Shape-induced anisotropy in antiferromagnetic nanoparticles

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High fraction of the surface atoms considerably enhances the influence of size and shape on the magnetic and electronic properties of nanoparticles. Shape effects in ferromagnetic nanoparticles are well understood and allow to set and control the parameters of a sample that affect its magnetic anisotropy during production. In the present paper we study the shape effects in the other widely used magnetic materials – antiferromagnets, – which possess vanishingly small or zero macroscopic magnetization. We take into account the difference between the surface and bulk magnetic anisotropy of a nanoparticle and show that the effective magnetic anisotropy depends on the particle shape and crystallographic orientation of its faces. Corresponding shape-induced contribution to the magnetic anisotropy energy is proportional to the particle volume, depends on magnetostriction, and can cause formation of equilibrium domain structure. Crystallographic orientation of the nanoparticle surface determines the type of domain structure. The proposed model allows to predict the magnetic properties of antiferromagnetic nanoparticles depending on their shape and treatment.

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I. INTRODUCTION

Magnetic nanoparticles (NP) are widely used as constitutive elements for the information technology (e.g., memory cells, spin valves, magnetic field controllers etc.). To drive and control the magnetic state of a particle and values of critical fields and currents, we can use not only internal properties of magnetic material, but also shape and size of the sample. As for ferromagnetic (FM) particles, shape effects allow to tailor the effective magnetic anisotropy and critical field values during production.

On the other hand, nowadays technologies use antiferromagnetic (AFM) nanoparticles along with (or sometimes instead of) FM ones. Experiments with AFM particles show that the reduction of size to tens of nanometres leads to noticeable changes of properties compared to the bulk samples: increase of lattice parameters in the magnetically ordered phase; increase of the magnetic anisotropy; pronounced decrease of AFMR frequency. Some of the finite size effects could be caused by the shape and faces orientations of nanoparticle. For example, according to the Néel predictions, small AFM particles exhibit uncompensated magnetic moment with the size- and shape-dependent value. Recent experiments with rather large (100-500 nm size) AFM particles discovered the shape effects similar to the shape-induced phenomena in FM materials: i) switching of AFM vector from crystallographic to particle easy-axis with an increase of aspect ratio; ii) correlation between the type of domain structure and such parameters as aspect ratio of the sample and orientation of faces.

However, the mechanism of the finite-size and shape effects in AFM nanoparticles is still an open issue.

Shape effects in AFM particles could, in principle, originate from a weak ferromagnetic moment thus reducing the difference between AFM and FM systems to quantitative one. On the other hand, certain dynamic and equilibrium properties of AFM, like peculiarities of the magnon spectra or coupling to the external magnetic field, could not be reduced to FM ones.

Understanding the mechanisms of the shape effects specific to AFM ordered systems is crucial for optimizing and fine-tuning the properties of AFM-based devices and clarifying the fundamental questions whether the shape effects reside in AFM with vanishingly small macroscopic magnetization, and which of peculiar AFM properties might depend on the particle shape. For this purpose we investigate the finite-size and shape effects in AFM particles, regardless of their macroscopic magnetization, combining two previously shown statements: i) the shape effects in AFM materials may originate from the long-range fields of “magnetoelastic” charges due to spontaneous magnetostriction below the Néel temperature (so called destressing fields); ii) “magnetoelastic” charges may arise from the surface magnetic anisotropy. We consider the particles with the characteristic size below the several critical lengths of monodomainization (which, for convenience, are referred to as “nanoparticles”).

The basic idea is to consider a priori the surface and bulk properties as different: to distinguish the constants of surface and bulk magnetic anisotropy and, as a consequence, equilibrium orientation of AFM vectors at the surface and in the bulk. We show that due to the long-range nature of elastic forces, the surface anisotropy contributes to the magnetic energy of the sample. This contribution is proportional to the particle volume, depends on the aspect ratio and crystallographic orientation of the sample faces, and affects equilibrium (single- and multi-domain) state of AFM nanoparticle.

The proposed approach requires consistent description of the magnetic and elastic subsystems of AFM particles and thus differs from the well-known formalism for the FM. M.I. Kaganov has already pointed out the role of the surface magnetic anisotropy effects on spin-flip transitions in magnetic materials, considering the magnetic moment at the surface as an additional parameter; however, his approach eliminates magnetoelastic and shape effects, while in our work we assume noticeable influence of these effects on the properties of the nanosized AFM particle.

II. MODEL

To describe the equilibrium magnetic state of a NP we need to introduce at least three additional (in comparison with bulk samples) parameters that characterize: i) shape, ii) size, and iii) orientation of sample faces.

We consider a thin flat rectangular particle (thickness $h \ll b < a$, Fig. 1), typical for experimental studies (see, e.g.,). The thickness $h$ of a particle is, however, large enough to ensure an AFM ordering (i.e., significantly larger than the magnetic correlation length).

The sample surface (see Fig. 1 b) consists of four faces with the normal vectors $\mathbf{n}_j = (\cos \psi_j, \sin \psi_j)$, $j = 1, \ldots, 4$ ($x, y$ are parallel to crystallographic axes). We disregard the upper and lower faces ($z = Z = const$) as they do not contribute to the effects discussed below. Equations that define the surface are ($Z \in [0, h]$, $X, Y$ are parallel to the
For such a model, the additional external (in thermodynamic sense) parameters of the NP are: i) aspect ratio $a/b$ (defines the shape), ii) width $b$ (defines characteristic size), and iii) angle $\psi$ (defines the orientation of the surfaces).

We consider a typical collinear AFM with two equivalent sublattices $\mathbf{M}_1$ and $\mathbf{M}_2$: the Néel (or AFM) vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$ plays a role of the order parameter. Far below the critical point the magnitude of the AFM vector is fixed (we assume $|\mathbf{L}| = 1$).

\[
\begin{align*}
X &= a/2, \quad Y \in [-b/2, b/2], \quad \mathbf{n}_1 = (1, 0), \quad \psi_1 = \psi, \\
Y &= b/2, \quad X \in [-a/2, a/2], \quad \mathbf{n}_2 = (0, 1), \quad \psi_2 = \psi + \pi/2, \\
X &= -a/2, \quad Y \in [-b/2, b/2], \quad \mathbf{n}_3 = (-1, 0), \quad \psi_3 = \psi + \pi, \\
Y &= -b/2, \quad X \in [-a/2, a/2], \quad \mathbf{n}_4 = (0, -1), \quad \psi_4 = \psi + 3\pi/2.
\end{align*}
\]

(1)

Figure 1. (Color online) Sample (a) and orientation of the Néel vector $\mathbf{L}$ (b) with respect to crystal axes $(x, y)$ and sample edges $(X, Y)$.

To obtain equilibrium distribution $\mathbf{L}(\mathbf{r})$ for the NP of given shape and size, we minimize the total energy $W$ which includes several terms of different nature. First of all, we may consider the surface as a separate magnetic phase\(^{18,21,22}\) with a small but finite thickness $\delta_{\text{sur}}$ (narrow peripheral region $S$ of thickness $\delta_{\text{sur}}$ in Fig. 2) and thus distinguish the bulk, $W_{\text{bulk}}$, and the surface, $W_{\text{sur}}$, contributions:

\[
W = W_{\text{bulk}} + W_{\text{sur}}.
\]

(2)

Then, we can also distinguish different contributions to the bulk energy, $W_{\text{bulk}}$, the most important are those that describe the magnetic anisotropy, $w_{\text{anis}}$, exchange, $w_{\text{exch}}$, and magnetoelastic, $w_{\text{m-e}}$ coupling. We assume that the bulk magnetic anisotropy corresponds to tetragonal symmetry with two equivalent easy directions in the NP plane $(x$ or $y$ in Fig. 1 b) and model respective contribution to the energy density as:

\[
w_{\text{anis}} = \frac{1}{2}K_{\parallel}L_z^2 - K_{\perp}(L_x^4 + L_y^4),
\]

(3)

where $K_{\parallel} \gg K_{\perp} > 0$ are the phenomenologic anisotropy constants.

Exchange interactions (responsible for inhomogeneous distribution of the Néel vector inside the sample) give rise to a gradient term

\[
w_{\text{exch}} = \frac{1}{2}\alpha(\nabla \mathbf{L})^2,
\]

(4)

where $\alpha$ is a phenomenological constant. Competition between the exchange coupling (4) and magnetic anisotropy (3) defines the characteristic size $\xi_{\text{DW}}$ of the domain wall (DW): $\xi_{\text{DW}} = (1/2)\sqrt{\alpha/K_{\perp}}$.

Magnetoelastic coupling in AFM materials can be pronounced pronounced (compared with FM ones) due to the presence of strong crystal field and, as a result, strong spin-orbit coupling (like in oxides LaFeO$_3$ or NiO). In the simplest case of the elastically isotropic material, the density of magnetoelastic energy is:

\[
w_{\text{m-e}} = \lambda_{\text{iso}} L^2 Tr\hat{u} + 2\lambda_{\text{anis}} \left[ \mathbf{L} \otimes \mathbf{L} - \frac{1}{3}I(\hat{u} - \frac{1}{3}I \mathbf{Tr}\hat{u}) \right],
\]

(5)

where $\hat{u}$ is the strain tensor, $I$ is the identity matrix, constants $\lambda_{\text{iso}}$ and $\lambda_{\text{anis}}$ describe isotropic and shear magnetostriction, respectively.

Final expression for the bulk energy is thus given by

\[
W_{\text{bulk}} = \int_V (w_{\text{anis}} + w_{\text{exch}} + w_{\text{m-e}} + w_{\text{elas}}) dV,
\]

(6)
where $u_{\text{elas}}$ is the elastic energy density (see, e.g.\textsuperscript{25}), $V = abh$ is the NP volume, all other terms are defined above.

At last, let us focus on the magnetic surface energy $W_{\text{sur}}$ which is of crucial importance for our model and needs special discussion. Experiments with the nanoscale AFM particles reveal significant difference between the magnetic ordering and hence the magnetic properties of the surface from those of the bulk. In particular, depending on the material, treatment, and other technological factors the NP surface may lack the long-range magnetic structure (paramagnetic or spin glass\textsuperscript{18,19}), or may have different type of ordering (e.g. multi- vs. two-sublattieral in the bulk\textsuperscript{20}), or different easy axis/axes. We consider the last case and assume, for the sake of simplicity, that the easy magnetic axis at the surface is perpendicular to the normal $n$; then, expression for the magnetic surface energy takes the form:

$$W_{\text{sur}} = K_{\text{sur}} \int_S (\mathbf{L}n)^2 dS = K_{\text{sur}} \sum_{j=1}^{4} \int_{S_j} (\mathbf{L}n_j)^2 dS, \quad (7)$$

where $K_{\text{sur}} > 0$ is a phenomenological constant. $W_{\text{sur}}$ obviously depends on orientation of edges: angles $\psi_j$, or, equivalently, vectors $\mathbf{n}_j$ (see (1)).

It is instructive to compare the specific surface magnetic anisotropy $K_{\text{sur}}/\delta_{\text{sur}}$ with the magnetic anisotropy constant $K_\perp$: they have the same order of value ($K_{\text{sur}}/\delta_{\text{sur}} \propto K_\perp$), if the broken exchange bonds play the main role in formation of the surface properties; while in the case of dominating dipole-dipole interactions $K_{\text{sur}}/\delta_{\text{sur}}$ can be much greater than $K_\perp$\textsuperscript{13}.

It should be stresses, that, in principle, all introduced phenomenological constants fall into two categories: internal\textsuperscript{23} (indexed “in”) and superficial (indexed “sur”); interactions of both types can contribute to the shape effects. However, in our model we distinguish only between the magnetic constants $K_{\text{sur}}/\delta_{\text{sur}}$ and $K_\perp$, taking this difference as the most important.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.pdf}
\caption{(Color online) Space distribution of AFM vector (arrows) in a single domain (a) and multidomain (b) states.}
\end{figure}

The expression (2) for the NP energy is the functional over the field variables $\mathbf{L}(\mathbf{r})$ (the AFM vector) and $\mathbf{u}(\mathbf{r})$ (the displacement vector). We reduce the number of independent variables to three assuming that: $i)$ vector $\mathbf{L}$ lies within the $xy$ plane and can be parametrised by a single angle $\varphi$ (see Fig. 1 b) because of a strong easy-plane anisotropy ($K_\parallel \gg K_\perp$); $ii)$ strain component $u_{zz}$ in a rather thin plate ($h \ll a, b$) can be considered as homogeneous and thus can be excluded from consideration (see\textsuperscript{25}). The standard minimum conditions generate the set of differential equations for one magnetic, $\varphi(\mathbf{r})$, and two elastic, $u_x(\mathbf{r})$, $u_y(\mathbf{r})$ variables in the bulk:

$$-\alpha \Delta \varphi + K_\perp \sin 4\varphi + 2\lambda_{\text{anis}}[-(u_{xx} - u_{yy}) \sin 2\varphi + 2u_{xy} \cos 2\varphi] = 0, \quad (8)$$

$$\Delta u_x + \nu_{\text{eff}} \nabla_x \nabla u = -\lambda_{\text{anis}}/\mu [\nabla_x \cos 2\varphi + \nabla_y \sin 2\varphi], \quad (9)$$

$$\Delta u_y + \nu_{\text{eff}} \nabla_y \nabla u = -\lambda_{\text{anis}}/\mu [\nabla_x \sin 2\varphi - \nabla_y \cos 2\varphi]. \quad (10)$$

Here, operators $\Delta$ and $\nabla$ are two-dimensional, $\nu_{\text{eff}} \equiv (1 + \nu)/(1 - \nu)$ is the effective Poisson ratio (instead of 3-dimensional $\nu$\textsuperscript{25}), $\mu$ is the shear modulus.

Equations for the AFM vector at the $j$-th face (variables $\varphi_{\text{sur}}$, see (1))

$$- K_{\text{sur}} \sin 2(\varphi_{\text{sur}} + \psi_j) + \alpha (\mathbf{n}_j \nabla) \varphi_{\text{sur}} = 0 \quad (11)$$
could be considered as the boundary conditions. They differ from the standard boundary conditions for AFMs (see, e.g., 26, 27) due to the presence of the additional surface term with \( K_{\text{surf}} \).

In the limit \( K_{\text{surf}} \to 0 \) the solutions of equations (9), (10), (11) are well known: the AFM vector \( \mathbf{L}(r) = \text{const} \) lies along one of the easy axes \( (\varphi_{\text{in}} = 0 \text{ or } \pi/2) \), the displacement vector \( \mathbf{u}(r) \) generates the homogeneous field of the magnetoelastically induced strain:

\[
\begin{align*}
    u^{(0)}_{xx} - u^{(0)}_{yy} &= -\frac{\lambda_{\text{anis}}}{\mu} \cos 2\varphi_{\text{in}}, \\
    u^{(0)}_{xy} &= -\frac{\lambda_{\text{anis}}}{2\mu} \sin 2\varphi_{\text{in}}.
\end{align*}
\]

In the massive (infinite) samples the spontaneous striction (12) causes magnetoelastic gap in the spin-wave spectrum (in assumption of “frozen” lattice), but does not affect the equilibrium orientation of the AFM vector (all the magnetostrictive terms in (8) cancel out, eliminating the shape effect).

For the finite-size samples with nonzero surface anisotropy \( (K_{\text{surf}} \neq 0) \) the easy direction at least in some near-surface regions unavoidably differs from that in the bulk and so, the spatial distribution of the AFM vector should be non-uniform. As a result, the sources of the displacement field – the non-zero gradient terms, or “magnetoelastic charges” – appear in the r.h.s of equations (9) and (10). In the following section we discuss this issue in more details.

III. SHAPE-INDUCED ANISOTROPY

The consistent theory of shape effects in AFMs should account for the long-range elastic and magnetoelastic interactions and thus should rest upon the complete set of equations (8)-(10). However, the displacement field \( \mathbf{u}(r) \) can be formally excluded from consideration once the Green tensor \( G_{jk}(r, r^\prime) \) for equations (9) and (10) is known (see Appendix A). In this case the spatial distribution of the AFM vector \( \mathbf{L}(r) \) should minimize the energy functional

\[
W[\mathbf{L}(r)] = \int_V (w_{\text{mag}} + w_{\text{exch}}) dV + W_{\text{surf}} + W_{\text{add}},
\]

which includes the additional term of magnetoelastic nature:

\[
W_{\text{add}} = \frac{2\lambda^2_{\text{anis}}}{\mu} \int_V \int_V \nabla_m [L_j(r)L_m(r)] G_{kj}(r, r^\prime) \nabla'_l [L_k(r^\prime)L_l(r^\prime)] dV dV' + \frac{2\lambda^2_{\text{anis}}}{\mu} \oint_S [L_j(r_{\text{surf}})L_m(r_{\text{surf}})] G_{kj}(r_{\text{surf}}, r_{\text{surf}}') L_k(r_{\text{surf}}') L_l(r_{\text{surf}}') dS_m dS'_l.
\]

Analysis of the Exp. (14) shows that any inhomogeneous distribution \( \mathbf{L}(r) \) gives nonzero, generally positive contribution to energy \( W_{\text{add}} \). Due to the “Coulomb-like” nature of the elastic forces \( (G_{jk}(r, r^\prime) \propto 1/|r - r^\prime|^3) \) this contribution scales as sample volume \( V \). In addition, nonlocality of the \( W_{\text{add}} \) term turns equations (8) to integro-differential ones and thus complicates the problem.

In the present paper we propose the simplified approach to solve equations (8) - (10) using the following peculiar features of antiferromagnets.

First, we consider the magnetostriction of AFMs as a secondary order parameter which means that in the thermodynamic limit (in neglect of boundary conditions) the homogeneous spontaneous strains (12) preserve the symmetry of the magnetically ordered state and orientation of the easy axis. In addition, though usually the magnetoelastic energy is comparable (up to the order of value) to the 4-th order magnetic anisotropy (i.e., \( K_4 \)) constant, it can be much less than the uniaxial magnetic anisotropy. Thus, assuming strong uniaxial surface anisotropy \( K_{\text{surf}} \gg K_4 \delta_{\text{surf}} \), we can disregard the influence of magnetoelastic strains on equilibrium orientation of AFM vector at the surface. However, this assumption does not restrict the relation between \( K_{\text{surf}} \) and the characteristic DW energy \( \sigma_{\text{DW}} \), because \( \sigma_{\text{DW}} \propto \sqrt{K_4 J_{\text{lat}}} \gg K_4 \delta_{\text{surf}} \) (where \( J \gg K_4 \) characterizes the exchange coupling, \( J_{\text{lat}} \) is the lattice constant, and we used the following relations: \( \sigma \propto J_{\text{lat}} \delta_{\text{surf}} \)).

Second, we propose the following hierarchy of characteristic length scales: the width of magnetic inhomogeneity is much less than the sample size, \( \xi_{\text{DW}} \ll a, b \), but much greater than interatomic distance, \( \xi_{\text{DW}} \gg a_{\text{lat}} \) (due to exchange enhancement); the width of elastic inhomogeneity has interatomic scale and thus is much less than \( \xi_{\text{DW}} \). Note, that the value of \( \xi_{\text{DW}} \) in nanoparticles with the large fraction of the surface atoms can be much smaller than that for the bulk samples due to variation of magnetoelastic and exchange coupling (see, e.g., 28). Thus, inequality \( \xi_{\text{DW}} \ll a, b \) keeps true in a wide range of the sample dimensions down to tens of nanometers (below this range applicability of the continual model is questionable).

Thus, within the above approximations, equilibrium orientation of the AFM vector at the surface results mainly from competition of the magnetic interactions: surface magnetic anisotropy and inhomogeneous exchange coupling,
once the bulk vector $\mathbf{L}_{\text{in}}$ is fixed. Orientation of $\mathbf{L}_{\text{in}}$, in turn, is defined by interplay between the bulk magnetic anisotropy and magnetostrictive contribution induced by spatial rotation of AFM vector in the thin ($\propto \xi_{DW}$) near-surface region (see Fig. 3). So, the effective shape-induced magnetic anisotropy and equilibrium distribution of AFM vector could be determined self-consistently according to the following procedure: $i$) to calculate $\mathbf{L}_{\text{surf}}$ starting from some (initially unknown) “seed” distribution of the AFM vector $\mathbf{L}_{\text{in}}$ in the NP bulk; $ii$) to substitute thus defined seed distribution into equations for the displacement vector and to determine corresponding field sources (magnetoelastic charges); $iii$) to calculate charge-induced average strains whose contribution into free energy is proportional to the sample volume; $iv$) to define the effective magnetic anisotropy which accounts for the average strains and calculate $\mathbf{L}_{\text{in}}$.

We note that the form of the seed distribution (and hence the free variable of the structure) is different for a single- and a multi-domain states. In the first case $\mathbf{L}_{\text{in}}$ is homogeneous within the bulk but can deflect from the magnetic easy axis, so, $\varphi_{\text{in}}$ is the appropriate free variable. In the second case we assume, in analogy with FM, that AFM vector within each of the domains is fixed and parallel to one of two equivalent easy axes; then, free variable coincides with the fraction of type-I (or type-II) domains.

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**Figure 3.** (Color online) Distribution of the Néel vector in the vicinity of $Y = b/2$ face, multidomain state. (a) Periodic (period $d$) domain structure, double arrows indicate orientation of AFM vectors inside domains and in the near-surface region (shaded horizontal stripe). (b) Space dependence of $L_x(\xi)$ (solid line) calculated from (15) provided that $\varphi_{\text{in}} = 0$. The horizontal line defines the center $\xi_0$ of a virtual full domain wall (dotted line). Shaded vertical bar indicates the position of surface region. Direction of DW normal coincides with the axis $\xi$ of the local coordinate system (inset).
### A. Seed distribution and magnetoelastic charges

In the simplest case of a single-domain state (Fig. 2a), there are two homogeneous regions: the “shell” (of the thickness $\delta_{\text{sur}}$) and the core. An equilibrium value $\varphi_{\text{in}}$ inside the core is fixed, constant (as $\Delta \varphi_{\text{in}} = 0$), but unknown (in some cases discussed below $\varphi_{\text{in}} = 0$ or $\pi/2$ that corresponds to one of the easy axes). We calculate the value $\varphi_{\text{sur}}^{(j)}$ at the surface from Eq. (11) with account of the standard expression for the domain wall profile:

$$\sin 2(\varphi(\xi) - \varphi_{\text{in}}) = 2\xi_{\text{DW}} \frac{d\varphi}{d\xi} = \frac{1}{\cosh((\xi - \xi_0)/\xi_{\text{DW}})}$$

(15)

Face normals generate the set of variables $\xi = (\pm X - a/2), (\pm Y - b/2)$ of the local coordinate system (see inset in Fig. 3b). Position $\xi_0$ of the DW center is calculated from the boundary conditions (see below). In (15) we neglect the possible difference between the DW width $\xi_{\text{DW}} = (1/2)\sqrt{a/K_\perp}$ in the near-surface region and in the core.

Substituting (15) in (11), we obtain the following equation for $\varphi_{\text{sur}}^{(j)}$:

$$\tan 2\varphi_{\text{sur}}^{(j)} = \frac{K_{\text{sur}} \sin 2\psi_j + \sigma_{\text{DW}} \sin 2\varphi_{\text{in}}}{\sigma_{\text{DW}} \cos 2\varphi_{\text{in}} - K_{\text{sur}} \cos 2\psi_j},$$

(16)

where $\sigma_{\text{DW}} = \sqrt{\alpha K_\perp}$ and $K_\perp$ is the characteristic energy of the domain wall. The values $\varphi_{\text{sur}}^{(j)}$ at the opposite faces coincide: $\varphi_{\text{sur}}^{(1)} = \varphi_{\text{sur}}^{(3)}, \varphi_{\text{sur}}^{(2)} = \varphi_{\text{sur}}^{(4)}$ (see (1)).

Analysis of Exp. (16) shows that the AFM vector at the surface can be either parallel to the edge: $\varphi_{\text{sur}}^{(j)} = \psi_j$, as shown in Fig. 2, (in the limit of large surface anisotropy, $K_{\text{sur}} \gg \sigma_{\text{DW}}$), or coincide with the bulk AFM vector: $\varphi_{\text{sur}}^{(j)} = \varphi_{\text{in}}$ (for the vanishing surface energy, $K_{\text{sur}} \ll \sigma_{\text{DW}}$). In the last case the surface influence and, correspondingly, shape effects disappear.

Note that the surface DW is “incomplete”: in general, DW center is located outside the sample (see Fig. 3b) and its coordinate $\xi_0^{(j)}$ depends on the surface anisotropy

$$\xi_0^{(j)} = \xi_{\text{DW}} \sinh^{-1} \frac{K_{\text{sur}} \sin 2\psi_j + \sigma_{\text{DW}} \sin 2\varphi_{\text{in}}}{\sigma_{\text{DW}} \cos 2\varphi_{\text{in}} - K_{\text{sur}} \cos 2\psi_j}.$$  

(17)

In a single-domain nanoparticle the AFM vector rotates from $L_{\text{sur}}$ to $L_{\text{in}}$ in a narrow, almost zero-width ($\ll \xi_{\text{DW}} \ll a, b$) region and so, we can model the spatial dependence of $\mathbf{L}(r)$ with a step-like function. Within this approximation, r.h.s. of equations (9) and (10) are nontrivial only at the surface; this fact makes it possible to use a homogeneous form of these equations for the bulk region of the NP:

$$\Delta \mathbf{u} + \nu_{\text{eff}} \nabla \text{div} \mathbf{u} = 0$$

(18)

with the following boundary conditions for the displacement vector:

$$(\mathbf{n} \cdot \nabla) \mathbf{u}_{\text{sur}} + \nu_{\text{eff}} \mathbf{n} \text{div} \mathbf{u}_{\text{sur}} = \mathbf{n} \hat{Q}^{m-e}.$$  

(19)

In (19) we introduced the tensor of magnetoelastic charges as follows:

$$\hat{Q}^{m-e} \equiv -\frac{\lambda_{\text{anis}}}{\mu} [L_{\text{sur}} \otimes L_{\text{sur}} - L_{\text{in}} \otimes L_{\text{in}}].$$

(20)

For a rectangular-shaped sample the charges at the opposite edges coincide: $\hat{Q}^{m-e}(\mathbf{n}_1) = \hat{Q}^{m-e}(\mathbf{n}_3), \hat{Q}^{m-e}(\mathbf{n}_2) = \hat{Q}^{m-e}(\mathbf{n}_4)$. We can express all the components of the $\hat{Q}^{m-e}$ tensor in terms of two nontrivial combinations, $Q_1^{m-e} = Q_{XX}^{m-e} - Q_{YY}^{m-e}$ and $Q_2^{m-e} = 2Q_{XY}^{m-e}$ (in $X, Y$ coordinates).

From definition (20) and the relations (16) it follows that

$$Q_1^{m-e}(\mathbf{n}_{1,2}) = \frac{\lambda_{\text{anis}}}{\mu} \left[ \cos 2(\varphi_{\text{in}} + \psi) - \frac{\sigma_{\text{DW}} \cos 2(\varphi_{\text{in}} + \psi) + K_{\text{sur}}}{\sqrt{K_{\text{sur}}^2 + 2K_{\text{sur}}\sigma_{\text{DW}} \cos 2(\varphi_{\text{in}} + \psi) + \sigma_{\text{DW}}^2}} \right],$$

(21)

and

$$Q_2^{m-e}(\mathbf{n}_{1,2}) = \frac{\lambda_{\text{anis}}}{\mu} \sin 2(\varphi_{\text{in}} + \psi) \left[ 1 - \frac{\sigma_{\text{DW}}}{\sqrt{K_{\text{sur}}^2 + 2K_{\text{sur}}\sigma_{\text{DW}} \cos 2(\varphi_{\text{in}} + \psi) + \sigma_{\text{DW}}^2}} \right].$$

(22)
Figure 4. (Color online) Magnetoelastic charges $Q^{m-e}$ (in $\lambda_{anis}/\mu$ units) vs. surface constant $K_{sur}$ calculated for single-domain state, $\psi = \pi/4$. Inset shows the charge distribution over the particles edges. Arrows indicate orientation of AFM vector at the surface and in the bulk.

Magnetoelastic charges (20) (as well as (21), (22)) are similar to “magnetostatic charges” at the surface of FMs but have another, magnetoelastic, nature (i.e., depend on magnetostriction), and depend on the surface magnetic anisotropy $K_{sur}$. Magnetoelastic charges disappear in the limiting case of small surface anisotropy $K_{sur} \ll \sigma_{DW}$ and reach the maximum possible value when $K_{sur} \gg \sigma_{DW}$ (as illustrated in Fig. 4). Like magnetostatic, magnetoelastic charges depend on the crystallographic orientation of the sample faces and vanish for those parts of the surface where $\mathbf{L}_n \parallel \mathbf{L}_{sur}$. From equations (18), (19) it follows that magnetoelastic charges produce long-range (decaying as $1/r^2$) elastic fields, which, in turn, lead to the “destressing” effects (similar to “demagnetising” effects in FMs).

Another way to interpret the formation of magnetoelastic charges presents itself in terms of incompatibility of seed spontaneous deformations at the surface and in the bulk. To this end, sufficient condition for charges to appear stems from the difference between the surface and bulk values of any physical quantity: magnetic (e.g. nonmagnetic or paramagnetic surface), magnetoelastic, or elastic (e.g. rigid shell).

### B. Average strains and shape-induced anisotropy

At the next, III, stage of the algorithm we solve equations (18), (19) for the displacement vector which, in general case, generates non-uniform field of additional (compared with (12)) elastic deformations. However, the main contribution to the magnetic anisotropy comes from the shear strains averaged over the sample volume (labeled as $\langle \ldots \rangle$):

\[
\langle u_{XX} - u_{YY} \rangle = -\frac{\pi}{1 + \nu_{eff}} \left\{ \left[ Q_{1}^{m-e}(n_2) + Q_{1}^{m-e}(n_1) \right] \left[ 1 + \nu_{eff} J_2 \left( \frac{a}{b} \right) \right] \\
- J_1 \left( \frac{a}{b} \right) \left[ Q_{1}^{m-e}(n_1) - Q_{1}^{m-e}(n_2) \right] \right\},
\]

\[
2 \langle u_{XY} \rangle = -\pi \left\{ \left[ Q_{2}^{m-e}(n_2) + Q_{2}^{m-e}(n_1) \right] \left[ 1 - \frac{\nu_{eff}}{1 + \nu_{eff}} J_2 \left( \frac{a}{b} \right) \right] \\
- J_1 \left( \frac{a}{b} \right) \left[ Q_{2}^{m-e}(n_1) - Q_{2}^{m-e}(n_2) \right] \right\},
\]

where $J_1(a/b)$, $J_2(a/b)$ are the dimensionless shape functions of the aspect ratio $a/b$ (see Fig. 5):

\[
J_1 \left( \frac{a}{b} \right) = \frac{2}{\pi} \left[ \arctan \frac{a}{b} - \arctan \frac{b}{a} + \frac{a}{4b} \ln \left( 1 + \frac{b^2}{a^2} \right) - \frac{b}{4a} \ln \left( 1 + \frac{a^2}{b^2} \right) \right],
\]

\[
J_2 \left( \frac{a}{b} \right) = \frac{1}{\pi} \left[ \arctan \frac{a}{b} - \arctan \frac{b}{a} + \frac{a}{4b} \ln \left( 1 + \frac{b^2}{a^2} \right) - \frac{b}{4a} \ln \left( 1 + \frac{a^2}{b^2} \right) \right].
\]
\[ J_2 \left( \frac{a}{b} \right) = \frac{4}{\pi} \left[ \frac{b}{a} \ln \left( 1 + \frac{a^2}{b^2} \right) + \frac{a}{b} \ln \left( 1 + \frac{b^2}{a^2} \right) \right]. \] (26)

Note that \( J_2(a/b) = J_2(b/a) \); \( J_1(a/b) = -J_1(b/a) \), so, \( J_1 = 0 \) for a square \( (a = b) \); in the opposite limiting case of high aspect ratio \( (a \gg b) \) \( J_1 \to 1, J_2 \to 0 \).

Figure 5. (Color online) Form-factors \( J_1, J_2 \) vs aspect ratio \( a/b \). Arrows show the points where the functions \( J_1 (a/b = 1) \) and \( K_4^{sh} (a/b \approx 16) \) change the sign.

Substituting Exps. (21), (22), (23), and (24) into Eq. (8) we arrive at the following equation for magnetic variable:

\[ K_\perp \sin 4\varphi_{in} + K_2^{sh} \sin 2(\varphi_{in} + \psi) + K_4^{sh} \sin 4(\varphi_{in} + \psi) = 0, \] (27)

where we introduce the shape-dependent coefficients \( K_2^{sh}, K_4^{sh} \), and take into account that \( \Delta \varphi = 0 \). In the limiting (and practically important) case \( K_{sur} \gg \sigma_{DW} \)

\[ K_2^{sh} = 2K^{m-e} J_1 \left( \frac{a}{b} \right), \quad K_4^{sh} = K^{m-e} \left[ 2J_2 \left( \frac{a}{b} \right) - 1 \right], \quad K^{m-e} = \frac{4\pi \mu_{eff} \lambda_{anis}^2}{(1 + \nu_{eff})\mu}. \] (28)

In general case the coefficients \( K_2^{sh} \) and \( K_4^{sh} \) depend on the constant \( K_{sur} \) of surface magnetic anisotropy and vanish when \( K_{sur} \ll \sigma_{DW} \) (see Appendix B).

Eq. (27) for the magnetic variables \( \varphi_{in} \) can be treated as the minimum condition for the effective energy density of the sample

\[ w_{eff} = \frac{W_{eff}}{V} = -\frac{1}{4} \left[ K_\perp \cos 4\varphi_{in} + 2K_2^{sh} \cos (2\varphi_{in} + \psi) + K_4^{sh} \cos 4(\varphi_{in} + \psi) \right], \] (29)

which, apart from the magnetic anisotropy, includes contributions from magnetoelastic and surface forces (the underlined terms). The last two terms in (29) cause the shape effects in AFM nanoparticle. To illustrate this result we consider some typical cases.
Let the sample edges be parallel to the easy magnetic axes ($\psi = 0$). In this case, as follows from (27) and (29), the term with $K_{2}^{sh}$ removes degeneracy of states $\varphi_{in} = 0$ and $\varphi_{in} = \pi/2$. This term is equivalent to uniaxial anisotropy, which selects the state with the collinear orientation of AFM vectors at the surface and in the bulk as energetically favorable. This means that the AFM vector is parallel to the long edge of the rectangle: if $a > b$, then $K_{2}^{sh} > 0$ and $L_{||}X (\varphi_{in} = 0)$. The second shape-induced term with $K_{4}^{sh}$ renormalizes the “bare” magnetic anisotropy constant, $K_{\perp} \rightarrow K_{\perp} + K_{4}^{sh}$; however, this effect makes no influence on the orientation of the AFM vector. For the square sample ($a = b$) the shape-induced correction has the same sign as $K_{\perp}$ ($K_{4}^{sh} > 0$) and thus does not affect equilibrium orientation of the AFM vector. The change of $K_{4}^{sh}$ sign appears for the samples with large aspect ratio ($a/b \approx 16$, see Fig. 5), where uniaxial anisotropy governs the orientation of the AFM vector, and shape-induced renormalization of $K_{\perp}$ is insignificant.

The role of the terms with $K_{4}^{sh}$ becomes noticeable when the faces (edges) of the square ($a = b$) sample are cut at the angle $\psi = \pi/4$ (i.e. along the “hard” magnetic axes). In this case the uniaxial anisotropy vanishes, $K_{2}^{sh} = 0$ and the effective magnetic anisotropy constant decreases: $K_{\perp} \rightarrow K_{\perp} - K_{4}^{sh}$. Assuming that $K_{\perp}$ and $K_{4}^{sh}$ have the same (spin-orbit) nature, we conclude that the shape can change the direction of the easy axes (if $K_{\perp} < K_{4}^{sh}$) or entirely compensate the 4-th order magnetic anisotropy (if $K_{\perp} \approx K_{4}^{sh}$), as it was recently observed in the experiments$^{12}$.

IV. MULTIDOMAIN STATE, DESTRESSING ENERGY AND CRITICAL SIZE

In the multidomain state the seed distribution can, in principle, model the domains and domain boundaries both in the bulk and at the surface. To simplify the problem we assume that distribution of the AFM vector $L_{surr}(r)$ within each face is homogeneous and $L_{surr}$ aligns due to the surface anisotropy ($K_{surr} \gg \sigma_{DW}$), as shown in Fig. 3 a. In this case, orientation of the AFM vector and, correspondingly, angle $\varphi$, can take one of two values within the bulk: $\varphi_{in}^{L} = 0$ or $\varphi_{in}^{II} = \pi/2$ (domains of two types, I and II). At the surface $\varphi_{I}^{(1)} = \varphi_{II}^{(3)}$ and $\varphi_{I}^{(2)} = \varphi_{II}^{(4)}$.

Magnetoelectric charges appear near the surface (due to the difference between $L_{surr}$ and $L_{in}$) and at the domain walls in the bulk (due to the difference between $L_{in}^{I}$ and $L_{in}^{II}$). The total charge of the full domain wall is zero because of the perfect compensation of the charges with opposite signs. So, the field of internal charges decreases rapidly with distance (as $1/r^6$, due to Coulomb-like nature of the “elastic” forces) and can be neglected.

Near-surface domain structure generates two types of the charges, $Q_{m-e}^{I}$ and $Q_{m-e}^{II}$, corresponding to two types of the domains with $L_{in}^{I}$ and $L_{in}^{II}$ (see Eq. (20)). Thus, distribution of $Q_{m-e}^{I,II}$ is space-dependent. We consider the simplest case of the stripe domain structure (see discussion of possible generalization below) and model it as

$$
\hat{Q}_{m-e}(\eta_{j}) = \left(\hat{Q}_{m-e}^{I} - \hat{Q}_{m-e}^{II}\right) f(\eta_{j}).
$$

Here $\eta_{j}$ is a local coordinate parallel to the $j$-th edge of the sample (for example, $\eta_{2} = -X$ in Fig. 3), and $f(\eta_{j})$ is a periodic function with zero mean value: $f(\eta_{j} + d) = f(\eta_{j})$, $\langle f(\eta_{j}) \rangle = 0$; $d$ is a domain structure period.

In the case of the fine domain structure, $d \ll b, a$, the averaged value $\langle \hat{Q}_{m-e}^{I} \rangle$ is independent of $j$ and coincides with that averaged over the particle volume. As in the single-domain state, the effective contribution from the averaged charges to the magnetic energy density is similar to (29):

$$
w_{destr} = -\frac{1}{4} \left\{ 2K_{2}^{sh} \left[ \cos 2(\varphi_{in} + \psi) \right] + K_{4}^{sh} \left[ \left( \cos 2(\varphi_{in} + \psi) \right)^2 - \sin 2(\varphi_{in} + \psi)^2 \right] \right\}.
$$

The term with $K_{2}^{sh}$ corresponds to the uniaxial shape-induced anisotropy. The second term, with $K_{4}^{sh}$, depends nonlinearly on the domain fraction and is analogous to the demagnetisation energy of FM. Previously$^{14}$ we named this contribution as *destr essing* energy, since it determines the equilibrium domain structure in the presence of the external fields (in the defectless samples).

We estimate the energy contribution of the second term in (30) using the analogy between the theory of elasticity and electro- (magneto-)statics: the total field of the alternating charge distribution with zero average decays exponentially into the sample at distances $d$: $u_{j} \propto \exp(-|X \pm a/2|/d), \exp(-|Y \pm b/2|/d)$. The corresponding contribution to the total energy density can easily be obtained by analogy with the well-known Kittel expressions for FMs (formulae (54), (63) in$^{19}$):

$$
w_{near-surr} = A \mu \left( \hat{Q}_{I}^{m-e} - \hat{Q}_{II}^{m-e} \right)^2 \frac{Sd}{V}
$$

where $A$ is a factor of the order of unity, $S$ is the surface area.

Comparison of (32) and (31) shows that $w_{near-surr}/w_{destr} \propto d/\ell \ll 1$ (where $\ell$ is the characteristic size sample). However, contribution $w_{near-surr}$, though small, defines the details of the domain structure (period, number of domains,
orientation and shape of DW). Also, as in the case of FM, a period of the equilibrium domain structure is determined by the competition between the energy \((32)\) (which increases with \(d\) increase) and the total DW energy density \(w_{\text{bound}} = \sigma_{\text{DW}} \ell S/(Vd)\) (which decreases with \(d\) increase). An optimal value \(d_{\text{opt}}\) (up to an unessential numerical factor) is

\[
d_{\text{opt}} \approx \sqrt{\frac{\ell \sigma_{\text{DW}}}{\mu \left( Q_{I}^{m-e} - Q_{II}^{m-e} \right)^2}}.
\]

The period \(d_{\text{opt}}\) of the domain structure defines the critical NP size \(\ell_{\text{cr}}\), below which the formation of AFM domains becomes energetically unfavourable:

\[
\ell_{\text{cr}} = d_{\text{opt}} = \frac{\sigma_{\text{DW}}}{\mu \left( Q_{I}^{m-e} - Q_{II}^{m-e} \right)^2}.
\]

Let us compare expressions \((33)\), \((34)\) with the similar expressions for the FM samples for two limiting cases.

**Strong surface anisotropy,** \(K_{\text{sur}} \gg \sigma_{\text{DW}}\). In this case, \(\left( Q_{I}^{m-e} - Q_{II}^{m-e} \right) \propto \lambda_{\text{anis}}/\mu\) and

\[
\ell_{\text{cr}} = d_{\text{opt}} = \frac{\sigma_{\text{DW}}}{\lambda_{\text{anis}}^2/\mu} \propto \frac{\sigma_{\text{DW}}}{K_{\perp}} \propto \xi_{\text{DW}}.
\]

Here we used the fact that the magnetic anisotropy in the AFM has the same nature as the magnetoelastic energy, resulting in \(K_{\perp} \propto \lambda_{\text{anis}}^2/\mu\).

**Weak surface anisotropy,** \(K_{\text{sur}} \ll \sigma_{\text{DW}}\). In this case \(\left( Q_{I}^{m-e} - Q_{II}^{m-e} \right) \propto \lambda_{\text{anis}} K_{\text{sur}}/(\sigma_{\text{DW}} \mu)\), and so,

\[
\ell_{\text{cr}} = d_{\text{opt}} = \frac{\sigma_{\text{DW}}}{\lambda_{\text{anis}}^2/\mu} \left( \frac{\sigma_{\text{DW}}}{K_{\text{sur}}} \right)^2 \propto \xi_{\text{DW}} \left( \frac{\sigma_{\text{DW}}}{K_{\text{sur}}} \right)^2 \gg \xi_{\text{DW}}.
\]

Thus, in AFMs, as opposed to FMs, the domain size and the critical particle size depend on the properties of the surface (in this particular case – on the magnetic surface anisotropy). In the presence of strong surface anisotropy the characteristic size of the domain is of the same order of magnitude as the DW width. A similar result is obtained in the FMs, provided that the magnetic anisotropy is of the same order as the shape anisotropy. In the limiting case of zero surface magnetic anisotropy the critical particle size tends to infinity, in agreement with expected absence of the shape effects in the large AFM samples (thermodynamic limit).

We emphasize that, in contrast to FMs, the equilibrium structure of AFMs is formed by the orientational domains only (the angle between vectors \(\mathbf{L}\) in neighboring domains \(\approx 180^\circ\)). The translational \(180^\circ\) domains in collinear AFMs (that have opposite \(\mathbf{L}\) directions) are physically indistinguishable and should be identified by the presence of the interfaces. This problem is out of the scope of the paper.

**V. DISCUSSION**

We developed a model that takes account of the magnetic surface anisotropy and magnetoelastic interactions and predicts the additional shape-dependent magnetic anisotropy in AFM. The surface anisotropy selects one of the easy magnetic axes as energetically favorable, while magnetoelastic long-range interactions transfer the influence of the surface on the entire NP bulk. Formally, we can describe such effects using the tensor of magnetoelastic charges \((20)\) localized at the NP surface.

Shape-induced magnetic anisotropy manifests itself in two ways: \(i\) as the uniaxial anisotropy, which splits energy of otherwise degenerated equilibrium orientations of AFM vector; \(ii\) as a “demagnetising” (destressing) factor, which promotes formation of a certain domain type.

The first effect occurs when the shape-imposed easy axis is perpendicular to the “proper” easy magnetic axis of the crystal (e.g., induced by an external magnetic field)\(^{10}\). The constants of intrinsic and shape-induced magnetic anisotropy are of opposite signs; so, there is a critical aspect ratio \(a/b\), at which spin-flop transition of the AFM vector takes place.

The second effect appears when the domain structure is reversibly changed\(^{20}\) under the action of external fields (magnetic or mechanical). In the flat rectangular NP with \(a \neq b\), the shape-induced anisotropy plays the same role as the external field, resulting in unbalance between domains of different types.
The constant of the surface anisotropy also determines the critical parameters limiting formation of the domain structure. For example, if the NP size is comparable with the domain structure period \( d \), formation of the domain walls and thus of domain structure is unfavourable. On the other hand, for the elongated samples with \( a \gg d \) but \( |K_2^{\text{sh}}((a/b)_{\text{cr}})| \geq K_\perp \) (see (29)), there is only one possible equilibrium orientation of the AFM vector and thus only one type of domains. In this case, the orientation of the easy axis depends not only on ratio \( a/b \), but also on the angle \( \psi \), which determines the orientations of the sample edges. So, the control of the AFM particles shape allows not only to create single-domain samples, but also to drive the magnetic ordering direction.

The magnetoelastic charges-based formalism allows to predict, at least qualitatively, the morphology of the domain structure depending on the size of the NP and crystallographic orientations of its edges. Note that charge contribution increases the energy density of NP compared with the case of an infinite crystal. So, charge-less (or zero mean) configuration is more favourable, as in FM. If the NP edges are parallel to the crystallographic axes, the surface charges disappear in the structure that shown in Fig. 6a, – when the domain of a certain type grows from the edge into the bulk as far as possible. This type of the domain growth was observed experimentally in\(^{12}\); the authors named it “edge effect”. Edge effect disappears if sample edges are rotated at angle \( \psi = \pi/4 \) with respect to easy magnetic axes. Really, in this geometry charge vanishes only in average (due to formation of the domain structure that is periodic along the edge, Fig. 6b). In this case, we assume that domain formation starts from the vertices of the rectangle, and the surface tension stresses can play a significant role in this process. A detailed discussion of this issue is beyond the scope of this paper.

![Diagram of domain states](https://example.com/diagram.png)

**Figure 6.** (Color online) Multidomain state of the nanoparticle with edges parallel (a) or at an angle 45° (b) relative to crystal axes. Arrows outside the rectangle indicate the orientation of the Neél vector at the surface.

The explicit form of the shape-induced anisotropy constants \((28), (29)\) depends on the magnetic properties of the surface. In our model we suggest that the magnetic ordering at the surface somehow differs from that in the bulk, e.g., by orientation of easy axis (see (1)). However, it is possible to generalize the model and consider other typical situations: e.g., the surface of the sample is paramagnetic, unlike the bulk. In this case, we expect that the shape effects will show up in the destressing energy (similar to \((31)\)), but the shape-induced anisotropy in the single-domain samples will be absent, as well as edge effect. Thus, the shape effects give us indirect information about the magnetic structure of the NP surface.

The proposed model predicts the occurrence of the domains in arbitrarily large samples, provided that \( K_{\text{sur}} \neq 0 \). Contribution of the magnetoelastic charges to the surface energy is proportional to the sample volume and competes with the anisotropy energy in samples of any size. On the other hand, increasing the characteristic size of the sample, we can reduce the influence of surface on the local properties up to the thermodynamic limit. Thus, we can question the existence of the upper critical size, above which the sample can be considered as single-domain. To find a rigorous answer, we need to solve a complicated problem, which is beyond the scope of our work; we confine ourselvess to a few physical considerations.

Formally, we may move to the case of physically large samples (to the thermodynamic limit) in two ways: either as \( \lim_{K_{\text{sur}} \to 0} \lim_{\ell \to \infty} \) or in the other order \( \lim_{\ell \to \infty} \lim_{K_{\text{sur}} \to 0} \). In the first case, the surface leads to the shape effects and the domain structure formation. Increasing the sample size, and thus the domain size, we obtain large homogeneous regions, in which the influence of domain walls and the surface can be neglected (this issue is discussed in details below). In the second case, we exclude the surface from consideration and get the homogeneous throughout the sample solution (12), which corresponds to the energy minimum. The domain structure is absent and the size of the sample is not important as a thermodynamic parameter.

We emphasize that our estimates of the domain structure period \((33)\) and lower critical sample size \((34)\) are based on the simplified Kittel’s model of striped domain structure with one characteristic period. While the optimal period is less than or equal to the critical sample size \((34)\), such choice of seed distribution seems reasonable. However, if the sample size (and \( d_{\text{opt}} \)) increases, the contribution of the charges \( \bar{Q}^{\text{m}}_{1,11} \) to the energy grows. At the same time, energy can be decreased by the branching (fractalisation) of the domain structure: the surface of “large” domain stimulates formation of small domains inside. Similar structures were observed in ferromagnetic and ferroelastic materials (such as martensites, in which deformation is the primary order parameter\(^{10}\)). In\(^{31}\) authors show that the scale
invariance of the two-dimensional Laplace equation causes the fractal nature of the ferromagnetic and intermediate state superconducting structures. In our case, assuming the Coulomb nature of the elastic forces, we can also expect that the system of equations (18) contains a similar (probably more difficult) fractal solution. We suppose that a multi-domain hierarchical structure, which contains ever smaller regions with various orientations of the AFM vector, may also appear in large AFM samples. This leads us to the following conclusions.

First, for large \( \ell \) we need to adjust the estimate (33) for the domain structure period \( d_{\text{opt}} \). Indeed, the total length of the domain walls in the fractal structure increases with the domain size \( d \) as \( d^{D_H} \), where \( D_H \) is the Hausdorff fractal dimension. Thus, the total energy of the domain walls changes as \( w_{\text{bound}} \propto \ell d^{D_H} \) (similar estimate for multiferroic BiFeO\(_3\) was made in \cite{32}), and the optimal domain size is \( d_{\text{opt}} \propto \ell^{1/(3-D_H)} \). For the striped domains \( D_H = 1 \), which yields (33); for branching structures, obviously, \( D_H > 1 \), and the dependence \( d_{\text{opt}}(\ell) \) is stronger. Second, in the fractal structure the ratio of the surface energy to the bulk energy decreases with increasing \( \ell \) slower than \( 1/\ell \), indicating the important role of the surface in large samples.

Finally, we note that branching domain structure also allows transition to thermodynamic limit: as we have already noted, for periodic structures the field of magnetoelastic charges is screened over distances of the order of \( d_{\text{opt}} \), and the local magnetic properties of homogeneous regions (such as orientation of AFM vector, AFMR frequencies, susceptibility, etc.) depend on the internal (bulk) parameters only, and the role of the surface energy is insignificant.

Let us discuss another, practical, approach to the concept of the upper critical dimension. Imagine that initially the multidomain sample is transferred to a single-domain, homogeneous state (without DWs) by an external field. The question is: will the domain structure appear after the field is switched off? As in the case of the FM materials, the answer depends on various parameters, including the size of the sample, and the magnitude of the DW formation activation barrier \( U_{\text{bar}} \). As we have already noted, the domain formation starts at the surface – from edges or vertices of the particle, depending on the crystallographic orientation of the surface. The domain nucleus creates the elastic stress field; energy density of this field decreases with distance (in analogy with the elastic energy of dislocation field) as \( [(Q_{\text{me}})^2/\mu] \ln r/r_0 \) (\( r_0 \) is a characteristic size of the order of the nucleus curvature radius). If \( U_{\text{bar}} > [(Q_{\text{me}})^2/\mu] \), then the domain walls preferably form in areas where the field of smagetoelastic charges located at the opposite edges add constructively. Hence, we estimate the upper critical size of the sample: \( \ell_{\text{cr}}^{\text{up}} \propto r_0 \exp U_{\text{bar}}/[(Q_{\text{me}})^2] \). In small particles, \( \ell < \ell_{\text{cr}}^{\text{up}} \), the interaction of charges located at the opposite edges is sufficient for the DW formation. If \( \ell > \ell_{\text{cr}}^{\text{up}} \), the sample may remain in the metastable single-domain state.

Note that we have considered the ideal, i.e. defectless, sample, eliminating the energy of twin boundaries and disclinations (the latter inevitably arise in the areas of convergence of three or more domains), and neglecting peculiarities of the AFM vector distribution near the vertices of the rectangle. Certainly, these factors should influence the domain structure formation and the effective magnetic anisotropy of the sample. However, we guess that only the surface relates the internal magnetic properties of NP and its form. We have shown that the shape effects can be caused by the long-range fields of non-magnetic nature – elastic forces – and so they should appear in the “pure” AFM samples (without FM moment as well). The effects described above should be more pronounced in the small (up to few critical lengths) samples: in this case, the formation of the magnetic structure is determined mainly by the surface and the influence of the defects can be neglected.

The results obtained show that the shape can be used as a technological factor which allows to drive, control and set the properties of antiferromagnetic nano-sized samples.

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**Appendix A: Green tensor method for the displacement field calculation**

Assuming that we know the distribution of the AFM vector \( \mathbf{L}(\mathbf{r}) \), let us examine Eqs. (9)-(10) for the elastic subsystem. Corresponding boundary conditions at the surface are:

\[
\frac{1 + \nu_{\text{eff}}}{1 - 2\nu_{\text{eff}}} \mathbf{n} \cdot \mathbf{u} + \left( n_x (\nabla_x u_x - \nabla_y u_y) + n_y (\nabla_y u_y + \nabla_y u_x) \right) = - \frac{2 \lambda_{\text{anis}}}{\mu} \mathbf{L}_{\text{surf}} \cdot (\mathbf{L}_{\text{surf}} \cdot \mathbf{n}). \tag{A1}
\]

To simplify, we skip the terms that describe the isotropic magnetostriction (as insignificant for further discussion). We denote the bulk forces vector by

\[
\mathbf{f} = - \left( \begin{array}{c} \nabla_x \cos 2\varphi + \nabla_y \sin 2\varphi \\ \nabla_x \sin 2\varphi - \nabla_y \cos 2\varphi \end{array} \right), \tag{A2}
\]
and the surface tension tensor (of the magnetostrictive nature) by \(2(\lambda_{\text{anis}}/\mu)L_{\text{sur}} \otimes L_{\text{sur}}\).

Let the functions \(G_{kj}(\mathbf{r}, \mathbf{r}')\) \((k, j = x, y)\) be the solutions of the equation:

\[
\Delta G_{kj}(\mathbf{r}, \mathbf{r}') + \nu_{\text{eff}} \nabla_k \nabla_l G_{lj}(\mathbf{r}, \mathbf{r}') = -\delta_{kj}\delta(\mathbf{r} - \mathbf{r}'),
\]

with the following boundary conditions:

\[
(n, \nabla)G_{kj}(\mathbf{r}_{\text{sur}}, \mathbf{r}') + \nu_{\text{eff}} [n_k \nabla_l G_{lj}(\mathbf{r}_{\text{sur}}, \mathbf{r}') + n_l \nabla_k G_{lj}(\mathbf{r}_{\text{sur}}, \mathbf{r}')] = 0.
\]

Here, \(\delta_{kj}\) is the Kronecker symbol, \(\delta(\mathbf{r} - \mathbf{r}')\) is the Dirac delta-function, \(n\) is the surface normal in point \(\mathbf{r}_{\text{sur}}\).

The functions \(G_{kj}(\mathbf{r}, \mathbf{r}')\) coincide with Green tensor for isotropic medium with fixed stresses (accurate within constants). In this case, we can represent the displacement vector as:

\[
u_j(\mathbf{r}) = \frac{2\lambda_{\text{anis}}}{\mu} \int_V G_{kj}(\mathbf{r}, \mathbf{r}') \nabla'_l [L_k(\mathbf{r}')L_l(\mathbf{r}')] \, dV' - \frac{2\lambda_{\text{anis}}}{\mu} \int_S G_{kj}(\mathbf{r}, \mathbf{r}'_{\text{sur}})L_k(\mathbf{r}'_{\text{sur}})L_l(\mathbf{r}_l) \, dS'_l.
\]

Substituting (A5) into energy expression (6) and taking into account boundary conditions (A4), we obtain elastic and magnetoelastic energy contributions:

\[
W_{\text{add}} = \frac{2\lambda_{\text{anis}}^2}{\mu} \int_V \int_V \nabla_m [L_j(\mathbf{r})L_m(\mathbf{r})]G_{kj}(\mathbf{r}, \mathbf{r}') \nabla'_l [L_k(\mathbf{r}')L_l(\mathbf{r}')] \, dVdV' + \frac{2\lambda_{\text{anis}}^2}{\mu} \oint_S L_j(\mathbf{r}_{\text{sur}})L_m(\mathbf{r}_{\text{sur}})G_{kj}(\mathbf{r}_{\text{sur}}, \mathbf{r}'_{\text{sur}})L_k(\mathbf{r}'_{\text{sur}})L_l(\mathbf{r}_l) \, dS_m dS'_l.
\]

Appendix B: Shape-induced contribution into the magnetic energy for an arbitrary constant \(K_{\text{sur}}\)

In the general case, magnetoelastic charges (21) and (22) depend on the ratio \(s = \sigma_{DW}/K_{\text{sur}}\), which we took as a unit when obtained Eqs. (28) and (29). Here, we generalize these expressions for arbitrary values of \(s\).

Substituting (21), (22) and (23), (24) into equation (8), we obtain expressions (27), where coefficients \(K_{2h}^s\), \(K_{4h}^s\) depend on variables \(\varphi^{(\text{in})}\):

\[
K_{2h}^s = K^{m-e} J_1 \left( \frac{a}{b} \right) \frac{\Lambda_+ + \Lambda_-}{\Lambda_+ - \Lambda_-} - \left( 1 + \nu_{\text{eff}} J_2 \left( \frac{a}{b} \right) \right) \frac{\Lambda_+ - \Lambda_-}{\Lambda_+ + \Lambda_-},
\]

\[
K_{4h}^s = K^{m-e} \left[ 2J_2 \left( \frac{a}{b} \right) - 1 \right] \left( 1 - \frac{s(\Lambda_+ + \Lambda_-)}{2\Lambda_+ \Lambda_-} \right) - J_1 \left( \frac{a}{b} \right) \frac{s(\Lambda_+ - \Lambda_-)}{2\Lambda_+ \Lambda_-}.
\]

Here,

\[
\Lambda_\pm \equiv \sqrt{1 + 2s \cos 2(\varphi_{\text{in}} + \psi) + s^2}.
\]

In the limiting case of the small magnetic anisotropy \((s \gg 1)\) both shape-dependent constants vanish:

\[
K_{2h}^s = K^{m-e} J_1 \left( \frac{a}{b} \right) \frac{2}{s} \to 0, \quad K_{4h}^s = -K^{m-e} J_1 \left( \frac{a}{b} \right) \frac{\cos 2(\varphi_{\text{in}} + \psi)}{s} \to 0.
\]

Equation (27) may perform as the minimum condition for the effective energy:

\[
u_{\text{eff}} = -\frac{1}{4} K_{\perp} \cos 4\varphi_{\text{in}} - \frac{1}{2s} K^{m-e} \left[ J_1 \left( \frac{a}{b} \right) (\Lambda_+ - \Lambda_-) + \left( 1 + \nu_{\text{eff}} J_2 \left( \frac{a}{b} \right) \right) (\Lambda_+ + \Lambda_-) \right]
\]

\[
- \frac{1}{12s} K^{m-e} \left[ 2J_2 \left( \frac{a}{b} \right) - 1 \right] \left( 3s \cos 4(\varphi_{\text{in}} + \psi) + 2(\Lambda_+ + \Lambda_-)^3 \right) - 2J_1 \left( \frac{a}{b} \right) (\Lambda_- - \Lambda_+)^3.
\]

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1 M. Petrik and B. Harbrecht, in 26 European Crystallographic Meeting (Darmstadt, Germany, 2010) pp. FA5–MS41–P11

2 M. Petrik and B. Harbrecht, Zeitschrift für Kristallographie Proc. 1, 253 (2011)
X. G. Zheng, H. Kubozono, H. Yamada, K. Kato, Y. Ishiwata, and C. N. Xu, Nature Nanotechn. 3, 724 (2008)
4 M. Petrik and B. Harbrecht, Zeitschrift fur anorganische und allgemeine Chemie 636, 2049 (2010)
5 C. Bahl, L. T. Kuhn, K. Lefmann, P.-A. Lindgard, and S. Morup, Physica B: Cond. Matt. 385-386, Part 1, 398 (2006)
6 L. Néel, C.R. Hebd. Sén. Acad. Sci. 228, 664 (1949)
7 L. Néel, “Low-temperature physics,” (New York: Gordon & Breach, New York, 1962) Summer school of theoretical physics, les houches (1961): Théorie des propriétés magnétiques des grains fins antiferromagnétiques: superparamagnétisme et super-antiferromagnétisme, p. 413, ed. C. de Witt, B. Dreyfus & P.-G. de Gennes
8 R. H. Kodama, A. E. Berkowitz, E. J. McNiff, Jr., and S. Foner, Phys. Rev. Lett. 77, 394 (1996)
9 S. Morup, C. Frandsen, F. Bodker, S. N. Klausen, K. Lefmann, P.-A. Lindgard, and M. F. Hansen, in Mossbauer Spectroscopy, ed. by P. Gütlich, B. W. Fitzsimmons, R. Ruffer, and H. Spiering (Springer Netherlands, 2003) pp. 347–357
10 E. Folven, A. Young, S. T. Retterer, J. E. Boschker, T. Tybell, Y. Takamura, and J. K. Grepstad, Nano Lett. 12, 2386 (2012)
11 E. Folven, A. Scholl, A. Young, S. T. Retterer, J. E. Boschker, T. Tybell, Y. Takamura, and J. K. Grepstad, Phys. Rev. B 84, 220410 (2011)
12 E. Folven, T. Tybell, A. Scholl, A. Young, S. T. Retterer, Y. Takamura, and J. K. Grepstad, Nano Lett. 10, 4578 (2010)
13 D. A. Garanin and H. Kachkachi, Phys. Rev. Lett. 90, 065504 (2003)
14 H. V. Gomonay and V. M. Loktev, Phys. Rev. B 75, 174439 (2007)
15 H. Gomonay, I. Kornienko, and V. Loktev, Cond. Matt. Phys. 13, 23701 (2010)
16 C. Kittel, Rev. Mod. Phys. 21, 541 (1949)
17 M.I. Kaganov, JETP 52, 779 (1980)
18 D. Tobia, E. Winkler, R. D. Zysler, M. Granada, and H. E. Troiani, Phys. Rev. B 78, 104412 (2008)
19 S. K. Mishra and V. Subrahmanyan, ArXiv e-prints (2008), arXiv:0806.1262
20 R. H. Kodama, S. A. Makhloof, and A. E. Berkowitz, Phys. Rev. Lett. 79, 1393 (1997)
21 N. Pérez, P. Guardia, A. G. Roca, M. P. Morales, C. J. Serna, O. Iglesias, F. Bartolomé, L. M. García, X. Batlle, and A. Labarta, Nanotechnology 19, 5704 (2008)
22 R. Narayan, R. Nagarajan, and R. Ranganathan, Phys. Rev. B 69, 054430 (2004)
23 The values of the internal constants in NP could differ significantly (by several times or even orders) from those for the bulk samples13,14 where the role of the surface is negligible.
24 Z. H. Sun, X. Y. Song, F. X. Yan, L. X. Sun, X. K. Yuan, and X. M. Liu, J. Phys. D: Appl. Phys. 42, 122004 (2009)
25 L. D. Landau and E. M. Lifshits, Theory of elasticity, 4th ed., Course of theoretical Physics, Vol. 7 (Fizmatgiz, Moscow, 1987) 246
26 V. Gamm and A. Zhukov, Sov. Solid state physics 22, 3188 (1980)
27 E. A. Brener and V. I. Marchenko, Phys. Rev. Lett. 97, 067204 (2006)
28 M. Petrik and B. Harbrecht, Solid State Phenomena 170, 244 (2011)
29 A. N. Lavrov, S. Komiyama, and Y. Ando, Nature 418, 385 (2002)
30 M. Nishida, T. Hara, M. Matsuda, and S. Ii, Materials Science and Engineering: A 481-482, 18 (2008)
31 E. D. Belokolos, J. Phys. A: Math. Gen. 34, 2331 (2001)
32 G. Catalan, H. Béa, S. Fusil, M. Bibes, P. Paruch, A. Barthélémy, and J. F. Scott, Phys. Rev. Lett. 100, 027602 (2008)