Magnetostriction and magnetoelastic domains in antiferromagnets

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(October 25, 2018)

The problem of the observable equilibrium domain structure in pure antiferromagnets (and other thermoelastics) is investigated with the use of continuous elasticity theory. It is shown that completely rigid surface produces the imaginary “incompatibility elastic charges” analogous to the surface magnetic charges in ferromagnets. Corresponding long-range field is shown to contribute into the “stray” energy of the sample that governs an appearance of the domain structure, the contribution from the “elastic charges” being proportional to the sample volume. Competition between the elastic “stray” field that favors inhomogeneous strain distribution, and external field that tends to homogenize the sample, provides the reversible reconstruction of the domain structure under the action of external magnetic field.
I. INTRODUCTION

The problem of antiferromagnetic (AFM) domains has a very long history and was discussed for many times (see, e.g., Ref. [3]). This problem is tightly related with the question of the domain structure (DS) origin in the substances that suffer thermoelastic phase transitions, i.e., transitions at which the principal order parameter is symmetrically conjugated with the shear components of the strain tensor. The transitions of such a kind are usually accompanied by appearance of the spontaneous strains along with the simultaneous change of the electronic structure (martensitic phase transitions) or onset of the vector or tensor order parameter of other nature (like magnetization, polarization, etc.). In the case when the values of the principal order parameter and/or corresponding strain tensor of low-symmetry (i.e., magnetic, ferrielectric, martensitic) phase are degenerated, the DS is thermodynamically equilibrium and reversible under the action of the external fields that can result in homogenization of the sample. The nature of the equilibrium DS of ferro- and ferrimagnets with opposite direction of magnetization inside the neighboring domains is well studied and is usually related with the finite size of the sample and demagnetization effect resulting from the surface magnetic charges. So, the question is “What is the reason of the clearly observed equilibrium DS in the cases when the noncompensated magnetization is equal to zero or vanishingly small (e.g., in weak ferromagnets)?” In the Ref. [3] we have supposed that one of the possible mechanism of equilibrium DS formation can be due to the surface effects, namely, the surface energy decrease yielded by the global variation of the sample shape. Although such a model has given qualitative and quantitative agreement with the magnetostriction experiments, its application, as it is evident from theoretical point of view, is restricted to the rather small samples for which the surface effects can be clearly detected.

In the present paper we make an attempt to generalize our previous results and to show that the formation of equilibrium DS in the course of thermoelastic phase transition can be explained by the effect of low-deformable sample surface, and what is principal, corresponding contribution into the sample energy being proportional not to the surface area but to the sample volume.

II. MODEL

Let us consider a sample with the volume \( V \) confined with the surface \( S \). Suppose, that above the critical temperature the sample is non-stressed and non-strained. Below the critical temperature the crystal lattice becomes unstable with respect to appearance of the spontaneous strains related with the fluctuations of the order parameter (e.g., magnetization, polarization, etc.). The spontaneous strains can result from the nonlinear elastic effects, as in the case of martensites, as well as be related with the appearance of spontaneous AFM moment, both cases allow no demagnetization effects.

In the infinite sample the only equation that defines distribution of spontaneous strain tensor \( \hat{u}(r) \) (\( r \) is a space coordinate) arises from the condition of the elastic free energy minimum with respect to \( \hat{u} \):

\[
\frac{\partial F_{el}(\hat{u})}{\partial \hat{u}(r)} = 0, \quad (1)
\]

where \( F_{el}(\hat{u}) \) is the free energy density. Equation (1) is equivalent to the condition of the absence of internal forces: \( \nabla \hat{\sigma}(r) = 0 \), where \( \hat{\sigma} \) is a stress tensor. Elastic free energy density \( F_{el}(\hat{u}) \) is usually independent upon the space derivatives of the strain tensor; so, the equilibrium strain is homogeneous and thus trivially satisfies the compatibility condition. In approximation of linear elasticity takes a form:

\[
\text{inc } \hat{u}(r) \equiv -\text{rotrot } \hat{u}(r) = 0. \quad (2)
\]

If we take into account the surface of the sample, given parametrically by equation \( r = r_S \), the situation changes in a crucial way. The elastic properties of the sample surface differ from that of the bulk, and homogeneous strain defined from (1), inevitably brings about the additional stresses in the sample. In formal, the surface can be accounted for through the surface energy term:

\[
F_{\text{surf}} = \int dS \left[ \alpha_S + \hat{\beta}_S \hat{u}(r_S) \right], \quad (3)
\]

where \( \alpha_S \) is a surface tension coefficient, \( \hat{\beta}_S \) is the tensor of the surface elastic modula, and \( \hat{u}(r_S) \) is a surface strain tensor. It should be emphasized that the first term in (3) is responsible for the set up of the equilibrium shape of the sample (it establishes the minimum surface area at given volume), the second term can be important when considering thermoelastic phase transition.

In what follows we will suppose that the shape of the sample is given technologically and does not relax during the time of experiment, and so, will disregard an influence of the surface tension (3). In other words, we consider the energy barrier for shape variation in the strained sample as high enough, so, that corresponding relaxation time is much greater than the inverse energies characteristic to the phase transition. This is the case for AFM transitions at low temperature and also for martensitic phase transitions that usually proceed with the rate close to sound velocity.

In the vicinity of the surface the spontaneous strains that occur during the thermoelastic phase transition can be represented as a sum of three summands:

\[
\hat{u}(r) = \frac{1}{3} \text{Tr}\hat{u} + \hat{u}_{\text{shear}} + \left\{ (n_S \hat{u}) \otimes n_S + n_S \otimes (\hat{u} n_S) - n_S \otimes n_S (n_S \hat{u} n_S) \right\}, \quad (4)
\]
where \( \mathbf{1} \) is a unit matrix, \( \mathbf{n}_S \) is a surface normal. The first term in (1) is an isotropic volume striction, the last three terms in brackets form the component that produces the shift along the surface normal \( \mathbf{n}_S \), and the second, traceless, term corresponds to the shear strains (with respect to \( \mathbf{n}_S \)). The first component is significant at the magnetic phase transitions and can be neglected during the martensitic ones, but at any rate it does not contribute into the symmetry change of the crystal and hence, as well as the last term, could not be removed by onset of DS in the sample. So, in what follows, we consider only the shear part of the strain tensor \( \hat{u}_{\text{shear}} \) and define the strain state with respect to isomorphous magnetostriction which contribution can be taken into account trivially.

If one assumes that the sample surface is not deformed during the phase transition, i.e., \( \hat{\beta}_S = 0 \), we should impose the standard boundary conditions for zero external stress

\[
\left\{ \frac{\partial F_{el}(\hat{u})}{\partial \hat{u}} \right\}_{r \in S} \mathbf{n}_S(\mathbf{r}_S) = 0, \tag{5}
\]

and additional conditions of the absence of surface strains, i.e.

\[
\mathbf{n}_S(\mathbf{r}_S) \times \hat{u}(\mathbf{r}_S) \times \mathbf{n}_S(\mathbf{r}_S) = 0, \tag{6}
\]

that can be easily deduced from the definition of the tangent component of the strain tensor. In (5), (6) \( \mathbf{n}_S(\mathbf{r}_S) \) is the surface normal at a given point.

Thus, the problem of equilibrium strain distribution in the definite-shaped sample below the phase transition temperature should be reformulated as follows: equations (5), (6) should be satisfied inside the sample volume for \( r \in V \setminus S \), and equations (3), (4) are fulfilled at the sample surface, \( r = r_S \in S \).

It is obvious that for the symmetry-governed transition, onset of homogeneous strain inevitably breaks the condition (6). The only exceptions being the cases of a rather thin disk- or plate-shaped sample with the plate surface coinciding with the homogeneous shear plane (8).

### III. “STRAY” ENERGY

The problem of the equilibrium DS in AFM can be formally solved in analogy with the well known problem of the domain distribution in ferromagnets; namely, we assume that equilibrium (and in general, inhomogeneous) shear strain of the sample that complies with the condition (6) consists of two parts:

\[
\hat{u}_{\text{shear}}(\mathbf{r}) = \hat{u}_{\text{ms}}(\mathbf{r}) + \hat{u}_{\text{ch}}(\mathbf{r}), \tag{7}
\]

where the first term, \( \hat{u}_{\text{ms}}(\mathbf{r}) \), corresponds to the eigen spontaneous magnetostriction-induced strain defined from the free energy minimum (1); and the second term, \( \hat{u}_{\text{ch}}(\mathbf{r}) \) is an additional strain field produced by the “incompatibility elastic charges” localized at the crystal surface with the density:

\[
\hat{e}_{\text{elas}}(\mathbf{r}) = -\mathbf{n}_S \times \hat{u}_{\text{ms}}(\mathbf{r}_S) \times \mathbf{n}_S \delta(\mathbf{n}_S(\mathbf{r} - \mathbf{r}_S)), \tag{8}
\]

where \( \delta \) is the Dirac’s delta-function, prime means derivative vs argument. Function \( \hat{u}_{\text{ch}}(\mathbf{r}) \) can be found explicitly from equation

\[
\text{inc} \ \hat{u}_{\text{ch}}(\mathbf{r}) = \hat{e}_{\text{elas}}(\mathbf{r}) \tag{9}
\]

as follows (for the details see Ref. (4))

\[
\hat{u}_{\text{ch}}(\mathbf{r}) = \frac{1}{4\pi} \int_V d\mathbf{r}_1 \hat{e}_{\text{elas}}(\mathbf{r}_1) - \hat{e}_{\text{elas}}(\mathbf{r}) = \frac{1}{4\pi} \int_S dS \left( \mathbf{n}(\mathbf{r} - \mathbf{r}_S) \cdot \hat{\mathbf{U}}(\mathbf{r}_S) \right) = \frac{1}{4\pi} \int d\Omega_\mathbf{r} \hat{\mathbf{U}}(\mathbf{r}_S), \tag{10}
\]

where

\[
\hat{\mathbf{U}}(\mathbf{r}) = \hat{u}_{\text{ms}}(\mathbf{r}) + \mathbf{n}_S \otimes \mathbf{n}_S \text{Tr} \hat{u}_{\text{ms}}(\mathbf{r}) - \mathbf{n}_S \otimes (\hat{u}_{\text{ms}}(\mathbf{r}) \mathbf{n}_S) - (\hat{u}_{\text{ms}}(\mathbf{r}) \mathbf{n}_S) \mathbf{n}_S.
\]

and \( d\Omega_\mathbf{r} \) is an increment of solid angle at which the surface point \( \mathbf{r}_S \) is seen from the given point \( \mathbf{r} \). Strain \( \hat{u}_{\text{ch}}(\mathbf{r}) \) produces the “twinning” stress

\[
\hat{\sigma}(\mathbf{r}) = \hat{c} \hat{u}_{\text{ch}}(\mathbf{r}), \tag{11}
\]

completely analogous to demagnetization field in ferromagnets. In (11) \( \hat{c} \) is the tensor of the elastic modula. Equilibrium strain distribution conditioned by nondeformed surface can thus be found from equation:

\[
\frac{\partial F_{el}(\mathbf{r})}{\partial \hat{u}(\mathbf{r})} = \hat{\sigma}(\mathbf{r}), \tag{12}
\]

that combines (1) and (11). It is easy to see that (12) can be also deduced from variation of free energy functional with respect to the strain tensor components that satisfy compatibility conditions (6) inside the sample (4).

\[
\Phi = \int_V d\mathbf{r} F_{el}(\mathbf{r}) - F_{\text{stray}}, \tag{13}
\]

where

\[
F_{\text{stray}} = \frac{1}{4\pi} \int_V d\mathbf{r} \int_S dS \left( \mathbf{n}(\mathbf{r} - \mathbf{r}_S) \cdot \hat{\mathbf{U}}(\mathbf{r}_S) \right) \hat{u}(\mathbf{r}) \hat{\mathbf{U}}(\mathbf{r}_S) \tag{14}
\]

is a twinning (stray) energy that was not considered in Ref. (4).

The above-developed approach makes it possible to solve the complicated problem of the domain distribution in the nonlinear media under the action of external field in terms of strain tensor which in contrast to the
shift vector is observable and symmetrically related with an order parameter.

To continue the analogy with the problem of ferromagnetic domains, we should emphasize some properties of the twinning (stray) fields in thermoelastics. First, as it is clearly seen from (10), “incompatibility charges” can produce homogeneous additional strain \( \hat{u}_{\text{ms}} \) and thus, a macrostress in the case of appropriate form of the sample. What is important, the energy contribution resulting from the macrostress is proportional to the sample volume \( V \) and thus is principal, regardless of sample size. On the contrary to ferromagnets, where demagnetization charges are the scalars, in the case of elasticity, the charges (see (6)) possess the tensor characteristics, so, the only obvious case for homogeneous twinning stress is the thin plate. This case will be considered in details below. The other interesting feature of the additional strain field (10) is that it is scaling-invariant. In other words, additional strain distribution inside the sample depends only upon the angle at which the surface is seen from the given point. So, isomorphic transformation of the sample does not change the additional strain distribution.

From the above considerations we can make rather general conclusion: in the case of temperature-induced phase transition the macroscopic symmetry of low- and high temperature phases is the same. If the transition is symmetry-breaking (at the microscopic scale), i.e. the microscopic order parameter is conjugated with nonisomorphous striction and produces the strains that locally reduce the symmetry of the crystal lattice, then, the macroscopic symmetry is restored due to the onset of the DS with the differently oriented strain tensors.

IV. EQUILIBRIUM DOMAIN STRUCTURE

Indeed, expressions (13) and (14) show that the shape of the sample essentially affects the strain distribution below the phase transition point. The simplest way to elucidate this point is to consider the case of thin plate below the phase transition point. The simplest way to elucidate this point is to consider the case of thin plate for which the additional strains and twinning stresses are homogeneous. Suppose, the plate surface is \( n_S \)-oriented with respect to crystal axes. If the plate normal \( n_S \) is directed along the principal symmetry axis \( C_n \) of the sample \( (n=3, 4, \text{ or } 6) \) then, the macroscopic symmetry of the sample must be restored below the phase transition point and all the types of the domains should be equally represented.

The above calculations convincingly show that appearance of the homogenous deviatoric (shear) strains in the finite-size sample can give rise to a considerable energy increase and thus are non-advantageous. There is a close analogy between the appearance of the long-range elastic fields in thermoelastics and dipole fields in ferromagnets (a similarity between the equations for electro-magnetic fields in substances and equations of elasticity theory was long ago noticed by R. de Wit in Ref. 16). The case of the weak ferromagnets needs the special treatment because the DS in corresponding compounds can be formed due to competition of demagnetization and twinning factors. This problem is out of scope of this paper.

The general expression (18) can also be used for the description of the martensitic phase transitions with the only corrections concerning adjustable habitus (zero-shift) plane instead of fixed sample surface.

Another important question that arises while analyzing the expression (18) is whether onset of twin (domain) structure is indeed thermodynamically advanta-
geous. The matter is that the inhomogeneous strain distribution gives rise to an increase of the full free energy due to the contribution of the domain walls. In order to show that the energy gain because of the twinning is greater than the loss from the domain walls contribution, let us follow the method applied for the ferromagnets and consider the DS consisting of two alternating types of the domains, characterized with the strain tensors \( \hat{u}_1 \) and \( \hat{u}_2 \), the period of the structure \( d_{DS} \) is much less then the plate thickness, \( d \). We suppose that the domains are coherently conjugated with each other and go out to the surface with the same strains that in the bulk.

Standard calculations based on the formulae (13) and (14) give rise to the following contribution into free energy from the “elastic charges”:

\[
F_{\text{stray}} = L^2 d \left\{ \langle \hat{u}_{\text{ms}} \rangle \hat{c} \langle \hat{u}_{\text{ms}} \rangle + n_S \otimes n_S \text{Tr} \langle \hat{u}_{\text{ms}} \rangle - n_S \otimes (\langle \hat{u}_{\text{ms}} \rangle n_S) - (\langle \hat{u}_{\text{ms}} \rangle n_S) \otimes n_S \right\}.
\] (17)

If we restrict ourselves with the shear strains \( \hat{u}_{\text{ms}} = \hat{u}_{\text{shear}} \), as it was assumed above (see (4) and (5)), then the stray energy takes a form

\[
F_{\text{stray}} = L^2 d \langle \hat{u}_{\text{ms}} \rangle \hat{c} \langle \hat{u}_{\text{ms}} \rangle \geq 0,
\] (18)

that explicitly reveals it positivity for any choice of shear strain. In analogy with the case of ferromagnets, in the absence of external field the stray energy \( F_{\text{stray}} \) can only be reduced by zeroing of the average strains. Expression (18) coincides with the expression (2) of Ref. 9 written previously from the phenomenologic considerations only.

The value of \( F_{\text{stray}} \) obviously depends upon the shape of the sample, in our case, from the orientation of plate surface with respect to crystal axes. If the plate normal \( n_S \) is directed along the principal symmetry axis \( C_n \) of the sample \( (n=3, 4, \text{ or } 6) \) then, the macroscopic symmetry of the sample must be restored below the phase transition point and all the types of the domains should be equally represented.

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Standard calculations based on the formulae (13) and (14) give rise to the following contribution into free energy from the “elastic charges”:...
\[ F_{ch} = L^2 d_{DS} \xi_1 \xi_2 (\hat{u}_1 - \hat{u}_2) \hat{c} (\hat{u}_1 - \hat{u}_2) \cos \vartheta, \]  
(19)  

where \( \vartheta \) is the angle between the plate surface and domain (twin) interface, \( \xi_1 (1 - \xi_2) \) is the volume fraction of the domain of the first type.

Domain walls (interfaces) also contribute into the free energy, corresponding expression is:

\[ F_{DW} = L^2 \sigma_{DW} \frac{d}{d_{DS}}, \]  
(20)  

where \( \sigma_{DW} \) is the surface energy of a single domain wall.

Comparison of equations (17), (18) and (19) shows that optimal DS period:

\[ d_{DS}^{opt} = \frac{\sqrt{\sigma_{DW} d}}{\sqrt{\xi_1 \xi_2 (\hat{u}_1 - \hat{u}_2)^2 \hat{c} (\hat{u}_1 - \hat{u}_2) \cos \vartheta}} \approx \sqrt{\frac{\sigma_{DW} d}{F_{el}}}. \]  
(21)  

So, contribution from the inhomogeneous part of strains and short-range periodic “charge” distribution is proportional to the ratio \( d_{DS} / d \ll 1 \) and is much less than the stray energy (17) that is proportional to the sample volume. This means that onset of the DS in thermoelastics crucially diminishes the free energy from the “elastic charges” localized on the sample surface, the energy increase related with the inhomogeneous strain distribution is much smaller and cannot compensate the energy related with homogenization of the domain structure for rather large samples.\[ \]  

V. CONCRETE EXAMPLES

To elucidate the consequences of the stray energy, let us consider a case of plate-shaped easy-plane pure AFM with the plane normal directed along the principal crystal axis. A good example of such an AFM is given by CoCl\(_2\) (symmetry group is \( D_{4d} \)) and low-doped YBa\(_2\)Cu\(_3\)O\(_{6+\delta}\) at \( x \leq 0.3 \) (symmetry group is \( D_{4h} \)) single crystals. Corresponding contribution into stray energy (17) is:

\[ F_{stray} = L^2 d \left\{ \frac{1}{2} c_{11} (u_{xz})^2 + (u_{yy})^2 \right\} + c_{12} (u_{xz})(u_{yy}) + 2 c_{66} (u_{xy})^2, \]  
(22)  

where \( z \)-axis is supposed to be directed along the plate normal. Full free energy with the account of the magnetic subsystem in the external magnetic field \( H \) (neglecting the small demagnetization effects) could be written as follows:

\[ F = L^2 d \sum_k \left\{ \xi_k [E_{an} + M_0 \frac{H}{2H_E} (H \hat{k})^2 + \hat{u}_k \lambda_{me} l_k \otimes l_k + \frac{1}{2} \hat{u}_k \hat{c} \hat{u}_k] \right\} + F_{stray}, \]  
(23)  

where \( E_{an} \) is the magnetic anisotropy energy, \( M_0 \) is saturation magnetization, \( 2H_E \) is the value of spin-flip field of exchange nature, \( l_k \) and \( \hat{u}_k \) are the vector of AFM and spontaneous magnetostriiction, respectively, in the \( k \)-th domain \( (k=1, 2, 3 \) for CoCl\(_2\) and \( k=1, 2 \) for YBa\(_2\)Cu\(_3\)O\(_{6+\delta}\)), \( \xi_k \) is the volume fraction of the \( k \)-th domain, \( \lambda_{me} \) the 4-rank tensors of magnetoelastic coefficient, the strain tensor is averaged over the sample volume as follows:

\[ \langle \hat{u} \rangle = \sum_k \xi_k \hat{u}_k. \]  

Minimization of the energy (23) with respect to components of AFM vectors, strain tensor and also to domain fraction \( \xi_k \) proves that due to redistribution of the domains, the effective magnetic field inside sample is zero until the external field attains the value

\[ |H| = H_{MD} \equiv \lambda_{eff} M_0 \sqrt{\frac{H_E M_0}{c_{eff}}}. \]  
(24)  

where \( \lambda_{eff} \) and \( c_{eff} \) are certain combinations of magnetoelastic and elastic constants that should be calculated with the account of concrete symmetry of the crystal. The value \( H_{MD} \) can be associated with the field of monodomenization of the sample.\[ \]

Below the filed of monodomenization the averaged shear strain

\[ \langle u_{shear} \rangle = \frac{\lambda_{eff} M_0^2}{c_{eff}} \left( \frac{H}{H_{MD}} \right)^2 = u_0 \left( \frac{H}{H_{MD}} \right)^2. \]  
(25)  

follows the quadratic field dependence normalized to the factor of monodomenization field value. In (25) \( u_0 \) describes the absolute value of the local spontaneous shear. Thus, the low-field dependence of any macroscopic property which depends upon the relative fraction of the elastic domains should follow the law of the “corresponding states”\[ ] i.e., should coincide for different substances in the reduced coordinates \( H/H_{MD} \) for the case \( H \leq H_{MD} \).

VI. CONCLUSIONS

The main results of the paper can be formulated as follows:

1. The equilibrium DS that arises in the course of thermoelastic phase transition results from the finite-size effects closely related with the properties of sample surface. The absolutely rigid surface produces the imaginary “incompatibility elastic charges” which long-range field contributes into the energy of the sample, corresponding contribution being proportional to the sample volume and thus provides the twinning effect.

2. Contribution from inhomogeneous distribution of the strain below the phase transition point is proportional to the DS period and in the case of large samples is vanishingly small.
3. Elastic stray energy is the reason of the onset of equilibrium DS in pure AFM. The domain distribution can be reversibly regulated by the external magnetic field.

4. Low-field dependence of macroscopic parameters should be the same for different samples if compared in the reduced coordinates $H/H_{MD}$ (the law of the “corresponding states”).

**ACKNOWLEDGMENTS**

The authors are grateful to Profs. M.A.Ivanov, V.I.Marchenko and S.M.Ryabchenko for numerous discussions on the problem of mechanism of the DS formation in AFM.

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10. Strictly speaking, the surface effects as treated in Ref. 8 can give rise to equilibrium DS only for special geometry of the sample, namely, for thin plate or disk with the normal taken along the principal crystal symmetry axis.
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13. Another point is that the isomorphous striction and shear strain are transformed according to different indecomposable representations of the crystal symmetry group. Separated consideration of both constituents means that we neglect in free energy of the crystal anharmonic terms which are usually small.

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14. This case is realized, for example, in the pure cobalt where fcc-hcp phase transition takes place. The both phases can be transformed to each other by the shift of closely-packed planes, so no incompatibility occurs on the interface between the fcc and hcp phases.
15. Another case of the thin plate with the normal in the direction of spontaneous elongation (contraction) seems to be unreal for the traceless part of strain tensor. Really, elongation (contraction) in plane normal direction correspond to contraction (elongation) in perpendicular direction lying in the plate plane and thus breaking the condition $\mathbf{R}$.
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17. Equations (12) in the divergent form can also be obtained from the functional (13) by varying it with respect to the shift vector components. The compatibility conditions in this case are obviously excessive.
18. An analogous expression was deduced in Ref. 9 on the basis of the empirical Saint-Venan principle.
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20. If the sample thickness is of the order of optimal DS period, the preferable configuration is homogeneous.
21. A kind of such conformity was found out in Ref. 8 for the high-filed dependence of magnetostriction where the sample can be treated as monodomain, the characteristic field being the spin-flip field value and corresponding dimensionless coordinates were $H/(2H_E)$. 