Phenomenological model of surface-induced anisotropy in magnetic nanostructures

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A phenomenological theory of surface-induced magnetic anisotropy has been developed to describe inhomogeneous distribution of surface interactions into the depth of nanowires with circular cross-section and spherical nanoparticles. For ellipsoidal nanoparticles and elliptical nanocylinders, by using the Neel's approach the surface uniaxial anisotropy coefficients have been derived as functions of nanostructure size and aspect ratios. The authors have shown that in elongated nanosystems, this specific anisotropy stabilizes homogeneous magnetic states in the system whereas in spherical nanoobjects, it tends to violate the magnetic state homogeneity and to induce specific inhomogeneous distributions of magnetization. In addition, the main peculiarities of magnetization switching processes in ellipsoidal nanoparticles have been investigated.

Для описания неоднородного распределения поверхностных взаимодействий вглубь нанопроволок с круговым поперечным сечением и сферических наночастиц разработана феноменологическая теория магнитной анизотропии, индуцированной поверхностью. Для эллипсоидальных наночастиц и эллиптических наноцилиндров, используя подход Нееля, определены коэффициенты поверхностной анизотропии как функции размеров и аспектных соотношений. Авторы показывают, что для удлиненных наносистем эта специфическая анизотропия стабилизирует однородные магнитные состояния, в то время как для сферических нанообъектов она стремится разрушить однородность состояний и вызвать особые неоднородные распределения намагниченности. В дополнение, исследованы основные особенности процессов перемагничивания в эллипсоидальных наночастицах.

In magnetic nanostructures, complex processes on surfaces result in a considerable uniaxial anisotropy which usually exceeds the intrinsic anisotropy and favors the magnetization to be oriented perpendicularly to the surface [1]. The main features of the induced anisotropy are strongly influenced by the size and geometrical shape of a nanoobject. For relatively large dimensions, the surface anisotropy (SA) can be successfully considered as a mere surface effect and phenomenologically described as an additional contribution to the surface energy with constant anisotropy coefficient (Néel's ansatz [2, 3]). But as the dimensions of a nanosystem decrease, the phenomenological theory of surface-induced uniaxial anisotropy should reflect the spatial variation of surface interactions into the depth [4]. Moreover, in elongated samples, SA manifests itself as an easy axis anisotropy and may fix the magnetization in preferable directions, suppresing the demagnetization.

In this paper, we apply the phenomenological model introduced in [5] to obtain the SA distributions for spherical nanoparticles and circular nanowires. For ellipsoidal nanoparticles and nanopillars with elliptical cross-section, we derive the values of SA coefficients as functions of aspect ratios and sizes. In addition, we investigate the peculiarities of magnetization processes in ellipsoidal nanoparticles.

Within the phenomenological theory, we assume that possible distributions of SA coefficient $K(\mathbf{r})$ can be described by an inhomogeneous "order-parameter- field derived from a general interaction functional independently from the magnetic subsystem [4]:

$$W = \int_V \left(a \left[(\nabla K)^2 + K^2 \sum_{i,j} (\partial_i n_j)^2 \right] + f_0(K) \right) dV =$$

$$= a \int_V \left(\left(\frac{dK}{dr} \right)^2 + K^2 G(\theta; r) + \frac{K^2}{\lambda^2} \right) dV. \tag{1}$$

Here, the first and the second term in (1) describe the stiffness of the field $K(\mathbf{r})$ with $\mathbf{n}$ being a unit vector perpendicular to the surface. $G(\theta; r) = \sin^2 \theta / r^2, dV = 2\pi r h dr$ in cylindrical symmetry; $G(\theta; r) = 2/r^2, dV = 4\pi r^2 dr$ for spherical symmetry. The third term $f_0(K)$ includes non-gradient terms of the interaction functional. We expand this contribution as $f_0(K) = \sum \alpha_n K^n$ and keep only the square term which plays the main role to describe the induced anisotropy.

$\lambda = \sqrt{a/\alpha}$ is the characteristic length in our theory. a, α are the stiffness constants which characterize the system resistance against the surface-induced interactions. In principle, the values of a and α can be derived from *ab initio* calculations of induced spin-orbit coupling in magnetic nanolayers [6]. In experiment, these parameters can be obtained only indirectly, for example, from the thickness dependences of the function $\Phi(d) = K_{eff} d$ where K_{eff} is the effective anisotropy, and d is either the film thickness for magnetic nanolayers or the radius for nanowires and nanoparticles. In [7, 8] the corresponding estimates resulted in $\lambda = 1.9A$ for Co/Au films and $\lambda = 26.4; 31.9A$ for Ni/Cu multilayer systems. Modern experimental techniques allow to observe the decrease of the induced magnetic anisotropy into the depth of magnetic nanolayers [9]. These methods can be used to obtain the stiffness constants values.

For a spherical nanoparticle, the minimization of (1) yields the solution (Fig.1(a)):

$$K_s(r) = K_s(R) \frac{R^2}{r^2} \frac{(-r \cosh(r/\lambda) + \lambda \sinh(r/\lambda))}{(-R \cosh(R/\lambda) + \lambda \sinh(R/\lambda))}. \tag{2}$$

For circular nanowires with a large aspect ratio height/radius, we subdivide the SA induced by lateral surface into two independent contributions:

$$w_s = K_{s1}(r)(\mathbf{m} \cdot \widehat{\mathbf{n}}_z)^2 - K_{s2}(r)(\mathbf{m} \cdot \widehat{\mathbf{n}}_r)^2. \tag{3}$$

The first term in Eq.(3) represents the uniaxial anisotropy while the second one has radial symmetry. Both characterize orientational effects. The distributions $K_{s1}(r)$ and $K_{s2}(r)$ can be obtained by independent minimization of functional (1) with $G(0; r) = 0$ and $G(\pi/2; r) = 1/r^2$, respectively:

$$\left. \begin{array}{l} K_{s1}(r) = K_{s1}(R) \dfrac{J_0(ir/\lambda)}{J_0(iR/\lambda)} \\[2mm] K_{s2}(r) = K_{s2}(R) \dfrac{J_1(ir/\lambda)}{J_1(iR/\lambda)} \end{array} \right\}, \tag{4}$$

where J_0, J_1 are the Bessel functions of first kind.

Depending on the ratios R/λ, the profiles (2) and (4) either vary smoothly within the nanoobject ($R \approx \lambda$) or essentially confined to the near-surface region ($R >> \lambda$, Fig.1). The radial components $K_s(r), K_{s2}(r)$ are zero in the nanoobject center (Fig.1a) whereas the axial component $K_{s1}(r)$ may be finite (Fig.1b).

The surface induced anisotropy may influence dominantly the magnetic properties and cause spin-reorientation transitions from the homogeneous magnetization state to a specific inhomogeneous distribution. In this case, the induced anisotropy aligns magnetic moments on the surface either along the normal $\mathbf{n}$ ($K_s(R) > 0$) or locally in the plane ($K_s(R) < 0$). General features of vortex states stabilized by surface anisotropy in cylindrical wires and spherical nanoparticles can be found elsewhere [10].

In elongated nanostructures (namely, in nanoellipsoids and in elliptical nanopillars), the SA plays the part of easy axis anisotropy and for systems with $K_s > 0$ favors the magnetization direction along
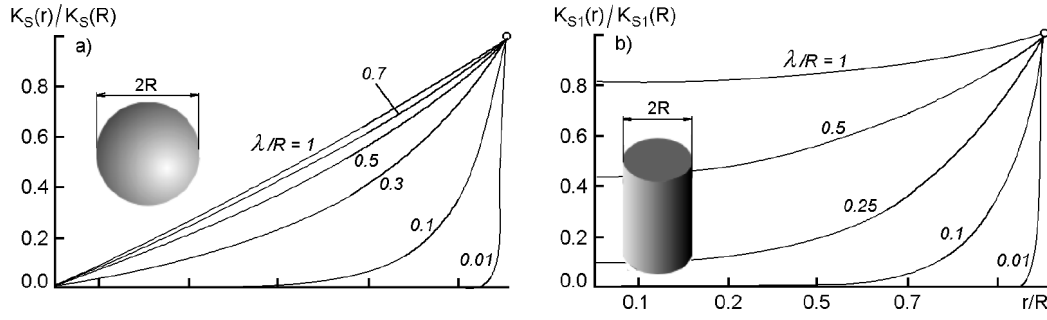
Fig. 1. Inhomogeneous distributions of the surface anisotropy coefficients into the depth of spherical nanoparticle (a) and circular nanowire (b) according to Eqs.(2),(4) for different ratios λ/R as indicated.
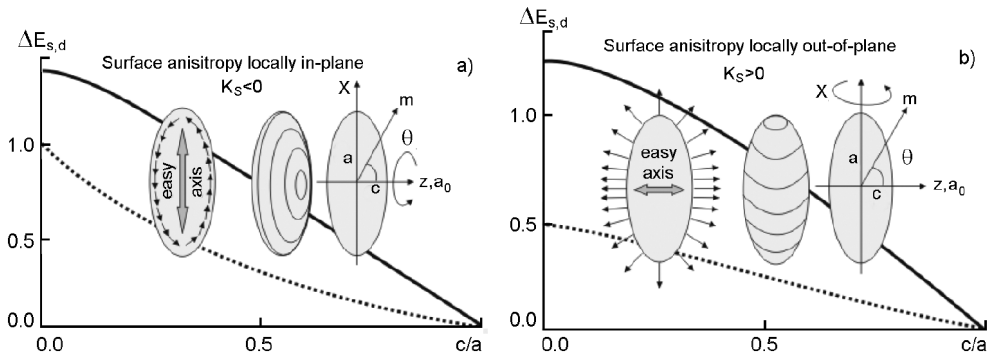

Fig. 2. Comparison of the demagnetization energy ΔE_d (dotted line) and the induced anisotropy energy ΔE_s (full line) for prolate (a) and oblate (b) ellipsoids.

the shorter dimension, i.e. stabilizes homogeneous states (see schematic distribution in Fig.2b). Thus, the uniaxial anisotropy weakens demagnetizing influence of surface and may completely suppress it. For systems with $K_s < 0$, the easy SA axis is along the longer dimension and strengthens the demagnetizing effects (see the scheme in Fig. 2a). In addition, the SA can be strongly increased by coating of magnetic nanosystems with various nonmagnetic materials [1].

Further, we shall consider SA with constant anisotropy coefficient $K_s > 0$ (the Néel's approach). Therefore, the energy density can be reduced to a functional form of a second-order anisotropy $w_s = -(K_s/c)[\Lambda_1(\mathbf{m} \cdot \mathbf{a_0})^2 + \Lambda_2(\mathbf{m} \cdot \mathbf{b_0})^2 + \Lambda_3(\mathbf{m} \cdot \mathbf{c_0})^2]$ (for ellipsoid with the unit vectors $\mathbf{a_0}, \mathbf{b_0}, \mathbf{c_0}$ along the ellipsoid axes), $w_s = (-K_{s1}/c)[\Lambda_1(\mathbf{m} \cdot \mathbf{a_0})^2 + \Lambda_2(\mathbf{m} \cdot \mathbf{c_0})^2]$ (for radial surface anisotropy of elliptical nanocylinder, $\mathbf{a_0}, \mathbf{c_0}$ are unit vectors along the elliptical cross-section axes). The "shape factors"Λ_i are written as elliptic functions of the ellipsoid sizes [11]. For ellipsoids of revolution with the eccentricity $k = \sqrt{1 - c^2/a^2}$, the coefficients Λ_i can be expressed by means of elementary functions [11].

Here, we compare the differences of demagnetizing factors $\Delta E_d = (N_a - N_c)$ [12] and shape factors $\Delta E_s = \Lambda_c - \Lambda_a$ [11] for a prolate ellipsoid (Fig.2a):

$$\Delta E_s = 3\frac{2k(k^2 - 3) + (k^4 + 2k^2 - 3)\log\left(\frac{1-k}{1+k}\right)}{8k^3} \tag{5}$$

and for oblate ellipsoid (Fig.2b):

$$\Delta E_s = 3\frac{k(3 - 2k^2)\sqrt{1 - k^2} + (4k^2 - 3)\arcsin(k)}{4k^3}. \tag{6}$$
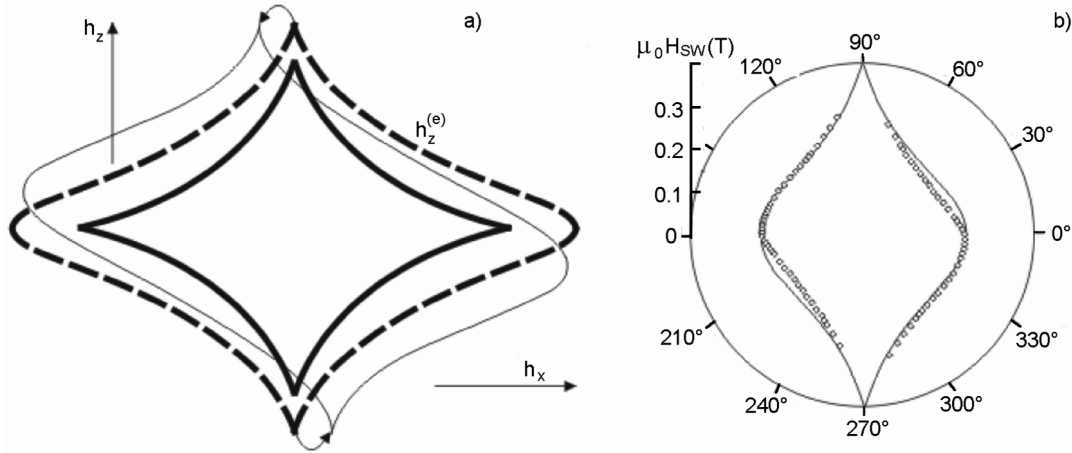
Fig.3. a) The mapping of the Stoner-Wohlfarth astroid (thick solid line) onto the plane of external field components (dashed line). The thin solid line is for the case $N_{xz} \neq 0$ (read text for details). b) The switching field H_{sw} of the BaFeCoTiO nanoparticle as a function of the angle θ (Data are taken from W.Wernsdorfer et al.[17]).

For elliptical nanocylinders with dimensions (a, c) in the cross-section and an aspect ratio $s = c/a$, we obtain:

$$\Delta E_s = \left[2s^2(1 + s^2)E\left[\frac{s^2 - 1}{s^2}\right] + 4s^2 K\left[\frac{s^2 - 1}{s^2}\right] - \right.$$
$$\left. - 2s((1 + s^2)E[1 - s^2] - 2s^2 K[1 - s^2])\right]/s(1 - s^2), \tag{7}$$

where K, E are elliptical integrals of first and second kind.

Both the demagnetizing factors N_i and the "shape factors"of ellipsoid and cylinder are geometrical factors, which depend only on the ratio of the geometrical axes. However, their impacts on the magnetic states are fundamentally different. The SA energy w_s is caused by short-range internal (local) magnetic interactions. On the contrary, the demagnetizing fields are due to the sample magnetization and have long-range (non local) character.

Finally, consider some peculiarities of the magnetization processes in elongated nanoparticles caused by the uniaxial SA. The equilibrium homogeneous magnetic states in the nanoparticle are formed as a result of the competition between internal magnetic field and SA [13, 14]. To elucidate the main features of such magnetic states, we consider switching processes in the xz plane. In this case, the reduced energy density of the system $\Phi(\theta) = (w \cdot c)/(K_s \Delta E_s)$ can be written as a function of the angle θ between $\mathbf{m}$ and $\mathbf{a_0}$:

$$\Phi(\theta) = -\cos^2 \theta - h \cos(\theta - \psi), \tag{8}$$

where $h = H/H_c \left(H_c = K_s \Delta E_s/(cM_0) \right)$ is the reduced internal magnetic field which makes angle ψ with $\mathbf{a_0}$ axis ($h_x = h \sin \psi$, $h_z = h \cos \psi$). The solutions of θ that minimize the potential $\Phi(\theta)$ (8) compose a phase diagram in components of internal magnetic field (h_x, h_z) (the famous Stoner-Wohlfarth astroid [15], thick solid line in Fig.3a).

To map this critical line on the plane spanned by the components of external magnetic field $(h_x^{(e)}; h_z^{(e)})$ one has to use the following relation [13, 14]:

$$\mathbf{h}^{(e)} = \mathbf{h} + \frac{4\pi c}{K_s \Delta E_s} \widehat{N} \mathbf{m}(\mathbf{h}) \tag{9}$$

($\widehat{N}$ is the demagnetizing tensor). The critical line $\mathbf{h}^{(e)}$ (dashed line in Fig.3a) is given by the analytical expression:

$$h_z^{(e)} = (1 - h_x^{2/3})^{3/2} + 4\pi N_{zz} \cos \theta. \tag{10}$$

The magnetic phase diagram in Fig.3a describes the main features of the model (8) as applied for single-domain nanoparticles and provides a basis for detailed analysis of the remagnetization processes (see e. g. [13, 14]). In particular, astroids $h_z^{(e)}$ (Fig.3a) with rounded edges have been observed in fcc Co [16] and BaFeCoTiO nanoparticles [17] (Fig.3b). In nanoparticles with misaligned anisotropy axes and ellipsoid geometrical ones (i.e. when $N_{xz} \neq 0$), the critical line $h_z^{(e)}$ undergoes a deformation with oblique sharp edges (thin solid line in Fig.3a).

To conclude, we have developed a simple phenomenological model of induced uniaxial surface anisotropy in magnetic nanoparticles and nanowires and have highlighted the main features of this specific enhanced anisotropy. For ellipsoidal nanoparticles and elliptical nanopillars, we have shown that SA stabilizes homogeneous states in the system suppressing demagnetizing effects. The surface anisotropy coefficients have been derived as functions of aspect ratios and nanoobject sizes. For spherical nanoparticles and circular nanocylinders, we have obtained the distribution of SA coefficient into the depth of a nanoobject and found out that surface anisotropy in this case may cause the inhomogeneous states of the magnetization.

Our results are in accordance with existing experimental data on switching processes in elongated magnetic particles [16, 17]. Note, that recent advances in nanometer scale technology (production of nanoparticles with different shapes [18, 19], different sizes and aspect ratios, with high perfection [20]) allow to study different aspects of surface magnetism and should confirm numerous effects predicted in our paper.

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Феноменологічна модель поверхневої анізотропії у магнітних наноструктурах

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Щоб описати неоднорідне розповсюдження поверхневих взаємодій у глибину нанопроволок з круговим поперечним перерізом і сферичних наночасток, разроблено феноменологічну теорію магнітної анізотропії, що индуцюється поверхнею. Для еліпсоїдальних наночасток і еліптичних наноциліндрів, використовуючи підхід Неєля, визначені коэфіцієнти поверхневої анизотропії як функції розмірів та аспектних співвідношень. Автори доводять, що ця специфічна анізотропія стабілізує однорідні магнітні стани, в той час як для сферичних нанооб'єктів вона намагається зруйнувати однорідність станів та викликати особливі неоднорідні розподідення намагнінення. Крім того, досліджено основні особливості процесів перемагнічення в эліпсоїдальних наночастках.