

Strain-Gradient-Induced Unidirectional Magnetic Anisotropy in Nanocrystalline Thin Permalloy Films


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Herein, the effect of inhomogeneous elastic deformation on the magnetic anisotropy of Ni_{71.5}Fe_{28.5} wt% nanocrystalline films is investigated. The in-plane controlled strains are induced in the film by bending of a glass substrate which has a thickness step in the middle. Ferromagnetic resonance measurements reveal the existence of in-plane unidirectional magnetic anisotropy. The anisotropy behavior directly correlates with the calculated strain gradients. It is shown that this correlation is well explained by the flexomagnetic effect, which establishes the relation between the magnetization and the inhomogeneous strains. The experimental value of the flexomagnetic coefficient for the thin Ni_{71.5}Fe_{28.5} wt% film is $1.5 \times 10^{-3} \text{ T m}$.

The flexoeffects are electromagnetomechanical effects in which the electric polarization or magnetization exhibits a linear response to inhomogeneous mechanical impact (elastic strain or stress gradient). Among these effects, the most studied one is flexoelectricity. The coupling between electric polarization and strain gradient in crystals was theoretically investigated as early as the 1960s by Mashkevich,^[1] Tolpygo,^[2] and Kogan.^[3] However, up to the end of the 1990s, the interest in flexoelectricity was very limited due to the small magnitude of the effect in bulk materials. Since the beginning of the 2000s, the situation has cardinally changed because of the considerable progress both in the field of the experimental study of the flexoelectric effect and in the field of the synthesis of new materials. The first systematic experimental studies on flexoelectricity in ferroelectric ceramics done by Ma and Cross,^[4–6] and Zubko et al.^[7] showed that the magnitude of the flexoelectric effect was much larger than was expected from the earlier theoretical estimates. This allowed Cross and coworkers^[8,9] to utilize the flexoelectric effect for the development of piezoelectric composites whose piezoelectric response was comparable with that of commercial piezoelectrics.

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 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/pssr.201900467>.

DOI: 10.1002/pssr.201900467

Recent developments in nanotechnology have also contributed to a significant increase in interest in flexoelectricity since large strain gradients possible at the nanoscale can result in significant flexoelectric coupling. The flexoelectric response of different nanostructures was studied in the works by Catalan et al.,^[10,11] Sharma and coworkers,^[12,13] Kalinin and Meunier,^[14] and Lee et al.^[15] In particular, it was shown that because of the lattice mismatch between thin ferroelectric films and the substrate, the large strain gradients can be induced in the films (up to 10^6 – 10^7 times larger than in bulk monocrystals^[15]), and the flexoelectric response of such films

significantly affects their functional properties. The recent discovery of phase coexistence in highly strained BiFeO₃ thin films attracted extra attention to flexoelectricity due to the enormous strain gradients (about 10^7 m^{-1}) arising from the lattice parameter mismatch at the boundary between the tetragonal-like and rhombohedral-like phases.^[16,17] Currently, the role of the flexoelectric effect in the physics of dielectrics and semiconductors is widely recognized, and the effect itself shows considerable promise for practical applications.^[18]

In the context of major achievements in the field of flexoelectricity, it seems quite surprising that flexomagnetic effect, which describes the coupling between magnetization and strain gradients, remains a largely unexplored phenomenon. There have been only few studies on the subject published to date.^[19–24] Lukashev and Sabirianov^[19] performed first-principles calculations of the flexomagnetic coefficient for the antiperovskite Mn₃GaN, which was about $0.2 \mu_B \text{ nm}$. Eliseev et al.^[20,21] theoretically showed that in the case of infinite medium, the flexomagnetic effect could exist in 69 of 90 magnetic classes, whereas in samples of finite size near the surface, it can exist for any magnetic class. Theoretical modeling of the flexomagnetic effect in a piezomagnetic nanobeam subjected to bending was reported in the study by Sidhardh and Ray.^[22] Experimental studies of the flexomagnetic effect are even more scarce. Up to now, only two works have been published, which present experimental evidences in support of the flexomagnetic effect. Zhang et al.,^[23] while investigating mixed-phase boundaries in perovskite BiFeO₃ thin films, have discovered a fivefold increase in spontaneous magnetization (up to 30 – 40 kA m^{-1}) in the vicinity of phase boundaries where the maximum strain gradients were formed. The authors related the observed increase in magnetization to the flexomagnetic effect and estimated its coefficient as $\approx 4 \mu_B \text{ nm}$. In another study of mixed-phase perovskite, La-5%-doped BiFeO₃, it was found that in those regions of the sample

where electric field-induced elongated stripes of the mixed-phase boundary were formed, the antiferromagnetic axis was oriented perpendicular to the elongation axis.^[24] Theoretical analysis showed that this magnetic behavior could be explained by the magnetic anisotropy induced near the phase boundaries by a shift of Fe ions with respect to the oxygen cage caused by strain gradients.

However, there is still no direct convincing proof of the existence of the flexomagnetic effect, and there are also no direct experimental measurements of its magnitude. In this Letter, we associate the strain gradient-induced unidirectional magnetic anisotropy revealed in the Ni_{71.5}Fe_{28.5} wt% nanocrystalline thin film with the manifestation of the flexomagnetic effect.

The magnetomechanical coupling is phenomenologically described by adding additional terms to the expression for thermodynamic potential density, i.e., the free energy density of the magnetostriction coupling F_{strict} and flexomagnetic coupling F_{flexo} ^[21]

$$F_{\text{me}} = F_{\text{strict}} + F_{\text{flexo}} = -q_{ijkl}u_{ij}M_kM_l + \frac{Q_{ijkl}}{2} \left(\frac{\partial u_{ij}}{\partial x_k} M_l - u_{ij} \frac{\partial M_l}{\partial x_k} \right) \quad (1)$$

where $M_{k,l}$ are components of the magnetization vector, u_{ij} are strain tensor components, q_{ijkl} are magnetostriction tensor component, and Q_{ijkl} are components of the flexomagnetic coupling tensor. Restricting our consideration to the specific case of the in-plane uniformly magnetized, elastically and magnetically isotropic thin film subjected to a bending uniaxial strain u along the Ox axis, Equation (1) can be transformed into the following form

$$F_{\text{me}} = -quM_x^2 + \frac{Q}{2} \frac{\partial u}{\partial x} M_x = -\mu_0 \frac{H_{2u}M_s}{2} \cos^2 \varphi - \mu_0 H_{1u}M_s \cos \varphi \quad (2)$$

where M_s is the saturation magnetization of the film, $\mu_0 = 4\pi \times 10^{-7} \text{ T m A}^{-1}$ is the magnetic constant, and φ is an in-plane angle between the magnetization vector and the Ox axis. This expression reflects a well-known fact^[25] that the magnetostriction contribution to the thermodynamic potential results in a uniaxial magnetic anisotropy, whose effective field is

$$H_{2u} = -\frac{1}{\mu_0} \frac{\partial F_{\text{strict}}}{\partial M_x} = \frac{2quM_s}{\mu_0} = \frac{3\lambda_s}{\mu_0 M_s} \frac{E}{1+\nu} u \quad (3)$$

where λ_s is the magnetostriction constant, E is the Young's modulus, and ν is the Poisson's ratio of the film. Note, however, that according to Equation (2), the flexomagnetic effect in this case should manifest itself by a unidirectional magnetic anisotropy, whose effective field is

$$H_{1u} = -\frac{1}{\mu_0} \frac{\partial F_{\text{flexo}}}{\partial M_x} = -\frac{Q}{2\mu_0} \frac{\partial u}{\partial x} = -\frac{Q}{2\mu_0} \nabla u \quad (4)$$

To verify this fact, we conducted a specially organized experiment.

An investigated magnetic film sample was produced by a vacuum thermal deposition of permalloy on polished glass $8 \times 24 \text{ mm}^2$ size substrate. As a source material, we chose Ni₇₁Fe₂₉ wt% alloy with the relatively large positive

magnetostriction constant $\lambda_s \approx 17 \times 10^{-6}$ ^[26] and the saturation magnetization $M_s \approx 1050 \text{ kA m}^{-1}$.^[27] To reduce the effect of substrate surface roughness on the magnetic film properties, the substrate was preliminarily covered by a 500 nm-thick SiO layer. To provide a controlled strain gradient in the film plane, the substrate, as shown in **Figure 1**, had a special profile—a thickness step in the middle, with the left side of the substrate having a thickness of 2 mm and the right one 1 mm. In addition, pockets of the size $0.5 \times 1 \text{ mm}^2$ designed for special fastenings were made on top of the substrate at the edges that were parallel to its short side (see **Figure 1**). Thus, the total size of the deposited film was $8 \times 22 \text{ mm}^2$. During the deposition process, the substrate temperature of 200°C was maintained, and an external magnetic field $\approx 16 \text{ kA m}^{-1}$ was applied in the film plane along its long side. The base pressure was lower than 10^{-8} bar, and the deposition rate was 1 nm s^{-1} . The nanocrystalline structure of the sample with the average crystallites' sizes of 8 nm was determined by transmission electron microscopy, whereas X-ray fluorescence analysis showed that the thickness of the film was 70 nm, and its composition was Ni_{71.5}Fe_{28.5} wt%. The electron diffraction pattern indicated a random distribution of the crystallites in the sample. As shown in the study by Herzer,^[28] such nanocrystalline materials with crystallite sizes much smaller than the exchange length have a very low coercivity due to magneto-crystalline anisotropy averaging.

To create an in-plane strain in the film, we put the sample into a specially designed holder that provided controlled bending of the substrate. The left half of the substrate was placed on a flat metal base, and the left edge of the substrate was pressed to the base with screws and a clamp (**Figure 1**). The right edge of the substrate was shifted downward by a distance of $h_z = 140 \mu\text{m}$ with a special bracket (not shown in **Figure 1**). As a result, the substrate was bent (as schematically shown in **Figure 1**), thereby inducing strain gradients in the film which we calculated using the finite element software COMSOL Multiphysics.

Magnetic properties on local areas of the Ni_{71.5}Fe_{28.5} wt% film were determined using the scanning spectrometer of ferromagnetic resonance (FMR),^[29] which allows recording FMR spectra for different orientations φ_H of the sweeping magnetic field H . The locality of measurements was 1 mm. The measurements of the sample were performed with a spatial step of 1 mm, whereas near the thickness step of the substrate, the spatial step of measurements was 0.5 mm. The microwave excitation frequency was

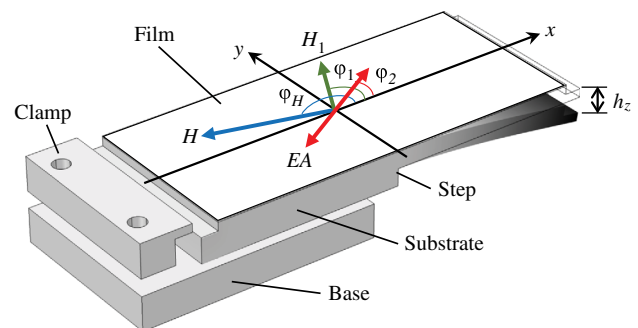


Figure 1. Sketch of a holder used to create controlled strains in the film by substrate bending and the coordinate system used in the FMR measurements and data analysis.

2274 MHz. To eliminate the magnetization nonuniformity, before each measurement the field $H = 23.9 \text{ kA m}^{-1}$ was applied in the film plane, and the FMR spectrum was recorded in the reverse sweep mode of the magnetic field.^[29] Magnetization saturation M_s and parameters of the in-plane uniaxial (field H_2 and easy axis [EA] magnetization direction φ_2) and unidirectional (field H_1 and direction φ_1) magnetic anisotropy were determined from the obtained angular dependences of the resonance field by fitting parameters of a theoretical model of a single-domain film to the experimental data.^[30,31] The coordinate system and the anisotropies orientations are shown in Figure 1.

In Figure 2 symbols show experimental angular dependences of the resonance field $H_R(\varphi_H)$ and FMR linewidth $\Delta H(\varphi_H)$, measured along the long axis of the bent sample at three points with coordinates $x = -1 \text{ mm}$ (square symbols), 0 mm (circle symbols), and 1.5 mm (triangle symbols). Solid lines in this figure show theoretical dependences. They coincide well with the experiment except for the FMR linewidth angular dependence $\Delta H(\varphi_H)$ at the point $x = -1 \text{ mm}$. This minor discrepancy for $\Delta H(\varphi_H)$ apparently indicates the existence of an additional relaxation mechanism at this point. An analysis of $H_R(\varphi_H)$ dependences reveals that at the central point $x = 0 \text{ mm}$ of the film only the uniaxial magnetic anisotropy H_2 presents. However, at points to the left and right of the central point, in addition to uniaxial anisotropy, the unidirectional magnetic anisotropy H_1 exists. At the point $x = -1 \text{ mm}$, the magnitude of H_1 is $\approx 0.14 \text{ kA m}^{-1}$ and

the direction of unidirectional anisotropy φ_1 is $\approx 180^\circ$. Furthermore, although the magnitude of unidirectional anisotropy at the point $x = 1.5 \text{ mm}$ is almost seven times smaller ($H_1 \approx 0.02 \text{ kA m}^{-1}$), it is clearly seen that it has an opposite direction $\varphi_1 \approx 0^\circ$.

The overall picture of the magnetic anisotropy parameters distribution across the entire area of the inhomogeneously strained film is shown in Figure 3. It is shown that both the uniaxial magnetic anisotropy H_2 (Figure 3a) and the unidirectional anisotropy H_1 (Figure 3b) vary primarily in one dimension parallel to the Ox axis, whereas along the width of the sample, these variations are insignificant. For analysis, we averaged magnetic characteristics along the Oy axis and showed the results in Figure 4. The error bars on these plots show the standard deviation of the averaged values.

To analyze and interpret the obtained data, we calculated the distribution of strain tensor components across the thin-film area using COMSOL Multiphysics. The absolute value of the longitudinal component of the strain tensor u_{xx} was more than an order of magnitude larger than values of in-plane components u_{xy} and u_{yy} . In addition, the variation of u_{xx} component along the Oy axis was negligibly small compared with the variation along the Ox axis. Therefore, our assumption that the bending-induced strains in the film are uniaxial and 1D is quite reasonable. The longitudinal dependences of the film's uniaxial strain $u(x) = u_{xx}(x)$ and its derivative $du(x)/dx$ are shown in Figure 4a,b.

The calculated dependence $u(x)$ correlates quite well with the dependence of the uniaxial magnetic anisotropy field $H_2(x)$, which is directly consistent with Equation (3). Note, however, that in the film, in addition to the uniaxial magnetic anisotropy H_{u2} induced by strain, a uniaxial magnetic anisotropy H_k was formed by an external magnetic field applied during deposition along the Ox axis. The magnitude of this anisotropy was $H_k \approx 0.8 \text{ kA m}^{-1}$, as determined from measurements of the same sample but without strain. Since the magnetostriction

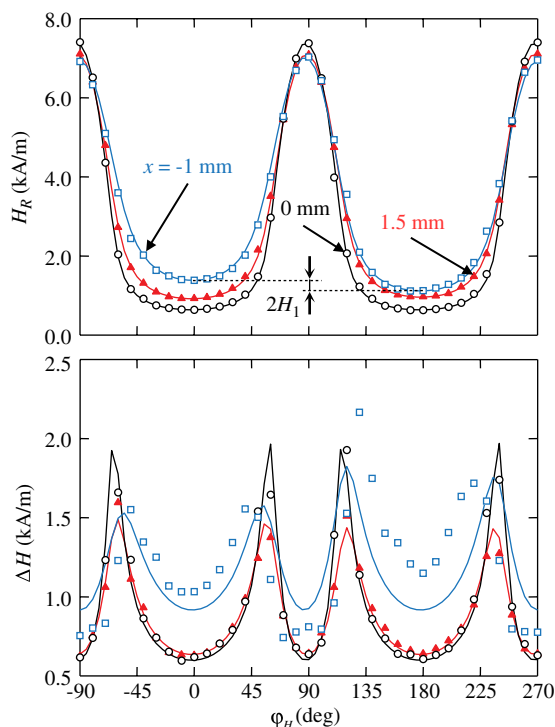


Figure 2. Dependences of the resonance field H_R (top) and FMR linewidth ΔH (bottom) on the sweeping field direction φ_H for the $\text{Ni}_{71.5}\text{Fe}_{28.5}$ wt% film, obtained from three local areas of the sample with coordinates $y = 0$ and $x = -1 \text{ mm}$ (square symbols), 0 mm (circle symbols), and 1.5 mm (triangle symbols). Symbols correspond to the experimental results, whereas lines are theoretical fits.

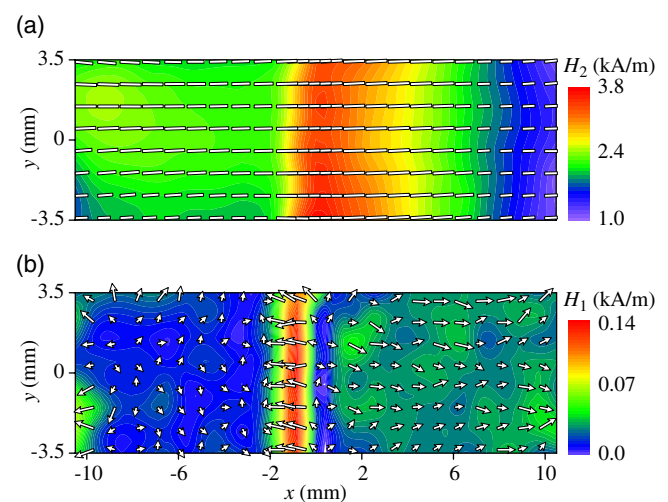


Figure 3. Distribution of parameters of the a) uniaxial and b) unidirectional magnetic anisotropy across the area of the thin $\text{Ni}_{71.5}\text{Fe}_{28.5}$ wt% film under the inhomogeneous strain caused by bending of the substrate with the thickness step profile. White bars and arrows show orientations of the easy axis magnetization and unidirectional anisotropy, respectively.

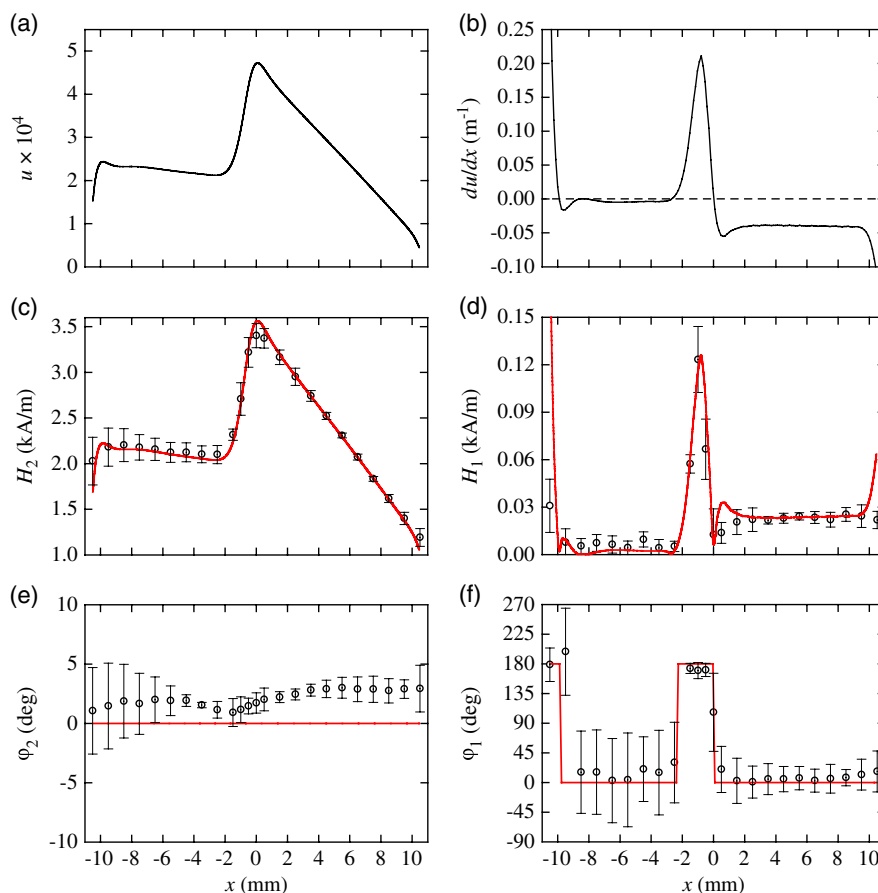


Figure 4. Dependences of the film's parameters averaged along the Oy axis on x coordinate. a,b) Uniaxial strain $u(x)$ and its derivative $du(x)/dx$. c,d) Uniaxial H_2 and unidirectional H_1 magnetic anisotropy field. e,f) Directions of EA (φ_2) and unidirectional anisotropy (φ_1). Symbols are experimental data, and lines are calculations.

constant λ_s for the composition of the considered film is positive, and $u > 0$, the observed uniaxial magnetic anisotropy field H_2 in the experiment is just a result of a simple sum $H_2(x) = H_k + H_{u2}(x)$, as shown in the study by Belyaev and Izotov.^[32] The calculated dependence $H_2(x)$ is shown in Figure 4c by a line. We used the following parameters for the theoretical model of the film: the saturation magnetization $M_s = 1022 \text{ kA m}^{-1}$ and the magnetostriction constant $\lambda_s = 16.3 \times 10^{-6}$ (experimental values determined for the strained sample), the Young's modulus $E = 200 \text{ GPa}$, and the Poisson's ratio $\nu = 0.3$ (standard values for the permalloy film taken from ref. [33]). The results of the calculations are in good agreement with the experimental data. A slight deviation of the experimental easy axis direction from the theoretical values (line in Figure 4e) is probably related to a certain misorientation of H_k and H_{u2} easy axes.^[32]

Of most interest is the behavior of the unidirectional magnetic anisotropy which, as shown in Figure 4b,d,f, directly correlates with the strain gradient. The direct relationship between the strain gradient and unidirectional anisotropy is also confirmed by a calculation using Equation (4). Theoretical curves (solid lines in Figure 4d,f), which show the magnitude and direction of the strain gradient-induced unidirectional anisotropy, agree well

with the experiment. A small discrepancy between theory and experiment observed at the film edges is associated with the difficulty to consider in the model the degree of influence of the real clamping of the substrate edges when calculating strains in the investigated sample.

The experimental data strongly suggest that the unidirectional magnetic anisotropy revealed in the $\text{Ni}_{71.5}\text{Fe}_{28.5}$ wt% film comes from the flexomagnetic effect. By fitting the flexomagnetic coupling parameter for the best agreement between theory and experiment, we obtained the flexomagnetic coefficient Q to be $1.5 \times 10^{-3} \text{ T m}$. The value $Q/\mu_0 \approx 1.2 \text{ kA}$ is 10^7 times larger than the theoretical value of the flexomagnetic coefficient obtained by Lukashev and Sabirianov by first-principles calculations but for antiperovskite Mn_3GaN ^[19] and the theoretical estimate for the flexomagnetic coefficient by Eliseev et al.^[21] Our obtained flexomagnetic coefficient is also $\approx 10^7$ larger than the value estimated from the experimental data for perovskite BiFeO_3 .^[23] As the flexomagnetic effect is a poorly explored phenomenon, there are almost no experimental data to compare with. We should admit that this enormous discrepancy in the magnitude between our obtained flexomagnetic coefficient and the previous theoretical and experimental estimates is perplexing. We hypothesize that this difference could be 1) due to the large magnetization of

permalloy compared with that of the perovskite BiFeO₃ and anti-perovskite Mn₃GaN and 2) due to giant strain gradients that might form at the grains boundaries of the nanocrystalline structure of the film subjected to bending that we did not consider in our calculations of strain.

In conclusion, we would like to emphasize that in this article, we experimentally revealed a new phenomenon—the unidirectional magnetic anisotropy induced by elastic strain gradients. Unlike the well-studied unidirectional anisotropy caused by the exchange coupling at the interface in the ferromagnetic/antiferromagnetic systems,^[34] as well as the unidirectional anisotropy in weak ferromagnets associated with the Dzyaloshinsky–Moriya interaction,^[35] the unidirectional magnetic anisotropy induced by strain gradients is a new phenomenon that has not been previously reported. We showed that this unidirectional anisotropy is caused by the flexomagnetic effect, which describes the coupling between magnetization and inhomogeneous strains. The experimental value of the flexomagnetic coupling coefficient Q for the Ni_{71.5}Fe_{28.5} wt% film was 1.5×10^{-3} T m. The obtained large value of the flexomagnetic coefficient Q in addition to giant strain and stress gradients that usually emerge due to the inhomogeneity of nanocrystalline structure indicates a substantial role of flexomagnetism in the formation and determination of the magnetic properties of nanocrystalline thin films. Therefore, we believe that flexomagnetic effect and unidirectional magnetic anisotropy induced by inhomogeneous strains should be the subject of intensive experimental and theoretical researches.

Acknowledgements

This work was supported by the Ministry of Education and Science of the Russian Federation, project no. RFMEFI60417X0179.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

flexomagnetic effect, strain gradients, unidirectional magnetic anisotropy

Received: August 20, 2019

Revised: September 27, 2019

Published online:

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