmetry (paraelectric phase) to the C_{2v}^{8} symmetry (ordered phase), and the ordering is accompanied by doubling of the unit cell volume and appearance of a two-component order parameter governing the amplitudes of the lattice distortions of the paraelectric phase which appear at this transition.[8] At the same time the energy attains a minimum at four points in the order parameter plane, which transform into one another under 90° rotations. The points correspond to four types of domain and a domain wall accompanied by 90° rotation corresponds to a change in the sign of the spontaneous polarization along the fourfold axis of the paraelectric phase and to a change in the sign of the shear spontaneous deformation (strain) in the basal plane. We shall call these the 90° or polarization walls. The domains differing in respect of rotation of the order parameter by 180° are identical in all their physical properties including polarization, but after a passage across such a 180° wall (called an antiphase wall) the structure is shifted by half the spatial period of the ferroelectric phase.

In a system with a two-component order parameter we can expect not only domain walls, i.e., plane arrays of singularities in the distribution of the order parameter (intrinsic defects), but also linear intrinsic defects. These linear defects will be called circulation (C) lines and they appear at contacts and intersections of domain walls or inside them if their symmetry allows the existence of different structures which are degenerate with respect to the energy, which splits these walls into subdomains. The walls which separate such subdomains are the C lines. The analog of these lines in ferromagnetism are the Bloch lines separating subdomains of Bloch walls.[9]

Here we present a qualitative and quantitative analysis of the structure of antiphase walls and C lines. We propose a method for creation of a concentrated force acting on a C line. The Lifshitz invariant in the free energy, induced by shear deformation, is responsible for this force. We show that the force applied to a C line at the contact between polarization and antiphase walls can displace the latter.

The range of existence of C lines in an antiphase wall are determined by investigating also the possible types of structures of an antiphase wall and phase transitions between them.

In Sec. 2 we describe a qualitative analysis of possible inhomogeneous structures in GMO, predicted on the basis of the properties of mapping of such structures onto the plane of a two-component order parameter. The force acting on a C line due to shear deformation is determined in Sec. 3. The next section (4) deals with an analysis of the structure of an antiphase wall and possible phase transitions in the wall. The motion of an antiphase wall under the action of the force defined in Sec. 3 and of the coercive force is discussed in Sec. 5.

2. DOMAIN WALLS AND C LINES IN GADOLINIUM MOLYBDATE

In this section we shall report a qualitative analysis of possible inhomogeneous distributions of the two-component order parameter of GMO using the properties of mapping of such structures onto the order parameter space. This qualitative analysis method is used widely in topological classification of inhomogeneous structures in systems with a complex order parameter[10] (the application specifically to improper ferroelectrics was made in Ref. 4, where relevant references are given).

The homogeneous state of the ferroelectric phase of GMO is quadruply degenerate and the four types of ground state correspond to four points in the $(q_1 q_2)$ plane of the order parameter, located on a circle (Fig. 1a). Introducing normal coordinates in this plane, we can say that these ground states correspond to the same modulus of the order parameter and four different angles (phases): $\varphi = 0, \pi/2, \pi$, and $3\pi/2$.[1)] The structures with domain walls correspond to mapping of the real space onto lines connecting these points in the $(q_1 q_2)$ plane. Mapping of a structure with a polarization 90° wall is demonstrated in Fig. 1b. Clearly, when the

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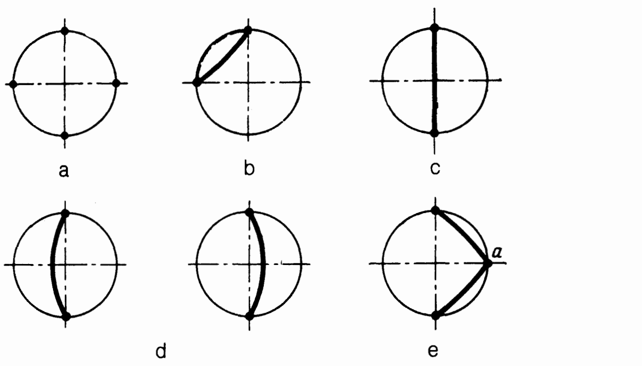

FIG. 1. Ground state and domain walls in gadolinium molybdate. Maps in the order parameter plane are shown.

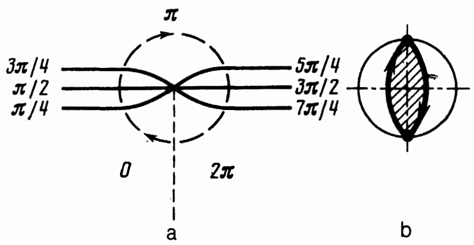

FIG. 2. Circulation line in an antiphase rotation wall. Figure 2a shows constant-phase lines (thin continuous lines) in real space as well as the line at which the phase changes by 2π (dashed curve). Figure 2b gives the map of the C line in the order parameter plane (shown shaded). The direction in which the map is traced is shown and it corresponds to the direction followed around the C line in real space, identified by the arrow in Fig. 2a.

anisotropy energy, i.e., the part of the energy dependent on the phase of the order parameter, tends to zero, a map of a domain wall tends to an arc of a circle (shown dashed in Fig. 1b). Such a domain wall is an analog of a rotation domain wall (Landau–Lifshitz wall) in magnetic materials where the anisotropy energy includes a relativistically small term.

Possible maps of a structure with an antiphase wall are shown in Figs. 1c–1e (see also Ref. 4). In the limit of strong anisotropy we can expect mapping of the structure of an antiphase wall onto the diameter of a circle (Fig. 1c). Such an antiphase wall will be called linear since inside this wall only one component of the order parameter differs from zero. Its analog in magnetism is a Bulaevskii–Ginzburg domain wall.[11] Lowering of the anisotropy energy may give rise to an antiphase wall of lower symmetry, when the corresponding map onto the $(q_1 q_2)$ plane encloses the point $q = 0$ (Fig. 1d). We shall call this a rotation wall. Since there are two ways of circumventing this point, which are energy-degenerate, it is possible to split an antiphase wall into subdomains with different circumnavigation paths. Linear walls between these subdomains represent circulation (C) lines discussed below.

In the limit of vanishing anisotropy a map of a 180° wall or of a 90° wall should approach an arc of a circle, but because of the fourfold rotation symmetry of the Hamiltonian, the 180° wall itself should become unstable against splitting into two 90° walls. The domain which appears between them is mapped onto point a in Fig. 1e.

Figure 2a shows the spatial structure of an antiphase rotation wall with a C line (constant-phase lines are shown), whereas Fig. 2b demonstrates mapping of this structure onto the order parameter plane. Then the domains with $\varphi = 0$ and π are mapped, as before, in the form of two points, whereas subdomains of a domain wall with φ varying from 0 to π and from π to 2π are represented by the thick lines passing to the left and right of the center of the plane and the core of a C line (surrounded by a dashed curve in Fig. 2a) is mapped to form the shaded region in Fig. 2b. Therefore, the map of the C line covers a finite area in the $(q_1 q_2)$ plane. The closed contour surrounding the C line in real space (shown in Fig. 2a dashed with an arrow indicating the direction of circumnavigation) is mapped to form a closed line surrounding the map and the direction of enclosure can be clockwise or anticlockwise (in Fig. 2b it is clockwise). These two directions of enclosure in the order parameter plane can be distinguished by attributing the C lines different circulations ± 1 of the phase of the order parameter (this was the reason why we introduced the term "circulation line"). However, this circulation differs fundamentally from the circulation of superfluid or magnetic vortices (Bloch lines). In our case its sign does not change when time is reversed, whereas in the case of superfluid and magnetic vortices the sign changes. Therefore, when a C line travels at a constant velocity, there is no gyrotropic force perpendicular to the velocity, which plays a very important role in the dynamics of superfluid and magnetic vortices (for an account of the gyrotropic force in ferromagnetic materials see Refs. 9 and 12 and the references cited there).

Figure 3a shows the structure after an antiphase wall near a C line breaks up into two polarization walls, whereas Fig. 3b shows a map of this structure. This structure is topologically indistinguishable from that in Fig. 2, but in real space inside an antiphase wall the domains which appear to the left and right of the line are characterized by $\varphi = \pi/2$ and $3\pi/2$ and they differ in the sign of polarization from the domains characterized by $\varphi = 0$ and π. However, an increase in the volume of the new domains that are formed as a result of breakup of an antiphase wall is limited by the finite density of the C lines in the original antiphase wall. Moreover, the structure with nodes (constrictions) shown in Fig. 4 is now observed.

We shall now consider contacts between polarization and antiphase walls which can be also identified with the C lines. The structure of such contacts and the corresponding maps on the order parameter plane are shown in Figs. 5 and

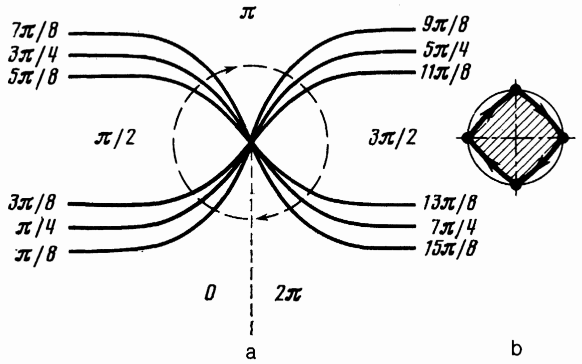

FIG. 3. Circulation line at the intersection between two polarization walls. The information given is the same as in Fig. 2.

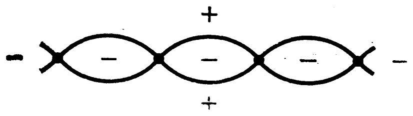

FIG. 4. Structure after splitting of an antiphase wall with C lines. The continuous lines are polarization walls; the points are the C lines. The signs (+ , −) of the polarization in the domains are shown.

6. We shall consider what happens if we try to transform these contacts into one antiphase wall by selecting several rotations of two polarization walls emerging from a contact (the direction of rotation is shown by arrows in Figs. 5a and 6a) and a subsequent continuous transformation (homotopy) of two polarization walls into one antiphase wall. Introduction of circulation as a topological invariant or charge means that in the course of this transformation we cannot change the number of times we traverse the boundary of the map of the structure around the center of the plane (q_1q_2) corresponding to zero order parameter. The physical reason for this is forbidden in that the boundary of the map *abc* in Figs. 5b and 6b is the map of a volume of polarization walls and vanishing of the order parameter inside a polarization wall requires a major increase in the energy proportional to the wall area. This increase in the energy is an activation barrier of *topological* origin and the structures which transform continuously into one another without overcoming this topological barrier, i.e., the structures with the same topological charge, are topologically equivalent or, in other words, they belong to the same homotopic class.[10] In the light of this discussion it is clear that the contact shown in Fig. 5 is topologically equivalent to an antiphase wall with a C line shown in Fig. 2 and the contact demonstrated in Fig. 6 is topologically equivalent to a homogeneous antiphase rotation wall. This is due to the fact that a topological charge, i.e., a circulation governed by the number and direction of enclosures along the boundary of a map around the point $q_1 = q_2 = 0$ is equal to $+1$ and 0 for the contacts in Figs. 5a and 6a, respectively. There may be also a contact which is intermediate between the cases shown in Figs. 5a and 6a: in this case the boundary of a map is identical with the diameter, i.e., it passes through the point where the order parameter has zero value (center of the circle). The circulation in the case of such a contact is an indeterminate quantity. This contact is topologically equivalent to a linear rotation wall

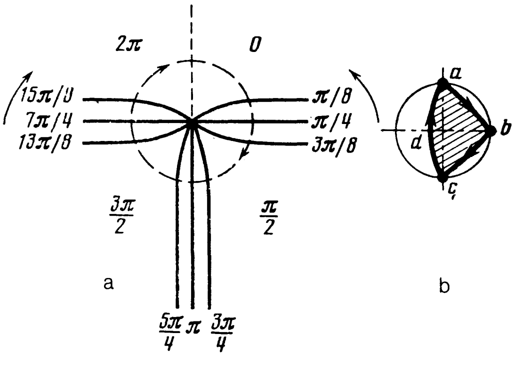

FIG. 5. Circulation line at the contact between polarization and rotation antiphase walls. The information given is the same as in Fig. 2.

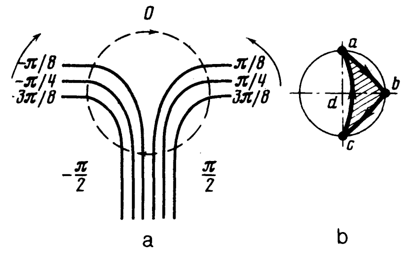

FIG. 6. Circulation line at a contact between polarization and rotation antiphase walls. The information given is the same as in Fig. 2, but there is no abrupt change in the phase, because the C line is characterized by zero circulation.

(Fig. 1c). However, in the problem under discussion these simple topological arguments cannot always have the same decisive role as in the case of other ordered condensed media, particularly in the case of ferromagnets when the anisotropy is usually weak and the maps of domain walls are always close to arcs of a circle. In the case of a ferroelectric characterized by a strong anisotropy the energies of antiphase rotation walls, mapped to form the contours *adc* in Figs. 5b and 6b, may hardly differ from the energy of a linear antiphase wall mapped along the diameter. This may mean that the topological barrier is no longer as high. On the other hand, there may be a barrier of nontopological origin preventing the merging of two polarization walls into one antiphase wall. For example, this is possible in that part of the phase diagram where a linear antiphase wall and two polarization walls are stable (see Sec. 4), although then one structure is absolutely stable and the other is only metastable. Moreover, there are forces of elastic origin which can orient polarization walls along the axes of the paraelectric phase[13] and prevent rotation, i.e., prevent merging into an antiphase wall. All this makes it desirable to consider not only topologically stable C lines with a specific nonzero circulation, but also special lines with zero or indeterminate circulation, because they may be stable although not in the usual topological sense.

A common property of all the C lines is that they correspond to a map with a finite area in the order parameter plane. The boundary of this map is a map of a closed contour in real space surrounding a C line at a large distance from it, i.e., a line which does not pass through its core. Then, if the spatial contour is traversed in a selected direction, the boundary of a map in (q_1q_2) may be circumvented clockwise or anticlockwise. As shown below, the properties of the C lines differing with respect to the direction in which the map is traversed defined in this way are different. Therefore, in order to distinguish such lines we have to generalize the concept of circulation defining it as the number and direction of circumventions of a map of a C line (around any internal point of this map, which now need not coincide with the point $q_1 = q_2 = 0$). In terms of this generalized definition of circulation the C lines in Figs. 5 and 6 have the same circulation of $+1$.

Our analysis may also be extended to more complex linear and planar singularities of the order parameter of an improper ferroelectric (for example, 270° or even 360° domain walls or intersections of more than two domain walls), but for the purpose of the present investigation it is sufficient to consider only the simplest situations.

3. FORCE ACTING ON A *C* LINE

A circulation line divides subdomains of a domain wall with different directions of rotation in the order parameter plane. Therefore, the force acting on a C line can be created only if there is an interaction which makes these subdomains inequivalent. This interaction may be the Lifshitz invariant induced by shear deformation and permitted by the transformation properties of the order parameter of gadolinium molybdate[8]:

$$\mathscr{F}_\varphi=\lambda\left[u_{xz}\left(q_1\frac{\partial q_2}{\partial y}-q_2\frac{\partial q_1}{\partial y}\right)+u_{yz}\left(q_1\frac{\partial q_2}{\partial x}-q_2\frac{\partial q_1}{\partial x}\right)\right], \quad (1)$$

where x, y, and z are selected along the axes of the paraelectric phase (here, z is the fourfold axis). Going over to the modulus q and the phase φ of the order parameter, we obtain

$$\mathscr{F}_\varphi=\lambda q^2\left[u_{xz}\frac{\partial\varphi}{\partial y}+u_{yz}\frac{\partial\varphi}{\partial x}\right]. \quad (2)$$

In the absence of the Lifshitz invariant the subdomains mentioned above are equivalent.[4]

We shall begin with the case of a C line in a planar antiphase wall. The resultant force F per unit length of a C line is equal to the difference between the integrals along a coordinate normal to an antiphase wall to the left and right of a C line. If we consider an antiphase wall in the yz plane, we obtain

$$F=\lambda u_{yz}\left(\int_{-\infty}^{\infty}q_l^2(x)\frac{\partial\varphi_l(x)}{\partial x}dx-\int_{-\infty}^{\infty}q_r^2(x)\frac{\partial\varphi_r(x)}{\partial x}dx\right), \quad (3)$$

where $q_l(x)$, $\varphi_l(x)$ and $q_r(x)$, $\varphi_r(x)$ are the order parameters in terms of polar variables to the left and right of a C line. We can readily rewrite Eq. (3) in the following form:

$$F=\lambda u_{xz}\oint_\Gamma q^2(\varphi)\,d\varphi, \quad (4)$$

where the contour Γ along which the integral is calculated represents the boundary of a map for a C line (Fig. 2b). The integral is equal to twice the map area. Clearly, this force should be directed along an antiphase wall.

In the more complicated case of a C line at a contact (Figs. 5 and 6) there should again be forces along all the walls, but the forces should now be added vectorially. For each domain wall emerging from a contact the expression for the force acting parallel to this wall may be written in a form similar to Eq. (4), but the contour Γ along which the integral is taken is a map of this domain wall (i.e., an open contour) and its magnitude is twice the area of the sector beginning from the point $q=0$ and bounded by the map in question. In the cases shown in Figs. 5 and 6 the forces acting on two parts of a polarization wall (contours ab and bc) are the same and they tend to shift the C line horizontally. However, the vertical force exerted by an antiphase wall is in such cases opposite in sign. However, if a linear antiphase wall emerges from a contact, it generally makes no contribution to the force acting on the C line.

It should be pointed out that in the cases under discussion dealing with C lines at contacts and a C line in an antiphase wall a reversal of the sign of circulation should be accompanied by a reversal of the sign of the resultant force, but in the case of the contacts shown in Fig. 6 the circulation should be regarded in the generalized sense (as defined at the end of Sec. 2).

4. PHASE TRANSITION IN AN ANTIPHASE WALL

In this section we shall present a quantitative theory of the structure of an antiphase wall and possible phase transitions between different types of such structures.

We shall describe the ferroelectric phase using the Landau expansion in powers of the components of the order parameter.[5,8] We shall write this expansion in terms of Cartesian coordinates:

$$\mathscr{F}=\frac{\alpha}{2}(q_1^2+q_2^2)+\frac{\beta}{4}(q_1^2+q_2^2)^2+2\beta_2q_1^2q_2^2+\frac{\gamma}{6}(q_1^2+q_2^2)^3+\frac{\varkappa}{2}\left[\left(\frac{\partial q_1}{\partial x}\right)^2+\left(\frac{\partial q_2}{\partial x}\right)^2\right] \quad (5)$$

as well as in terms of polar coordinates:

$$\mathscr{F}=\frac{\alpha}{2}q^2+\frac{\beta_1}{4}q^4-\frac{\beta_2}{4}q^4\cos4\varphi+\frac{\gamma}{6}q^6+\frac{\varkappa}{2}\left[\left(\frac{\partial q}{\partial x}\right)^2+q^2\left(\frac{\partial\varphi}{\partial x}\right)^2\right], \quad (6)$$

where $\beta_1=\beta+\beta_2$ This Landau expansion can be obtained, for example, from Eqs. (25) and (27) of Ref. 5 when the reference line for measuring the angle φ in the (q_1q_2) plane is selected suitably. It is also assumed that all the changes in the order parameter occur solely along one of the axes of the paraelectric phase (x axis).

This expansion was used in calculations dealing with 90° polarization walls.[5,6] Calculations relating to 180° antiphase walls[6,7] yielded contradictory results. Fouskova and Fousek[6] found a solution only for a linear antiphase wall and reached the conclusion that antiphase rotation walls cannot exist in the present model. In fact, they could not find a solution for an antiphase rotation wall because they sought an antiphase rotation wall which was nearly ideal in the rotation sense and exhibited mapping to form a semicircle. On the other hand, Ishibashi and Dvořak[7] concluded that there is always an antiphase wall structure with an energy lower than the energy of a linear antiphase wall, i.e., the latter is not optimal from the energy point of view and an antiphase wall can only be of the rotation type. This is in conflict with our analysis given above, which shows that there are regions of stability of linear and rotation walls and the existence of a rotation wall can be established in principle allowing also for small quantities γ in the Landau expansion.

The equilibrium structure of a domain wall should be found from the Euler–Lagrange equations obtained by variation of the free energy functional. In terms of Cartesian components q_1 and q_2, these equations are

$$\begin{aligned}-\varkappa\frac{\partial^2q_1}{\partial x^2}+\alpha q_1+\beta(q_1^2+q_2^2)q_1+4\beta_2q_2^2q_1+\gamma(q_1^2+q_2^2)^2q_1&=0,\\-\varkappa\frac{\partial^2q_2}{\partial x^2}+\alpha q_2+\beta(q_1^2+q_2^2)q_2+4\beta_2q_1^2q_2+\gamma(q_1^2+q_2^2)q_2&=0.\end{aligned} \quad (7)$$

The system of equations (7) has a solution for a linear antiphase wall,[6] for which $q_2=0$ and q_1 varies from $-q_{00}$ to q_{00}, where

$$q_{00}^2=(-\beta+(\beta^2-4\alpha\gamma)^{1/2})/2\gamma \quad (8)$$

is the equilibrium value of the order parameter in a domain. We then have

$$q_1(x)=q_0(x)=q_{00}\frac{e^{2Kx}-1}{[(e^{2Kx}+1)^2+4q_{00}^2A^{-1}e^{2Kx}]^{1/2}},\tag{9}$$

where

$$A=\frac{3}{2}\frac{\beta}{\gamma}+2q_{00}^2,\quad K=\left(\frac{\gamma}{3\varkappa}(q_{00}^2+A)q_{00}^2\right)^{1/2}.$$

The solution simplifies greatly in the special case when $\beta>0$ and $\gamma=0$, which corresponds to a model of a second-order phase transition:

$$q_2=0,\ q_1(x)=q_0(x)=q_{00}\,\mathrm{th}(x/d),\ q_{00}^2=-\alpha/\beta,\ d^2=2\varkappa/\alpha.\tag{10}$$

We shall consider the stability of this solution by a method used frequently in the past to investigate phase transitions in domain walls.[14,15] We have to expand the energy functional to second order in the small deviations q_2 and $q=\bar{q}_1-q_0$ from the solution (9) and determine the eigenvalues λ of this functional. The problem reduces to the following two linear differential equations:

$$-\varkappa\frac{\partial^2\bar{q}}{\partial x^2}+\alpha\bar{q}+3\beta q_0^2\bar{q}+5\gamma q_0^4\bar{q}=\lambda\bar{q},\tag{11}$$

$$-\varkappa\frac{\partial^2 q_2}{\partial x^2}+\alpha q_2+(\beta+4\beta_2)q_0^2q_2+\gamma q_0^4q_2=\lambda q_2.\tag{12}$$

A linear antiphase wall is stable if all the eigenvalues λ are nonnegative. The modes corresponding to $q_2=0$ and $\bar{q}\neq0$ cannot give rise to an instability, because the smallest eigenvalue λ for these modes is 0 (such a mode corresponds to translation of a domain wall). An instability of a linear antiphase wall may be related, however, to the appearance of a negative eigenvalue λ for the mode corresponding to $q_2\neq0$ and $\bar{q}=0$.

It was found possible to carry out a complete analytic investigation of a phase transition in a domain wall associated with this instability in the case when a bulk phase transition is of the second order and it lies far from the tricritical point ($\beta>0$, γ small). We shall consider this situation. If $\gamma=0$, the eigenvalue λ for the mode with $q_2\neq0$ passes through zero and becomes negative at $\delta=2\beta_2-\beta<0$. This can easily be shown because for $\gamma=0$ and $\beta_2/\beta=1/2$ Eq. (12) is completely identical to Eq. (11) which has zero eigenvalue. Therefore, if $\beta_2/\beta=1/2$ a linear antiphase wall is unstable against the appearance of a second component of the order parameter which makes the wall rotational.[2)] We shall consider the nature of the phase transition in the structure of a wall as a result of this instability. We shall do this bearing in mind that the nonlinear Euler–Lagrange equations of the system (7) have for $\delta=2\beta_2-\beta=0$ and $\gamma=0$ a class of solutions obtained by Ishibashi and Dvořak[7]:

$$q_1=\frac{q_{00}}{2}\left(\mathrm{th}\frac{x+D}{d}+\mathrm{th}\frac{x-D}{d}\right),\qquad q_2=\frac{q_{00}}{2}\left(\mathrm{th}\frac{x+D}{d}-\mathrm{th}\frac{x-D}{d}\right).\tag{13}$$

A remarkable property of this class of solutions is that the energy is independent of D, which thus becomes a degeneracy parameter, whereas a state with $\delta=0$ and $\gamma=0$ is a multicritical point in the phase diagram of a wall. If $D=0$ the solution given by the system (13) becomes identical with that given by Eq. (10), whereas for $D\gg d$ it corresponds to two polarization domain walls separated by a distance $2D$. Therefore, a neutral equilibrium with respect to transformation of a linear antiphase wall into a rotational wall and breakup of the latter into two polarization walls is established at a multicritical point. This degeneracy is lifted if $\delta\neq0$ and/or $\gamma\neq0$. To first order in these parameters the energy of an antiphase wall, measured from the energy of a linear antiphase wall at a multicritical point ($D=0$, $\gamma=0$, $\delta=0$), can be written in the form

$$\Delta\mathcal{F}=\frac{\gamma}{6}\int dx[(q_1^2+q_2^2)^3-q_0^6]+\delta\int dx\, q_1^2q_2^2,\tag{14}$$

where q_1 and q_2 is given by the solution (13) of Ishibashi and Dvořak, whereas q_0 is the solution for a linear wall described by the system (10).

After evaluating the simple but messy integrals in Eq. (14), we obtain

$$\Delta\mathcal{F}(D)=\tilde{\gamma}\left[\frac{1-z^2}{z^5}\left(\frac{1}{2}\ln\frac{1+z}{1-z}-z-\frac{z^3}{3}\right)-\frac{1}{5}\right]-\frac{\tilde{\delta}}{2}\left[\frac{1-z^2}{z^3}\left(\frac{1}{2}\ln\frac{1+z}{1-z}-z\right)-\frac{1}{3}\right],\tag{15}$$

where $z=\tanh(2D/d)$ increases from 0 to 1 when D is increased from 0 to ∞, and the following new parameters are introduced:

$$\tilde{\gamma}=\gamma q_{00}^6d=\gamma\left(\frac{2\varkappa}{\alpha}\right)^{1/2}\left(\frac{\alpha}{\beta}\right)^3,\quad \tilde{\delta}=\delta q_{00}^4d=\delta\left(\frac{2\varkappa}{\alpha}\right)^{1/2}\left(\frac{\alpha}{\beta}\right)^2.\tag{16}$$

An analysis of the dependence of ΔF on D makes it possible to plot the phase diagram in the $(\tilde{\delta},\tilde{\gamma})$ plane. At low values of $D\ll d$, we can expand Eq. (15) in terms of z:

$$\Delta\mathcal{F}=[-{}^2/_{35}\tilde{\gamma}+{}^1/_{15}\tilde{\delta}]z^2=[-{}^8/_{35}\tilde{\gamma}+{}^4/_{15}\tilde{\delta}](D/d)^2.\tag{17}$$

It follows from this expression that a linear antiphase wall ($D=0$) is stable, i.e., it corresponds to an energy minimum, if it lies below the AOB line in Fig. 7, where

$$\tilde{\gamma}<{}^7/_6\tilde{\delta}.\tag{18}$$

At high values $D\gg d$, we can expand Eq. (15) as a series in $1-z$, which gives

$$\Delta\mathcal{F}=-\frac{\tilde{\gamma}}{5}+\frac{\tilde{\delta}}{6}+\left[\tilde{\gamma}-\frac{\tilde{\delta}}{2}\right](1-z)\ln\frac{1}{1-z}=-\frac{\tilde{\gamma}}{5}+\frac{\tilde{\delta}}{6}+\left[\tilde{\gamma}-\frac{\tilde{\delta}}{2}\right]8\frac{D}{d}\exp\left(-\frac{4D}{d}\right).\tag{19}$$

It follows from Eq. (19) that the energy of an antiphase wall is an increasing function of D for $D\to\infty$ in the region below the straight line COD (Fig. 7), where

$$\tilde{\gamma}<\tilde{\delta}/2.\tag{20}$$

Therefore, this inequality defines the range of instability of two polarization walls against merging in the one antiphase wall. The phase diagram can be constructed if we compare the absolute values of the energies of antiphase ($D=0$) and of two polarization ($D=\infty$) walls. According to Eq. (19), below the line EF (Fig. 7), where

$$\tilde{\gamma}<{}^5/_6\tilde{\delta},\tag{21}$$

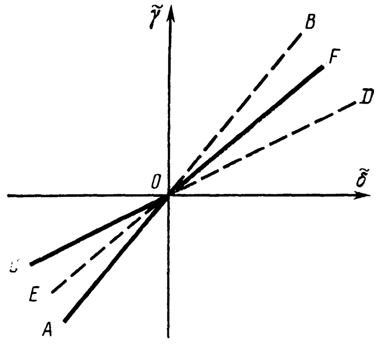

FIG. 7. Phase diagram of an antiphase wall near a multicritical point characterized by $\tilde{\gamma} = \gamma q_{00}^6 d = 0$ and $\tilde{\delta} = \delta q_{00}^4 d = 0$ [see Eqs. (5) and (10)]. The region below *AOF* is a linear antiphase wall which is absolutely stable, the region above *COF* represents two polarization walls which are absolutely stable. The sector *COA* is an antiphase rotation wall which is absolutely stable, the sector *BOF* is a linear antiphase wall which is metastable, and the sector *FOD* represents two polarization walls which are metastable against merging into a linear antiphase wall. The ray *OF* is a first-order phase transition line and the rays *OC* and *OA* are second-order phase transition lines.

the energy of a linear antiphase wall is less.

Consequently, the $\tilde{\delta}$–$\tilde{\gamma}$ plane splits into three regions separated by three lines emerging from a multicritical point $\tilde{\delta} = \tilde{\gamma} = 0$ (represented by the heavy lines in Fig. 7). In the region below *AOF* a linear antiphase wall is absolutely stable. This region is separated by the *OF* line of the first-order phase transition from the region above *COF*, where two polarization walls are preferable from the energy point of view to any antiphase wall, and by the *AO* line corresponding to the second-order phase transition from the region *AOC*, where a stable rotation wall exists (the energy ΔF is minimal for a finite value of D). On the *OC* line of the second-order phase transition a rotation antiphase wall it splits into two polarization walls. In the vicinity of the *OF* line of the first-order phase transition a rotation antiphase wall it splits into two polarization walls. In the vicinity of the *OF* line of the first-order phase transition there are "supercooling" and "superheating" regions where either a linear antiphase wall (sector *BOF*) or two polarization walls (sector *FOD*) are metastable. Clearly, the *C* lines can appear in an antiphase wall in the region *AOC* where the rotation type of wall is stable.

However, our phase diagram of an antiphase wall near the multicritical point $\gamma = \delta = 0$ cannot be used directly to describe transitions in the structure of an antiphase wall in GMO, because the volume phase transition in GMO is of the first order and it is close to the tricritical point, i.e., in the case of this compound we have $\beta < 0$ in the Landau expansion and in a wide range of temperatures the inequality $\beta^2 \ll \gamma|\alpha|$ is obeyed. We can expect however that the anisotropy parameter β_2/β governing the stability of a linear antiphase wall in the analysis given above can be replaced in the vicinity of a bulk tricritical point by a temperature-dependent parameter

$$\beta_2/(\beta^2-4\alpha\gamma)^{1/2} \sim \beta_2/2(|\alpha|\gamma)^{1/2}.$$

This is confirmed by an analysis of Eq. (12) designed to find the eigenvalues for the second component q_2 of the order parameter. We can easily show that in the case of sufficiently small values of $|\beta|$ the only parameter which determines the potential (i.e., the factor in front of q_2) and, consequently, the sign of the smallest eigenvalue is the dimensionless parameter

$$(\beta+4\beta_2)/(|\alpha|\gamma)^{1/2} \sim 4\beta_2/(|\alpha|\gamma)^{1/2};$$

the smallest value of λ is certainly positive if this parameter is large (strong anisotropy) and certainly negative if this parameter is small (weak anisotropy). Therefore, somewhere at $4\beta_2/(|\alpha|\gamma)^{1/2} \sim 1$ there should be a phase transition in the structure of an antiphase wall. This qualitative analysis of the stability shows that the range of existence of a stable linear antiphase wall is located near the temperature of the bulk phase transition (for small values of $|\alpha|$). However, cooling may result in a loss of stability by the wall [if the parameter $4\beta_2/(|\alpha|\gamma)^{1/2}$, which decreases as a result of cooling, can reach the critical value], but it is not certain whether this is accompanied by a transition to a rotation wall or by splitting into two polarization walls. A complete solution of this problem will require a more thorough quantitative analysis of the transition of the structure of an antiphase wall near a bulk tricritical point and also more information on the numerical values of the parameters of the Landau expansion for GMO.

5. FORCED MOTION OF AN ANTIPHASE WALL

As is known, an antiphase wall separates physically equivalent regions, i.e., there is no physical field that can make "antiphase domains" inequivalent and can thus create pressure on an antiphase wall. This limits greatly the possibility of creating forced motion of an antiphase wall (this wall can be set in motion by interaction of an arriving polarization wall driven by an electric field[1]). The proposed mechanism of forced motion of an antiphase wall does not require radical modification of the domain structure, which accompanies the motion of a polarization wall, and it is a more selective effect which acts only on an antiphase wall. The mechanism involves applying a force to a contact between antiphase and polarization walls (Fig. 5). Such a force is created under the influence of elastic stresses; it was evaluated in Sec. 3 [Eq. (4)]. This force tends to shift both antiphase and polarization walls. However, there are considerable forces of elastic origin, which orient the polarization wall along the axis of the paraelectric phase and prevent bending. Such forces are not created by bending of an antiphase wall, because it separates domains with equivalent elastic properties. Therefore, we shall assume that the force acting on a contact shifts an antiphase wall by displacing its end along an immobile rectilinear polarization wall. This displacement should be hindered by the coercive forces. Let us assume that in the initial position an antiphase wall coincides with the *xz* plane, whereas a polarization wall coincides with the *yz* plane (i.e., the contact is along the *z* axis). The displacement $y(x)$ of the antiphase wall then satisfies the equation of balance of the force proportional to the surface tension of the wall σ and the coercive force of density f:

$$-\sigma\frac{\partial^2 y}{\partial x^2}+f=0. \qquad (22)$$

Equation (22) gives the displacement of an antiphase wall in the region $0 < x < x_0$, where the surface tension force exceeds the dry friction (coercive) force. If $x > x_0$, the wall does not move, i.e., $y=0$. Equation (22) is solved subject to the boundary conditions $y=0$ and $\partial y/\partial x=0$ at $x=x_0$ and has a solution of the type

$$y=\frac{f}{2\sigma}(x-x_0)^2. \qquad (23)$$

The condition $\partial y/\partial x=0$ at $x=x_0$ ensures the absence of a kink of the wall at this point, which may appear only in the presence of a concentrated force. Such a concentrated force F acts at a contact; it is governed by Eq. (4) and it is equal to the flux of the momentum along the wall:

$$F=-\sigma\frac{\partial y}{\partial x}\bigg|_{x=0}=fx_0. \qquad (24)$$

After elimination of x_0 from Eqs. (23) and (24), we obtain the relationship between the force F at a contact and the displacement of a contact $y_0=y(0)$:

$$y_0=\frac{fx_0^2}{2\sigma}=\frac{F^2}{2f\sigma}=Cu_{xz}^2. \qquad (25)$$

The length C is given by

$$C=\frac{\lambda^2}{2f\sigma}\left(\int_\Gamma q^2(\varphi)\,d\varphi\right)^2\sim\frac{(\lambda q_{00}^2)^2}{f\sigma}. \qquad (26)$$

In the absence of sufficient information on the constants of the theory, we can obtain only an order-of-magnitude estimate of the length C in the temperature interval characterized by $(|\alpha|\gamma)^{1/2}\gg\beta$. Then, because

$$\sigma\sim\frac{|\alpha|q_{00}^2}{K}\sim q_{00}^4(\varkappa\gamma)^{1/2},$$

we find the following order-of-magnitude relationship

$$C\sim\lambda^2/f(\gamma\varkappa)^{1/2}.$$

Using the standard dimensional estimates for λ, γ, and $\varkappa$, we obtain

$$C\sim\varepsilon_{\rm at}a/f,$$

where $\varepsilon_{\rm at}\sim10^{11}$–$10^{13}$ erg/cm^3 is the characteristic density of the atomic energy and $a\sim10^{-8}$ cm is the characteristic interatomic distance. The coercive force depends on the scale and amplitude of inhomogeneities of a sample and it is not known for an antiphase wall. However, in a rough estimate we can use its typical values for a polarization wall found in Ref. 16:

$$f\approx5\cdot10^3\ \text{erg/cm}^3.$$

Using the values given above, we thus find that $C\sim0.2$–20 cm. Therefore, a strain of 10^{-3} gives rise to a displacement of $\sim$20–2000 Å.

6. CONCLUSIONS

A qualitative analysis of the structure of circulation lines in an improper ferroelectric is given above. These lines represent linear singularities of an antiphase wall or contacts of antiphase and polarization walls, and they correspond to a finite area of a map in the plane of a two-component order parameter. It is shown that C lines can be influenced, i.e., that it is possible to create concentrated forces acting on these lines. A force of this kind is created by a deformation coupled to a phase gradient, i.e., which occurs in the free energy in the form of the Lifshitz invariant induced by deformation. This force can also be used to act on an antiphase wall via a C line at a contact between antiphase and polarization walls. The estimates obtained, which allow for the existence of the coercivity, suggest that it should be possible to observe experimentally this effect.

Phase transitions in the structure of an antiphase wall were also considered. The phase diagram of this wall was constructed near a multicritical point where there is a neutral equilibrium in the state of the wall. It was found that there is a region in this diagram where a stable antiphase rotation wall exists and C lines can form. In the case of gadolinium molybdate, in which a bulk phase transition is close to the tricritical point, the state of an antiphase wall is governed by the effective anisotropy parameter $\beta_2/(|\alpha|\gamma)^{1/2}$, which depends on temperature. When temperature is lowered (and $|\alpha|$ is increased), this parameter becomes smaller and can reach a critical value at which a linear antiphase wall becomes unstable. If $\beta_2/(|\alpha|\gamma)^{1/2}$ is sufficiently small, this wall can become unstable against breakup into two polarization walls, but this instability occurs either in the form of one first-order phase transition directly to a state with two polarization walls or in the form of two second-order phase transitions passing through an intermediate state in the form of an antiphase rotation wall (separated by C lines into subdomains). The transition from an antiphase wall to two polarization walls should be accompanied by the appearance of a chain structure with nodes of intersection of polarization walls (C lines), of the kind shown in Fig. 4. It should be possible to observe experimentally a structure of this type. A detailed determination of the phase diagram of an antiphase wall in gadolinium molybdate will require further quantitative analysis of phase transitions in this wall in the case of values of $|\alpha|\gamma/\beta^2$, which are not small; moreover, more reliable information is needed on the parameters of the Landau expansion for the free energy in gallium molybdate.

[1)] In contrast to the treatments in Refs. 4–8, we selected the line from which the phase was measured in such a way that $\varphi=0$ corresponded to one of the ground states.

[2)] According to Ref. 7, a linearized phase wall is unstable and in the case of a strong anisotropy we have $\beta_2/\beta>1/2$. This follows from the inequality (34) in Ref. 7. However, according to our calculations this inequality should have the opposite sign, which corresponds to a stable linear antiphase wall.

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