ON ASYMPTOTIC SOLUTIONS IN THE MICROSCOPIC THEORY OF FERROELECTRIC DOMAIN STRUCTURES

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Recently, a general microscopic formalism was proposed by one of us (W.Z.) which permits a uniform description of ferromagnetic as well as ferroelectric domain structures. The main idea of this approach resides in employing inhomogeneous rotations of the spins or electric dipoles, respectively, and setting up a suitable variational principle. As variational parameters one can choose, generally, either the rotating angles or the direction cosines of the rotating axes. To facilitate the calculations, the former description is preferably used though in principle both the procedures are strictly equivalent. The present paper studies the applicability and efficiency of the latter procedure by applying it to some specific domain structures of ferroelectric crystals and imposing asymptotic boundary conditions. The variational principle is derived for the case of an orthorhombic dipole-lattice, and the Euler-Lagrange equations are solved in the limit cases when the deviation from a cubic lattice is either remarkable or negligible (both in a specific sense). Effective formulae are given for the thickness and energy of three types of inter-domain walls, and the results are compared with those obtained when using conventional methods. Moreover, a satisfactory qualitative explanation of the influence of particular homogeneous lattice-deformations on the direction of polarization and type of domain structure can be given.

1. *Introduction*

As recently shown by Ziętek [1], the microscopic formalism given in [2] which has already been applied, with considerable success, to ferromagnetic domain structures (*e.g.*, [3—8]) can easily be generalized to that extent as to be applicable to ferroelectric domain structures, too. This formalism, in its generalized form, has already been employed in [9] where the asymptotic and periodic solutions corresponding to domain structures of perovskites are investigated and the influence of a tetragonal deformation of the cubic crystal lattice on these solutions examined. Rotations as well as elongations of the elementary polarization vectors are considered in [9] though the equations are solved in the limit cases only when either rotation or elongation occurs. For the case of rotations, however, the rotating angles are chosen as variational parameters, the direction cosines of rotating axes being put constant before starting the calculations. None the less, it is pointed out in [9] and exhaustively discussed in [1] that one can as well choose the direction cosines as variational parameters and put the rotating angle, φ, constant and equal π from the very beginning, without restricting in

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any way the degrees of freedom of the problem. Furthermore, it is also pointed out in [1] that both the procedures though strictly equivalent when rigorously treating the problem may lead, practically, to somewhat different results because of the simplifications and various approximations that, of necessity, have to be done when intending to arrive at effective results. It seems thus expedient to examine this alternative description, and this is the task of the present paper.

2. *General assumptions*

Following [1, 9] we shall use the notion of the "dipole lattice" and assume it to be orthorhombic, the corresponding lattice constants being a, b, c in the directions x_1, x_2, and x_3, respectively (see Fig. 1). Let P_μ^α be the elementary polarization vector assigned to the site

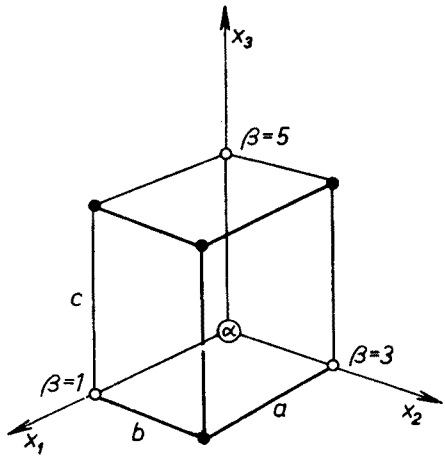

Fig. 1. Elementary cell of orthorhombic dipole-lattice, co-ordinate frame, denotation of lattice constants and labelling of nearest neighbours (*cf.* scheme (11))

$\alpha(\mu=1,2,3)$ and $A^{\alpha\beta}_{\mu_1\mu_2}$ $B^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4}$ the interaction tensors of second and fourth rank, respectively. In the absence of an external electric field the Hamiltonian will then have the form [1]

$$H = \sum_{\alpha\beta} \{A^{\alpha\beta}_{\mu_1\mu_2} P^\alpha_{\mu_1} P^\beta_{\mu_2} + B^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} P^\alpha_{\mu_1} P^\alpha_{\mu_2} P^\beta_{\mu_3} P^\beta_{\mu_4}\}, \tag{1}$$

where $\alpha \neq \beta$. (With respect to tensor indices μ Einstein's summation rule is used.)

The rotating matrices $R^\alpha_{\mu_1\mu_2}$ introduced in [2] and applied, inter alia, in [1, 9] have to be specified according to our assumtion that the rotating angles φ^α are for all sites the same and equal π. Thus, the matrices become simply

$$R^\alpha_{\mu_1\mu_2} = 2e^\alpha_{\mu_1} e^\alpha_{\mu_2} - \delta_{\mu_1\mu_2}, \tag{2}$$

e^α_μ being direction cosines of the rotating axis associated with site α. Besides.,

$$(e^\alpha_\mu)^2 = 1. \tag{3}$$

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Let us denote by P Debye's conventional elementary polarization vector [1], and let P_μ be a vector ("generating" vector, see [1, 9]) of length P. Then,

$$P^\alpha_{\mu_1} = R^\alpha_{\mu_1\mu_2} P_{\mu_2}, \tag{4}$$

if elongations of the polarization vectors are not taken into account [9], and the Hamiltonian (1) becomes

$$H = \sum_{\alpha\beta} \{\tilde{A}^{\alpha\beta}_{\mu_1\mu_2} P_{\mu_1} P_{\mu_2} + \tilde{B}^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} P_{\mu_1} P_{\mu_2} P_{\mu_3} P_{\mu_4}\}, \tag{5}$$

$$\tilde{A}^{\alpha\beta}_{\mu_1\mu_2} = \tilde{A}^{\alpha\beta}_{\nu_1\nu_2} R^\alpha_{\nu_1\mu_1} R^\beta_{\nu_2\mu_2}, \tag{6}$$

$$\tilde{B}^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} = B^{\alpha\beta}_{\nu_1\nu_2\nu_3\nu_4} R^\alpha_{\nu_1\mu_1} R^\alpha_{\nu_2\mu_2} R^\beta_{\nu_3\mu_3} R^\beta_{\nu_4\mu_4}. \tag{7}$$

Further, let $\varrho^{\alpha\beta}_\mu$ be the spatial vector between sites α and β, its length being $\varrho^{\alpha\beta}$. Then, it is convenient to introduce the dimensionless quantities

$$\eta^{\alpha\beta}_\mu = \varrho^{\alpha\beta}_\mu / \varrho^{\alpha\beta}. \tag{8}$$

Assuming the interaction tensors to be of dipolar and quadrupolar type one can write [7, 9]

$$A^{\alpha\beta}_{\mu_1\mu_2} = A^{\alpha\beta}\{\delta_{\mu_1\mu_2} - 3\eta^{\alpha\beta}_{\mu_1}\eta^{\alpha\beta}_{\mu_2}\}, \tag{9}$$

$$B^{\alpha\beta}_{\mu_1\mu_2\mu_3\mu_4} = B^{\alpha\beta}\eta^{\alpha\beta}_{\mu_1}\eta^{\alpha\beta}_{\mu_2}\eta^{\alpha\beta}_{\mu_3}\eta^{\alpha\beta}_{\mu_4}, \tag{10}$$

where the coupling functions $A^{\alpha\beta}$ and $B^{\alpha\beta}$ depend merely on the distance between sites α and β.

For the sake of simplicity, we shall restrict our considerations to interactions between nearest elementary polarization vectors[1].

If the lattice constants a, b, c of the orthorhombic lattice differ not too much from each other, each site α has six nearest neighbours β that correspond to those ones in the (simple) cubic lattice. In the following, let "central" sites be numbered with α and neighbouring ones with ($\beta = 1, 2, \ldots, 6$), the numeration of the latter being as indicated in Fig. 1. The configuration vectors (8) are then as follows

β	1	2	3	4	5	6
$\varrho^{\alpha\beta}_\mu$	$+a$	$-a$	$+b$	$-b$	$+c$	$-c$
$\eta^{\alpha\beta}_\mu$	$+1$	-1	$+1$	-1	$+1$	-1
	$\searrow\swarrow$		$\searrow\swarrow$		$\searrow\swarrow$	
	$\delta_{1\mu}$		$\delta_{2\mu}$		$\delta_{3\mu}$	

(11)

where the values in the columns have to be multiplied by the corresponding $\delta_{\lambda\mu}$, as marked in the last row.

[1] That interactions between farther sites can be taken into cosideration as well is shown in [8]. As for conditions under which this can be done and the accuracy of the procedure see, however, Appendix I in [9] and remarks in [1].

Finally, since the interaction is restricted to the first-order neighbourhood we may confine ourselves to the first-order Taylor-series approximation [1, 9] when expressing the variational parameters assigned to the neighbouring sites by those of the corresponding central site, *i.e.*

$$e_\nu^\beta = (1+\varrho_\mu^{\beta\alpha}\partial_\mu^\alpha)\, e_\nu^\alpha, \qquad \partial_\mu^\alpha \equiv \frac{\partial}{\partial x_\mu^\alpha}. \tag{12}$$

Moreover, we shall assume the (single) crystal to have rectangular shape with dimensions L_μ in the directions x_μ, and simplify the problem to the one-dimensional case (plate domain structure) in that we specify

$$e_1^\alpha = 0, \tag{13}$$

and assume the remaining direction cosines to be functions of x_1 only (domains parallel to the co-ordinate plane $x_2 0 x_3$).In this instance no electric charges are produced on the inter--domain walls as the component of the polarization vector normal to the wall is constant on passing through the wall.

It should perhaps be mentioned that due to the specification (9) and restriction to certain a neighbourhood the influence of free electric charges at the surface of the crystal on the internal domain structure is automatically excluded from our considerations (*cf.* Appendix VI in [9]), and due to (11) inhomogeneous lattice deformations (such as internal stresses) are not taken into account. Hence, all lattice sites are mutually equivalent and we can drop the superscripts in the coupling functions in Eqs. (9) and (10) using instead the simpler notation shown below:

β	1,2	3,4	5,6
$A^{\alpha\beta}$	A_a	A_b	A_c
$B^{\alpha\beta}$	B_a	B_b	B_c.

(14)

3. *Variational principles*

As an example, let us study the case when domains are polarized in direct ons parallel to the walls, *i.e.* perpendicularly to the co-ordinat axis x_1 because of Eq. (13). To be able to determine (in the co-ordinate plane $x_2 0 x_3$) the direction of polarization of the domains for both the cases $b<c$ as well as $b>c$, it is convenient to perform the calculations twice according to the following two specifications of the generating vector

$$P_\mu = P\delta_{\mu 2}, \tag{15a}$$

$$P_\mu = P\delta_{\mu 3}. \tag{15b}$$

In either case the transformed interaction tensors (6) and (7), thus the Hamiltonian (5) too, become relatively simple, partly because of the specifications (9) and (10), partly because Eqs. (15a) and (15b) imply in (4) that only the respective columns $R_{\mu,2}^\alpha$ and $R_{\mu,3}^\alpha$ of the

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rotating matrices (2) be needed. Besides, the first elements in these columns are zero because of Eq. (13).

Now, due to Eqs. (3) and (13) one can express the matrix elements (2) by means of a single direction cosine. Let it be e_2^α for the case (15a) and e_3^α for the case (15b). Similarly, when expressing the direction cosines e_μ^β ascribed to a neighbouring site β by those of the corresponding central site α, according to Eq. (12), one can use again Eqs. (3) and (13) in order to reduce the (first-order) derivatives to a single one (*i.e.* with respect to a single direction cosine, as specified just above). Hence, the variational problem can be formulated by using only one component of the unit vector which determines the direction of the rotating axis in every site, thus the functional derivable from (5) in the usual way (*cf.* [1, 2, 9]) will depend on a single function and its first derivative. As the procedure has been oftentimes demonstrated (see [1—3, 7—9], especially [9]) and a general description can be found in [1], we will omit here all the details and merely indicate the successive steps that have to be done when passing from the Hamiltonian (5) to the functional. First, one has to insert expressions (9), (10) and (2) into (6) and (7), and the resulting quantities into (5). Then, one can specify the generating polarization-vector P_μ by substituting in (5) either (15a) or (15b). The next step resides in utilizing Eqs. (3) and (13) and eliminating in (5) either the direction cosines e_3^α and e_3^β or e_2^α and e_2^β, according to whether specification (15a) or (15b) is used, respectively. After doing this one has to replace in (5) the direction cosines assigned to the neighbouring sites β (*i.e.* e_2^β for the case (15a) and e_3^β for the case (15b)) by the direction cosines (and their first derivatives) of the central sites α, by the aid of Eq. (12). Due to the last step the sum over β in (5) becomes evaluable for there are no variational parameters (*i.e.* direction cosines) with index β. All one needs in evaluating this sum is the form of the coupling functions in (9) and (10) and the configuration vectors (8), which in our case (non-deformed orthorhombic dipole-lattice, co-ordinate axes along principal crystallographic directions, restriction to first-order-neighbourhood interaction) are given by (11) and (14). When carrying out the calculations according to the above scheme one arrives at the following functionals, corresponding for $\lambda = 2$ and $\lambda = 3$ respectively to the specifications (15a) and (15b)

$$H[\vartheta_\lambda] = \frac{P^2}{abc} L_2 L_3 \int \{u_0(\cos 2\vartheta_\lambda - 1)\dot{\vartheta}_\lambda^2 + (-1)^\lambda u_4 \cos 4\vartheta_\lambda + u_8 \cos 8\vartheta_\lambda + \text{const}_\lambda\} \, dx_1, \tag{16}$$

where $\dot{\vartheta}_\lambda = \partial\vartheta_\lambda/\partial x_1$ and

$$e_\lambda = \cos \vartheta_\lambda, \tag{17}$$

$$u_0 = A_a a^2, \tag{18}$$

$$u_4 = 3\,(A_c - A_b) + P^2\,(B_b - B_c), \tag{19}$$

$$u_8 = \frac{P^2}{4}\,(B_b + B_c). \tag{20}$$

(Note that λ takes only the values 2 and 3).

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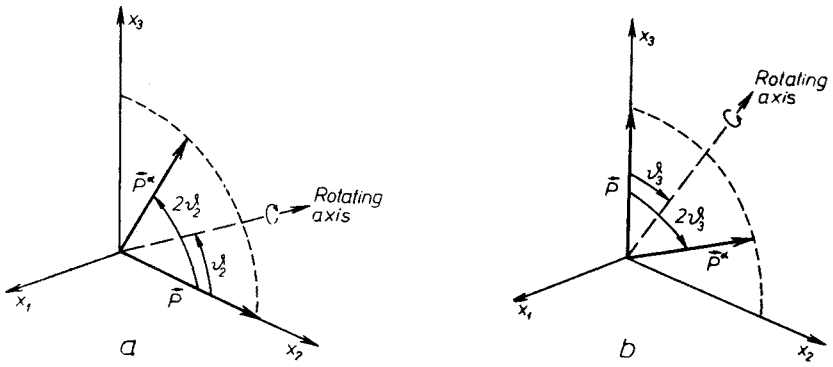

Fig. 2. Geometrical meaning of variational parameters ϑ_λ and position of generating vector P_μ for $\lambda = 2$, (*a*), and $\lambda = 3$, (*b*) (*cf.* Eqs. (2), (4), (13), (15a, b) and (17))

The angles ϑ_2 and ϑ_3 are indicated in Fig. 2 which illustrates the position of the generating vector P_μ in both the cases, as well as the position of the elementary polarization vector P_μ^α as a result of rotating the vector P_μ by the angle π around an axis lying in the co-ordinate plane $x_2 0 x_3$ and forming the angle ϑ_λ with the generating vector.

4. *Solution of Euler-Lagrange equations*

As the integrand in (16) does not depend explicitly on x_1, one can write the Euler-Lagrange equation in the form of a first-order differential equation, namely,

$$\dot{\vartheta}_\lambda^2 = \frac{(-1)^\lambda u_4 \cos 4\vartheta_\lambda + u_8 \cos 8\vartheta_\lambda + C_\lambda}{u_0(\cos 2\vartheta_\lambda - 1)}, \tag{21}$$

the constant C_λ to be determined from the boundary conditions.

We shall solve equation (21) in the limit cases when either u_8 or u_4 is negligible, that is when either inequality $|u_4| \gg |u_8|$ or $|u_4| \ll |u_8|$ holds. In the first instance our requirement implies the dipolar coupling to be much stronger than the quadrupolar one, and the difference between lattice constants b and c to be sufficiently large to ensure the condition 12 $|A_b - A_c| \gg P^2 |B_b + B_c|$ as may easily be concluded from Eqs. (19) and (20). In the latter instance we conclude from similar considerations that the difference between those two lattice constants must be sufficiently small, that is the lattice must be almost cubic (at least in the $x_2 0 x_3$ plane). The lower limit for the difference $|b - c|$ in the first case and the upper limit in the second depend substantially on the (relative) intensity of both types of interaction and the character it changes with distance. It is not the purpose of the present paper to analyse this question in further details, nor to list ferroelectric substances and experimental conditions for which those limit assumptions may hold. Suffice it to say, however, that there are a number of perovskites that in the vicinity of their transition temperatures are likely to fit the required conditions (*cf.* [10, 11]), either when passing from the paraelectric (cubic lattice) to the first ferroelectric phase (tetragonal lattice) or from the first to the second ferroelectric phase (orthorhombic lattice).

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Although it is not yet quite clear how to specify the coupling functions $A^{\alpha\beta}$ and $B^{\alpha\beta}$, especially the latter one (*cf.* [12—17]), nevertheless, they must correspond to the first two coefficients of Devonshire's thermodynamic potential [18, 9] and these are known to be functions of temperature and stress. Hence, in a microscopic theory the (effective) coupling functions though depending solely on the inter-atomic distance when properly chosen can be expected to describe phase transitions as well provided the temperature-dependence of the lattice constants is known. One should thus be able to fix, for instance, the temperature regions in which our approximate solutions are justified.

Let us now solve equation (21) in those two limit cases.

$$1^\circ\colon\ |u_4| \gg |u_8|.$$

In this instance we put $u_8 = 0$ in (21). In order to determine the constant C_λ we have to impose appropriate boundary conditions. As we are actually concerned in asymptotic solutions (two infinite domains separated by a finite wall, *cf.* [9]) we demand

$$\dot{\vartheta}(x_1 = \pm\infty) = 0. \tag{22}$$

One easily proves that this condition does not determine C_λ until the value of ϑ_λ at plus or minus infinity is fixed. There are two values that lead to real solutions and correspond to domains polarized (at infinity) in the main crystallographic directions; namely,

$$\vartheta_\lambda(x_1 = +\infty) = \pi/2 \tag{23A}$$

and

$$\vartheta_\lambda(x_1 = +\infty) = \pi/4. \tag{23B}$$

(See however Appendix) Accordingly, the constant in Eq. (21) takes the values

$$C_\lambda = (-1)^{\lambda+1}u_4, \tag{24A}$$

$$C_\lambda = (-1)^\lambda u_4, \tag{24B}$$

and the equation itself becomes

$$\dot{\vartheta}_\lambda^2 = (-1)^\lambda \frac{u_4(\cos 4\vartheta_\lambda - 1)}{u_0(\cos 2\vartheta_\lambda - 1)}, \tag{25A}$$

$$\dot{\vartheta}_\lambda^2 = (-1)^\lambda \frac{u_4(\cos 4\vartheta_\lambda + 1)}{u_0(\cos 2\vartheta_\lambda - 1)}, \tag{25B}$$

respectively. These equations can be given the simple form

$$\dot{\vartheta}_\lambda = \pm 2\sqrt{(-1)^\lambda u_4/u_0}\cos\vartheta_\lambda, \tag{26A}$$

$$\dot{\vartheta}_\lambda = \pm\sqrt{(-1)^{\lambda+1}u_4/u_0}\,\frac{\cos 2\vartheta_\lambda}{\sin\vartheta_\lambda}. \tag{26B}$$

A straightforward integration provides the solutions

$$\tan(\vartheta_\lambda/2) = \tanh(x_1\sqrt{(-1)u_4/u_0}), \tag{27A}$$

$$2\cos\vartheta_\lambda = \sqrt{2}\tanh(2x_1\sqrt{(-1)^{\lambda+1}u_4/u_0}) \tag{27B}$$

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when choosing respectively the intervals

$$-\pi/2 \leqslant \vartheta_\lambda \leqslant \pi/2, \tag{28A}$$

$$3\pi/4 \geqslant \vartheta_\lambda \geqslant \pi/4 \tag{28B}$$

for $-\infty \leqslant x_1 \leqslant +\infty$. The last integration constant has been determined according to the conditions

$$\vartheta_\lambda(x_1 = 0) = 0, \tag{29A}$$

$$\vartheta_\lambda(x_1 = 0) = \pi/2. \tag{29B}$$

As we see for $u_0/u_4 > 0$ the real solutions are: (27A) with $\lambda = 2$ and (27B) with $\lambda = 3$, and *vice versa* for $u_0/u_4 < 0$ we have the real solutions: (27A) with $\lambda = 3$ and (27B) with $\lambda = 2$.

Solutions (27A) describe domains polarized in parallel directions and separated by a 360°-wall which is polarized opposite to the domains. Solutions (27B) correspond to domains that are polarized in antiparallel directions and separated by a 180°-wall, this being polarized perpendicularly to the domains. According to the value of λ which is determined by the sign of the product $u_0 u_4$ the direction of polarization of the domains is parallel to the co-ordinate axis x_λ in the first case, and to $x_{5-\lambda}$ in the latter. This is illustrated in Fig. 3 where the solutions (27A) are marked with A (shape of curve independent of λ) and solutions (27B) with B (*cp.* Eqs. (15a), (15b), (17) and Fig. 2).

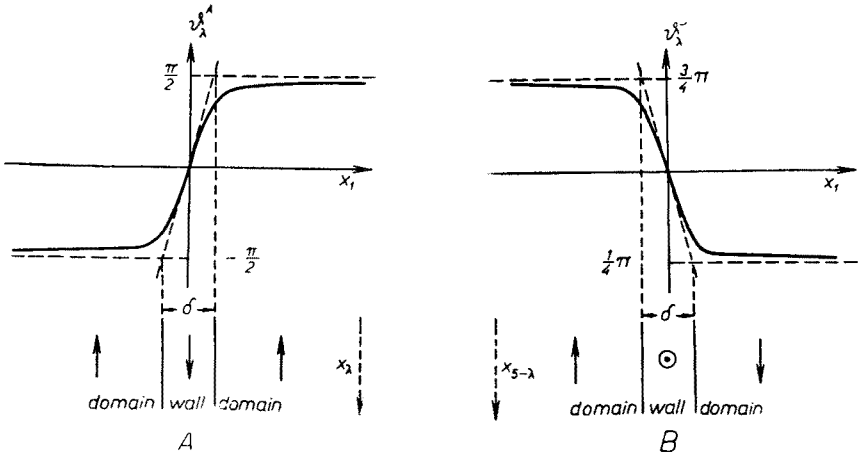

Fig. 3. Schematic curves and domain structures corresponding to asymptotic solutions (27A) (360° -wall, (A)) and (27B) (180° -wall, (B))

These results have an interesting interpretation when assuming the moduli of the coupling functions (14) monotonically to decrease with distance. This is certainly true whatever the correct form of those functions looks like, since the interaction between the elementary polarization vectors must weaken at larger and strengthen at smaller distances, at least in

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case of elastic lattice deformations. In this instance, however, inequality $c>b$ implies $u_0u_4<0$ and, conversely, $c<b$ implies $u_4u_0>0$ (provided the quadrupolar interaction is weaker than the dipolar one, or else opposite in sign). Hence, whether 360° or 180°-domains, in the first case they are polarized parallel to co-ordinate axis x_3 (c-axis), in the second parallel to x_2 (b-axis), thus perpendiculary to the c-axis. These effects are in agreement with experimental observations, as the spontaneous tetragonal lattice deformation of cubic ferroelectrics when passing the first transition temperature corresponds to the first effect (first phase transition of perovskites, $c>b=a$), and the rotation of the direction of easiest polarization from a parallel into a perpendicular position to the c-axis when pressing the crystal in the c-direction can be regarded as an examplification of the second effect (*cf.* [10, 11]). True, it is mostly the 180°-domains that are observed in those experiments, never 360° ones — as to our knowledge. Whether the latter domains are physically realizable at all is an open question. Although in ferromagnetic crystals this type of inter-domain walls certainly exists, or can be produced (by means of an external magnetic field), in ferroelectric crystals it possibly cannot be expected to appear because of the large local internal stresses that such a wall would produce.

Let us turn back to equation (21) and consider the other limit case, *i.e.*

$$2°:\ |u_4| \ll |u_8|.$$

Now, we put $u_4 = 0$ in equation (21) and maintain condition (22). To determine the constant C_λ in (21) we consider two types of boundary conditions for the value of the angle ϑ at infinity[2,3], namely,

$$\vartheta(x_1 = \pm\infty) = \{(2n+1)\pm 1\}\frac{\pi}{8}, \tag{30A}$$

$$\vartheta(x_1 = \pm\infty) = (2n\pm 1)\frac{\pi}{8} \tag{30B}$$

where $n = 0, \pm 1, \pm 2, \ldots$. According to whether (30A) or (30B) is assumed we get

$$C_\lambda = -u_8, \tag{31A}$$

$$C_\lambda = +u_8, \tag{31B}$$

and equation (21) becomes

$$\dot{\vartheta}^2 = \frac{u_8(\cos 8\vartheta - 1)}{u_0(\cos 2\vartheta - 1)}, \tag{32A}$$

$$\dot{\vartheta}^2 = \frac{u_8(\cos 8\vartheta + 1)}{u_0(\cos 2\vartheta - 1)}, \tag{32B}$$

[2] When putting $u_4 = 0$ in (21) one can drop the index λ as the type of the equation does not depend on the value of λ. The only difference resides in this case in the interpretation of the angle which may correspond either to Eq. (15a) or (15b) (*cp.* Fig. 2).

[3] The boundary conditions (23A) and (23B) can similarly be generalized, namely, $\vartheta_\lambda^A(x_1 = \pm\infty) = (2n\pm 1)\pi/2$ and $\vartheta_\lambda^B(x_1 = \pm\infty) = \{2(2n+1)\pm 1\}\pi/4$, respectively.

respectively. After some manipulations one obtains

$$\dot{\vartheta} = \pm \sqrt{u_8/u_0}\, \frac{\sin 4\vartheta}{\sin \vartheta}, \tag{33A}$$

$$\dot{\vartheta} = \pm \sqrt{-u_8/u_0}\, \frac{\cos 4\vartheta}{\sin \vartheta}, \tag{33B}$$

and thus the solutions

$$C' \exp\left(\pm 8x_1 \sqrt{-u_8/u_0}\right) = \left|\frac{\sin\vartheta + \frac{\sqrt{2}}{2}}{\sin\vartheta - \frac{\sqrt{2}}{2}}\right|^{\sqrt{2}} \left|\frac{\sin\vartheta - 1}{\sin\vartheta + 1}\right|, \tag{34A}$$

$$C' \exp\left(\pm 8x_1 \sqrt{-u_8/u_0}\right) = \left|\frac{2\cos\vartheta - \sqrt{2-\sqrt{2}}}{2\cos\vartheta + \sqrt{2-\sqrt{2}}}\right|^{\sqrt{2+\sqrt{2}}} \left|\frac{2\cos\vartheta + \sqrt{2+\sqrt{2}}}{2\cos\vartheta - \sqrt{2+\sqrt{2}}}\right|^{\sqrt{2-\sqrt{2}}} \tag{34B}$$

From Eqs. (30A) and (30B) it follows that the angles in the above solutions may vary within the intervals

$$n\pi/4 \leqslant \vartheta \leqslant (n+1)\pi/4, \tag{35A}$$

$$(2n-1)\frac{\pi}{8} \leqslant \vartheta \leqslant (2n+1)\frac{\pi}{8}, \tag{35B}$$

and hence the constants C' can be determined from the conditions

$$\vartheta(x_1 = 0) = (2n+1)\frac{\pi}{8}, \tag{36A}$$

$$\vartheta(x_1 = 0) = n\frac{\pi}{4}. \tag{36B}$$

However, when deeper examining the solutions and taking into account conditions (22) and equations (33A) and (33B) one easily verifies that certain intervals have to be excluded, *i.e.* n is not an arbitrary integer. It can be shown that n must be such as to fulfill the conditions[4]

$$n \neq 4k, \qquad n \neq (-1)^k(2k-1); \tag{37A}$$

$$n \neq 4k, \tag{37B}$$

where $k = 0, \pm 1, \pm 2, \ldots$. (See also Appendix.)

[4] Quite analogously, from Footnote 3 it follows that the intervals (28A) and (28B) can respectively be generalized in the following manner: $(2n-1)\frac{\pi}{2} \leqslant \vartheta_A^{\lambda} \leqslant (2n+1)\frac{\pi}{2}$ and $(4n+1)\frac{\pi}{4} \leqslant \vartheta_{\lambda}^{B} \leqslant (4n+3)\frac{\pi}{4}$, where $n = 0, \pm 1, \pm 2, \ldots$ as in the latter case the exclusion of not permitted intervals which follow from Eqs. (22), (26B) and (27B) is automatically ensured.

As seen from Eqs. (34A) and (34B) the first solution is real if $u_0 u_8 > 0$, and the second if $u_0 u_8 < 0$. Since the widths of the intervals (35A) and (35B) are identic and amount $\pi/4$, both solutions describe 90°-walls and correspond to the schematic curves and domain structures shown in Fig. 4 (*cp.* Fig. 2). The only difference is that in the first case the domains are polarized in the directions x_2 and x_3, *i.e.* along principle crystallographic axes (*cf.* Fig. 1), the inter-domain wall being polarized in a direction which bisects the angle between the polarization directions of adjacent domains (*i.e.* parallel to crystallographic directions of

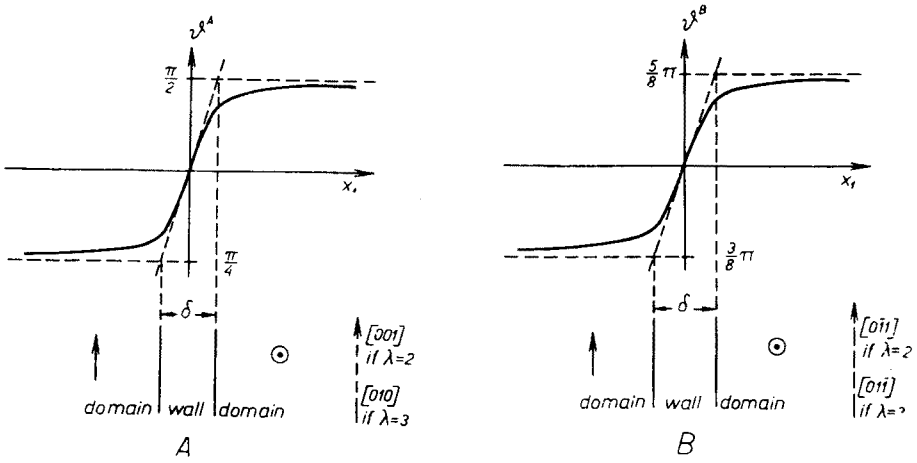

Fig. 4. Schematic curves and domain structures corresponding to asymptotic solutions (34 A, B) (90° —walls)

type [011] in a cubic lattice), whereas in the second case the situation is exactly opposite, that is domains are polarized in directions of type [011] (in reference to a cubic lattice) and walls in principle crystallographic directions of type [010] or [001] (*cp.* Fig. 2). Although this type of wall exists neither in ferroelectrics nor in ferromagnets and is a pure consequence of neglecting the influence of surface charges (*i.e.* neglect of electrostatic or magnetostatic self-energy of crystal, see [9—11, 19—23]), it is interesting in that it shows our approximation $u_4 = 0$ to be equivalent to the cubic-lattice approximation, and on the other hand, indicates the crystallographic directions of type [011] to be possible directions of easiest polarization (see also Appendix). Particularly the latter conclusion seems worthy to be emphasized as it suggests that more accurate calculations can reasonably be expected to permit a suitable description of the rotation of polarization directions when passing the second transition temperature in perovskites (tetragonal-orthorhombic phase transition) or compressing the ferroelectric crystal in the tetragonal phase along the fourfold axis (rotation of tetragonal axis). As in both the cases the second coefficient of Devonshires phenomenologic potential changes its sign (*cf.* [10, 11, 18]), and this coefficient must correspond to the quadrupolar coupling function in (10), thus also to u_8 (*cp.* Eq. (20)), the solutions (34A) and (34B) that are assigned to opposite signs of the product $u_0 u_8$ may quite well correspond to those effects mentioned above. A further argument in favour of our expectation can be found in [9] where the periodic solutions of a similar differential equation

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are discussed (*cf.* Appendix II in [9]; therein, the constants u_2 and u_4 correspond to the constants u_4 and u_8 used in the present paper).

It is to be noted that conditions (37A) and 37B) are purely mathematical ones and cause no physical restrictions when remembering that solutions (34A) and (34B) permit either choice of the specifications (15a) and (15b) of the generating vector (see Eqs. (31A) and (31B), *cp.* Footnote 2).

5. *Calculation of thickness and energy of inter-domain walls*

The thickness δ of an inter-domain wall can easily be calculated from the formula

$$\delta = \frac{|\vartheta(x_1 = +\infty) - \vartheta(x_1 = -\infty)|}{|\dot{\vartheta}(x_1 = 0)|}. \tag{38}$$

according to Fig. 3 and 4, and the energy σ per cm^2 of the wall is given by

$$\sigma = \frac{H_\vartheta - H_0}{L_2 L_3}, \tag{39}$$

where H_ϑ and H_0 denote respectively the value of the functional (16) after inserting the solutions of the Euler-Lagrange equation and $\vartheta(x_1 = -\infty)$ and evaluating the integral (*cf.* [7, 9, 20]).

By the aid of these formulae we shall calculate the thickness and the energy of the 180°-wall and 90°-wall. In the first case, we make use of Eqs. (22), (25B), (26B), (28B) and (29B) and arrive at the results

$$\delta_\lambda = \pi a [A_a(-1)^\lambda / 2\{3(A_b - A_c) + P^2(B_c - B_b)\}]^{\frac{1}{2}}, \tag{40}$$

$$\sigma_\lambda = \frac{P^2}{bc} [2A_a(-1)^\lambda \{3(A_b - A_c) + P^2(B_c - B_b)\}]^{\frac{1}{2}}, \tag{41}$$

when putting $u_8 = 0$ in (16) and utilizing Eqs. (18) and (19). In the second case we employ quite analogously Eqs. (22), (32A) or (32B), (33B) or (33B), (35A) or (35B), and (36A) or (36B), respectively, and take into account conditions (37A) and (37B). Thus, we obtain

$$\delta_A = \frac{\pi a \sqrt{2+\sqrt{2}}}{4P} \left[\frac{A_a}{B_b - B_c}\right]^{\frac{1}{2}}, \tag{42A}$$

$$\sigma_A = \frac{P^3}{bc} [A_a(B_b + B_c)]^{\frac{1}{2}} \left\{\frac{4}{15}(2+\sqrt{2}) + \frac{1}{8} [\ln (34A)]_{\frac{\pi}{4}}^{\frac{\pi}{2}}\right\} \tag{43A}$$

for the solution (34A) (*cf.* (30A), *et sq.*), and

$$\delta_B^{\text{even}} = \frac{\pi a}{2P} \left[-\frac{A_a}{B_b + B_c}\right]^{\frac{1}{2}}, \tag{42B-1}$$

$$\delta_B^{\text{odd}} = \frac{\pi a \sqrt{2}}{4P} \left[-\frac{A_a}{B_b + B_c}\right]^{\frac{1}{2}}, \tag{42B-2}$$

$$\sigma_B^{\text{even}} = \frac{P^3}{bc}\,[-A_a(B_b+B_c)]^{\frac{1}{2}}\left\{\frac{8}{15}\sqrt{2+\sqrt{2}} - \frac{1}{8}\,[\ln(34\text{B})]_{\frac{1}{8}\pi}^{\frac{3}{8}\pi}\right\}. \tag{43B-1}$$

$$\sigma_B^{\text{odd}} = \frac{P^3}{bc}\,[-A_a(B_b+B_c)]^{\frac{1}{2}}\left\{\frac{4}{15}\left(\sqrt{2+\sqrt{2}}+\sqrt{2-\sqrt{2}}\right) + \frac{1}{8}\,[\ln(34\text{B})]_{\frac{1}{8}\pi}^{\frac{3}{8}\pi}\right\} \tag{43B-2}$$

for the solution (34B) (*cf.* (30B), *et eq.*), in either case putting $u_4 = 0$ in (16) and making use of Eqs. (18) and (20). The symbols ln (34A) nad ln (34B) in Eqs. (43A), (43B-1) and (43B-2) denote logarithms of the right-hand sides of Eqs. (34A) and (34B), respectively; they results from the integration in Eq. (39) and have to be evaluated by inserting the indicated limits into the respective expressions[5].

The notation "even" and "odd" refers to the fact that in the "B"-case the value of the derivative (33B) at the point $x_1 = 0$ (which is needed in Eq. (38)) as well as the value of the integral (16) (see Eq. (39) and final result for σ_B given in Footnote 5) depend on whether the integer n in Eqs (36B) and (35B) is even or odd (note however the exclusion condition (37B)).

As we see from Eqs (40) and (41), the 180°-wall vanishes if the orthorhombic (or tetragonal) crystal lattice becomes cubic, at least in the $x_2 0 x_3$-plane, *i.e.* if $b = c$, since

$$\lim_{b\to c}\delta_\lambda = \infty, \qquad \lim_{b\to c}\sigma_\lambda = 0. \tag{44}$$

This can be related to the ferroelectric-paraelectric phase transition of perovskites [10, 11]. Besides, owing to λ both the quantities, δ_λ and σ_λ, can always be made real independent of whether $b>c$ or $b<c$. The only difference is in the direction of polarization of domains which is always parallel to the larger lattice constant (*cp.* discussion of solution (27B) in the preceding section), in agreement with experiment.

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[5] The evaluaton of the integral in (16) for the case of solution (27B) is quite elementary. As regards solutions (34A) and (34B), it is also simple when utilizing Eqs (32A), (33A) and (32B), (33B) in the following way:

$$\frac{anc}{P^2}\,\sigma_A = \int [u_0(\cos 2\vartheta - 1)\,\dot{\vartheta}^2 + u_8(\cos 8\vartheta + 1)]\,dx_1 = 2u_8\int \cos 8\vartheta\,dx_1 =$$

$$= 2\sqrt{u_0 u_8}\int \frac{\cos 8\vartheta \sin\vartheta}{\sin 4\vartheta}\,d\vartheta = 2\sqrt{u_0 u_8}\left[\int \frac{\sin\vartheta}{\sin 4\vartheta}\,d\vartheta - 2\int \sin 4\vartheta \sin\vartheta\,d\vartheta\right] =$$

$$= 2\sqrt{u_0 u_8}\left[\frac{1}{8}\ln(34\text{A}) - \frac{1}{3}\sin 3\vartheta + \frac{1}{5}\sin 5\vartheta\right],$$

$$\frac{abc}{P^2}\,\sigma_B = 2u_8\int \cos 8\vartheta\,dx_1 = 2\sqrt{-u_0 u_8}\int \frac{\cos 8\vartheta \sin\vartheta}{\cos 4\vartheta}\,d\vartheta =$$

$$= 2\sqrt{-u_0 u}\left[2\int \cos 4\vartheta \sin\vartheta\,d\vartheta - \int \frac{\sin\vartheta}{\cos 4\vartheta}\,d\vartheta\right] =$$

$$= 2\sqrt{-u u_8}\left[\frac{1}{3}\cos 3\vartheta - \frac{1}{5}\cos 5\vartheta - \frac{1}{8}\ln(34\text{B})\right].$$

As for the 90°-wall, we have three slightly different formulae for the thickness which is finite, and as many for the energy which due to the logarithms is infinite[6]. The latter result is rather surprising as for the same type of wall we obtained in [9] finite values for the energy.

Since in [9] we chose the rotating angle φ as variational parameter (see Sections 1 and 2 of the present paper), it proves that the choice of variational parameters can have influence upon the numerical results. Notwithstanding this difficulty one can attain to finite values by introducing sort of "renormalized" energy, *i.e.* by simply subtracting the logarithms from the expressions (43A), (43B-1) and (43B-2). The respective remainders are then finite and can be interpreted as the measurable energies. However, even then we have for the case "B" (*i.c.* domains polarized in directions of the type [011], see Fig. 4 and discussion at end of Section 4) two formulae for the thickness as well as the energy of the wall, the "even" and the "odd" one. One easily proves, by taking into account Eqs. (33B), (35B) and (37B), that only for the "even" case solution (34B) is symmetric (note condition (36B)). Thus, when choosing the symmetric solution for the case $u_0 u_8 < 0$ (*i.c.* sign $A_a = -$ sign $(B_b + + B_c)$, *cf.* Eqs. (18), (20) and (34B)) we have for the thickness of the wall the formula (42B-1) and for the renormalized energy, instead of (43B-1),

$$\sigma_B^{\text{even/ren}} = \frac{8P^3\sqrt{2+\sqrt{2}}}{15bc}\,[-A_a(B_b+B_c)]^{\frac{1}{2}}. \tag{45}$$

Moreover, beside (37B) we have the additional condition

$$n \neq 2k+1. \tag{46}$$

Analogously, the renormalized energy that follows from (43A) for the case $u_0 u_8 > 0$ reads

$$\sigma_A^{\text{ren}} = \frac{4P^3(2+\sqrt{2})}{15bc}\,[A_a(B_b+B_c)]^{\frac{1}{2}}. \tag{47}$$

Now, when calculating the thickness $\delta_\varphi^{\text{cubic}}$ and the energy $\sigma_\varphi^{\text{cubic}}$ of such 90°-walls by means of the "φ-representation" (see Eqs. (49), (50), (62), (66), (70) and (85) in [9] where these quantities are denoted by δ_0 and $2\sigma_0$, respectively) we get for the cubic lattice

$$\delta_\varphi^{\text{cubic}} = \frac{\pi a}{2P}\,|AB|^{\frac{1}{2}}, \tag{48}$$

$$\sigma_\varphi^{\text{cubic}} = \frac{2P^3}{a^2}\,|AB|^{\frac{1}{2}}, \tag{49}$$

[6] This may readily be shown either by inserting the respective integration limits into Eqs. (34A) and (34B), or, even simpler, by suitably changing certain integrals when calculating the energy (39). For instance, when calculating σ_A according to Footnote 5 one can write

$$\int \frac{\sin\vartheta}{\sin 4\vartheta}\,d\vartheta = \sqrt{u/u_0}\int dx_1$$

because of Eq. (33A), or

$$\ln\,(34\text{A}) = 8x_1\sqrt{u/u_0} + \text{const}$$

by virtue of Eq. (34A). As $-\infty \leqslant x_1 \leqslant +\infty$, both the expressions are evidently divergent.

for either type of 90°-wall[7]. Here, A and B denote respectiv elythe dipolar and quadrupolar coupling constants of the cubic dipole-lattice (*cf.* [9]). In contradistinction to Eqs. (48) and (49), Eqs. (42A) and (47) do not coincide with Eqs. (42B-1) and (45) when passing to the cubic-lattice case, *i.e.* when putting $a = b = c$, $A_a = A_b = A_c \equiv A$, $B_a = B_b = B_c \equiv B$. Instead we have

$$\sigma_{\varphi}^{\text{cubic}} > \sigma_{B}^{\text{even/ren/cubic}} > \sigma_{A}^{\text{ren/cubic}}, \tag{50}$$

$$\delta_{\varphi}^{\text{cubic}} > \delta_{B}^{\text{even/cubic}} > \delta_{A}^{\text{cubic}}, \tag{51}$$

$$\sigma_{\varphi}^{\text{cubic}} : \sigma_{B}^{\text{even/ren/cubic}} : \sigma_{A}^{\text{cubic}} = 15 : 4\sqrt{4+2\sqrt{2}} : 4(1+\sqrt{2}), \tag{52}$$

$$\delta_{\varphi}^{\text{cubic}} : \delta_{B}^{\text{even/cubic}} : \delta_{A}^{\text{cubic}} = 2\sqrt{2} : 2 : \sqrt{2+\sqrt{2}}, \tag{53}$$

$$\sigma_{B}^{\text{even/ren/cubic}} = 0.7\sigma_{\varphi}^{\text{cubic}}, \quad \sigma_{A}^{\text{ren/cubic}} = 0.65\sigma^{\text{cubic}}, \tag{54}$$

$$\sigma_{B}^{\text{even/cubic}} = 0.7\sigma_{\varphi}^{\text{cubic}}, \quad \sigma_{A}^{\text{cubic}} = 0.65\sigma_{\varphi}^{\text{cubic}}, \tag{55}$$

Eqs. (54) and (55) being approximate.

There are three conclusions following from our results which are worth while emphasizing. First, the thickness as well as the (renormalized) energy of the 90°-wall as actually calculated are smaller than the respective values obtained in [9]. Second, these quantities actually differ according to whether the domains are polarized in crystallographic directions of type [001] ($AB>0$) or [011] ($AB<0$), and, third, they are smaller in the former case. All the conclusions are comprised in inequalities (50) and (51). With respect to experimental data they have to be considered as rather satisfactory for the numerical value ofr the energy of the wall as roughly estimated in [9] was still pretty high (265 erg/cm^2).

It must be noted, however, that it is hardly the different choice of variational parameters that might explain the difference between the present results and those obtained in [9]. Instead, it is rather the additional cosine-series-approximation one usually is forced to do when working in the "φ-representation" that should solely be responsible for this discrepancy (*cf.* Eq. (36) in [9]). Thus it would seem reasonable to draw the conclusion that the "e_{μ}-representation" as demonstrated in the present paper is more advantageous, to some extent, than the "φ-representation" for it permits to derive the variational principle with a better approximation and thus to seize more subtle effects, in spite of the approximate solution of the Euler-Lagrange equation (21) (*i.e.* when either $u_4 = 0$ or $u_8 = 0$ can be assumed). On the other hand, it must not be forgotten that the (less accurate) variational principles derived by means of the "φ-representation" lead to Euler-Lagrange equations that often can rigorously be solved, even when imposing periodic boundary conditions [3—9, 23]. Hence, which of the two representations is to be decided for depends upon the problem under consideration, and the proper choice requires every time some preliminary analysis.

[7] The coefficients u_0 in the present paper and in [9] are exactly the same, while the coefficient u_8 used in the present paper corresponds to the coefficient u_4 in [9]. Furthermore, to simplify the notation we actually write P instead of εP which means that P is to be understood as the elementary polarization of the given material, whereas in the notation εP it denotes Debye's elementary polarization unit (see Eqs. (2), (3), (6), and (85) in [9]).

6. *Final remarks*

Our examination proves that in some cases the description of the domain structure can be more efficient, at least with respect to certain effects, when choosing the direction cosines e_μ as variational parameters and putting the rotating angle $\varphi = \pi$. Although we are concerned with a microscopic formalism, this may also be true in case phenomenological methods are employed as the basic idea which consists in introducing rotational parameters is in both the approaches identic. However, it is also true that the differential equations one arrives at in the "e_μ-representation" are usually more complicated than those obtainable in the "φ-representation"[8], their integration being therefore much more difficult if at all possible, particularly when periodic solutions are aimed at. For example, it is difficult to find the general non-periodic solution of Eq. (21). Were we able to do it we would have a solution that in the limit cases $u_8 \to 0$ or $u_4 \to 0$ should coincide with solutions (27A), (27B) or (34A), (34B), respectively. Such a solution would describe the transition, for instance, from 90°-walls to 180°-walls as a consequence of a continuous tetragonal deformation of the cubic dipole-lattice. From this point of view, the less accurate "φ-representation" happens to be favourable in so far as it much easer enables a suitable description of such processes (*cf.* [9]).

APPENDIX

The boundary conditions (23A) and (23B) (see also Footnote 3) are by far not the only ones that can be imposed upon Eq. (21) when putting $u_8 = 0$. On the contrary, there are infinite many asymptotic boundary conditions for which solutions of this simplified differential equation do exist, although they have to be chosen according to a certain rule if imaginar solutions are excluded. So for instance the boundary conditions

$$(2n-1)\frac{\pi}{8} \leqslant \vartheta_\lambda \leqslant (2n+1)\frac{\pi}{8}, \tag{56}$$

when imposed upon the simplified Eq. (21), *i.e.*

$$\dot{\vartheta}^2 = \frac{(-1)^\lambda u_4 \cos 4\vartheta_\lambda + C_\lambda}{u_0(\cos 2\vartheta_\lambda - 1)}, \tag{57}$$

imply

$$C_\lambda = 0, \tag{58}$$

as condition (22) still holds. Hence, Eq. (57) becomes

$$\dot{\vartheta}_2^\lambda = (-1)^\lambda (u_4/u_0)\frac{2\sin^2 2\vartheta_\lambda - 1}{2\sin^2 \vartheta_\lambda}. \tag{59}$$

Because of the denominator in Eq. (59), not all the intervals (56) are permissible. The integer n in (56) must obey the exclusion condition (37B). If, moreover, we demand the solution

[8] For the general form of these equations see [1].

to be symmetric further intervals are interdicted, according to condition (46). In this case $\lambda = 2$ provides real solutions for $u_0 u_4 < 0$, and $\lambda = 3$ for $u_0 u_4 > 0$.

One easily verifies that Eq. (59) though differing from Eq. (32B) describes 90°-walls of similar character, *i.e.* the adjacent domains are polarized in crystallographic directions of the type [011] (*cf.* Eqs. (15a), (15b), (37B), (46), (56) and (59), and Fig. 2 and 4 (B)).

Similarly, it can be proved that boundary conditions of the type

$$\frac{1}{3}\pi \leqslant \vartheta \leqslant \frac{2}{3}\pi, \tag{60}$$

when imposed upon Eq. (57) imply $2C_\lambda = (-1)^\lambda u_4$ and lead to 120°-walls, boundary conditions of the type

$$\frac{5}{12}\pi \leqslant \vartheta_\lambda \leqslant \frac{7}{12}\pi, \tag{61}$$

imply $2C_\lambda = (-1)^{\lambda+1} u_4$ and lead to 60°-walls, boundary conditions

$$\frac{7}{16}\pi \leqslant \vartheta_\lambda \leqslant \frac{9}{16}\pi, \tag{62}$$

imply $2C_\lambda = (-1)^{\lambda+1} u_4 \sqrt{2}$ and lead to 45°-walls, and so on. For all those intervals condition (22) is automatically fulfilled, and the respective solutions will be symmetric.

The rule of selecting the permissible intervals is rather simple. Let $\vartheta_\lambda(x_1 = -\infty) \equiv \vartheta_\lambda^-$ and $\vartheta_\lambda(x_1 = +\infty) \equiv \vartheta_\lambda^+$. Then, ϑ_λ^- and ϑ_λ^+ must be such as to satisfy the following conditions:

i)
$$0 < \left(\frac{\pi}{2} - \vartheta_\lambda^-\right) = \left(\vartheta_\lambda^+ - \frac{\pi}{2}\right) < \frac{\pi}{2}, \tag{63}$$

ii)
$$\cos 4\vartheta_\lambda^- = \cos 4\vartheta_\lambda^+, \tag{64}$$

iii)
$$\operatorname{sign}(\cos 4\vartheta_\lambda - \cos 4\vartheta_\lambda^+) = \text{const} \quad \text{for } \vartheta_\lambda^- < \vartheta_\lambda < \vartheta_\lambda^+, \tag{65}$$

as $C_\lambda = (-1)^{\lambda+1} u_4 \cos 4\vartheta_\lambda^+$, according to Eqs. (22) and (57). The angle between directions of polarization of adjacent domains amounts $2(\vartheta_\lambda^+ - \vartheta_\lambda^-)$, and this angle determines the type of wall (*cf.* Fig. 2). The generalization of these conditions beyond the interval $(0, \pi)$ is quite obvious. Of course, the above conditions are ascribed to Eq. (57) and subject to proper modification according to the differential equation to be solved.

It is thus clear that boundary conditions (35A) and (35B), too, are particular ones when examining Eq. (21) in the other limit case, that is when putting $u_4 = 0$, and that the full set of admissible boundary conditions can similarly be selected as in the preceding case.

Hence, we see that, in general, the Euler-Lagrange equation has a great variety of solutions which all resemble (asymptotic or periodic) domain structures whose domains are polarized in directions that are determined by the boundary conditions imposed upon the problem. True, these conditions are not quite arbitrary, and the variety of domain-struc-

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ture-resembling solutions is thus somehow restricted. None the less, there will still be (usually infinitely many) different domain structures that are described by a single differential equation. This important fact seems to be permanently overlooked by many authors dealing with domain-structure investigations theoretically [10—17, 20, 21].

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