Domain structure in biaxial Fe/Cr films induced by lateral fluctuations of the magnetic anisotropy

A. A. Rzhovsky
Institut für Festkörperforschung IFF-9 “Elektronische Eigenschaften,” Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany and Ioffe Physical Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

B. B. Krichevtsov
Ioffe Physical Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

D. E. Bürgler and C. M. Schneider
Institut für Festkörperforschung IFF-9 “Elektronische Eigenschaften,” Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

(Received 3 November 2007; revised manuscript received 9 April 2008; published 30 May 2008)

The magnetic microstructure of single-crystalline Fe(001) thin films has been studied by magnetic second harmonic generation and conventional magneto-optical Kerr effect methods. The layers were grown on GaAs/Fe/Ag(100) substrates, capped by a Cr overlayer, and displayed a fourfold in-plane magnetic anisotropy.

I. INTRODUCTION

The magnetic properties of the epitaxial Fe/Cr film system as well as the behavior of exchange-coupled Fe/Cr/Fe structures based on it have been the subject of intensive studies for a long time. This material combination was the first to exhibit the giant magnetoresistance (GMR) effect, a discovery that started off the field of spintronics. GMR is still of major interest for applications and devices in spintronics, and has been probed by the method of magnetization-induced second harmonic generation and conventional magneto-optical Kerr effect methods. The layers were grown on GaAs/Fe/Ag(100) substrates, capped by a Cr overlayer, and displayed a fourfold in-plane magnetic anisotropy.

We observe the formation of a multidomain structure at magnetic fields \( |H| \leq 0.5 \) kOe, when the field is applied in a narrow range of azimuthal angles close to the hard axis direction \( (\Delta \xi < 1^\circ) \). The domains are characterized by the same longitudinal magnetization component \( M_z \) and transverse components \( M_y \) of opposite signs. We developed an approach to extract the change of the relative contributions of domains with different magnetization directions during magnetization reversal from the experimental data. The formation of a domain structure can be explained by taking into account the lateral fluctuations of the magnetic anisotropy energy caused by defects and stress at the interfaces and the role of the biquadratic exchange coupling between interfacial Fe and Cr magnetic moments.

The magnetic properties of Fe/Cr and Fe/Cr/Fe have been studied by a variety of methods such as Brillouin light scattering (BLS), ferromagnetic resonance (FMR), neutron scattering, or the magneto-optical Kerr effect (MOKE), and a compilation of the major results can be found in several review articles. Recently, the transformation between different magnetic states in exchange-coupled structures Fe/Cr/Fe has been probed by the method of magnetization-induced second harmonic generation (MSHG). We have shown that MSHG opens a pathway to explore peculiarities of the magnetic state and its field-induced evolution in thin films, which have not been observed previously by using conventional methods, e.g., MOKE or BLS. In particular, when an in-plane magnetic field was applied close to the direction of the hard axis in an Fe/Cr/Fe structure—displaying a ferrimagnetic alignment at \( H = 0 \)—we observed an additional magnetic state transformation by means of MSHG. For the correct interpretation of the experimental results obtained by the different methods (FMR, BLS, MSHG) as well as for the practical application of Fe/Cr structures, a detailed knowledge of their magnetic properties, in particular, the domain structure at different values and orientations of the magnetic field, is needed.

In this work, we employ the MSHG approach to probe and analyze the domain structure of anisotropic Fe/Cr films. The MSHG response in the case of a multidomain state has been studied both experimentally and theoretically. With the help of MSHG the domain images of antiferromagnetic CrO₂,10 hexagonal rare-earth manganites \( RMnO₃ \),11,12 as well as magnetic garnet films13,14 have been obtained. The MSHG features in the case of a multidomain structure in contrast to a single-domain case were theoretically considered in Refs. 15–17. In particular, a complementary mechanism associated with a second harmonic generation signal arising from domain walls was proposed in Ref. 15. This particular mechanism is based on the spatial magnetization gradients in the domain wall. For the situation of magnetic stripe domains it was predicted that the angular profile of the SHG intensity comprises a diffraction pattern of SHG beams characterized by diffraction orders \( m \). The detailed theory of SHG and MSHG from magnetic multilayers including the case of a multidomain structure and calculations of the angular profiles of these phenomena based on the electric point-dipole radiation theory were recently presented in Ref. 17.

The appearance of a domain structure in systems with in-plane uniaxial magnetic anisotropy when a magnetic field...
is applied along the hard axis, as well as in systems with out-of-plane magnetic anisotropy when a magnetic field is applied perpendicular to the easy axis, as well known. The formation of this structure is associated with fluctuations of the magnetic anisotropy appearing for different reasons related to the methods of preparation, the homogeneity, uniformity, morphological structure of the film, and so on. In Ref. 20 the formation of a multidomain structure during the magnetization reversal process initiated by an in-plane magnetic field in polycrystalline Au/Co/Au structures having strong perpendicular magnetic anisotropy has been studied. The observed phenomenon was associated with the existence of nanosized uncoupled atomic terraces with different local magnetic anisotropies. Nevertheless, to the best of our knowledge in systems with biaxial magnetic anisotropy and particularly in epitaxial, monocrystalline Fe/Cr(100) films grown by molecular beam epitaxy (MBE), the existence of a nonuniform magnetic state in a wide range of magnetic field including \( H=0 \) has not yet been reported.

The domain structures in poly- and monocrystalline Fe/Cr and Cr/Fe films have been studied by different methods. In the work of Fischer et al. in Ref. 21, by means of magnetic transmission x-ray microscopy, images of in-plane magnetic domains in Cr(3 nm)/Fe(50 nm)/Cr(6 nm) films grown by thermal evaporation on Si_3N_4 substrates were obtained with a magnetic field applied in the film plane. In polycrystalline films with a weak uniaxial in-plane magnetic anisotropy a typical ripple structure with preferred domain orientations was observed. The latter were attributed to small magnetic fields being present during the evaporation process. The domain structure of thin (2 nm) Fe monocrystalline films grown by MBE on bulk Cr(100) using secondary electron microscopy with polarization analysis was studied in Ref. 22. At temperatures above \( T_N \) of bulk Cr, the ultrathin films are left in a monodomain state with the magnetization along the easy axis after application and removal of the external magnetic field. However, at temperatures lower than \( T_N \), a transition to a multidomain state takes place, with the magnetization still lying in the film plane, but the magnetization vector rotated by 90° in some places. The reason for the appearance of the multidomain state with the orthogonal direction of \( \mathbf{M} \) at \( T < T_N \) is associated with the presence of an atomic scale roughness at the interface, which can result in perpendicular coupling of Fe and Cr interfacial magnetic moments. Note that such an unusual behavior was observed in Fe films with a thickness of less than 5 nm. In Ref. 23 the domain structure of epitaxial Fe/Cr(100) thin films was studied by means of element-sensitive photoemission spectroscopy based on magnetodichroic effects in photoinduced Auger electron emission. The images of 180° domains in Cr films and Fe substrates were separately obtained with spatial resolution of 10 \( \mu \text{m} \), and regions of bilinear and biquadratic exchange coupling in a Fe/Cr/Co sandwich were identified.

In this contribution we show that in monocrystalline bi-axially anisotropic Fe/Cr films of 10–50 nm thickness a peculiar magnetic multidomain structure with equal components of the longitudinal magnetization \( M_x \) and transverse components \( M_y \) of different sign can be realized at \( H=0 \), after applying a saturating magnetic field in a narrow angle range slightly deviating from the hard axis direction. The analysis of the MSHG experimental data obtained allows us to define the change of the relative contributions of domains with different \( M_y \) components during the magnetization reversal process.

II. EXPERIMENTAL ASPECTS

The Fe films (thickness \( d=10–50 \text{ nm} \)) were grown by molecular beam epitaxy onto GaAs(001) substrates, with an Ag(150 nm)/Fe(1 nm) buffer layer being deposited prior to the Fe film growth in order to provide better epitaxy. The structure was covered by a Cr(2 nm) protective cap layer. The quality of the films has been monitored in situ by reflection high-energy electron diffraction during the growth process. After deposition the sample was removed from the chamber and fixed onto a sample holder allowing a 360° rotation around the surface normal. All measurements were performed at room temperature (\( T=294 \text{ K} \)).

The geometry of the MSHG experiment is schematically shown in Fig. 1. A magnetic field up to 3 kOe was applied parallel to the sample surface in the plane of the incident light (longitudinal geometry). The MSHG field dependencies were measured in an applied magnetic field by sweeping in the range 0⇒3 kOe⇒−3 kOe⇒3 kOe. The exciting light pulses at \( \lambda=800 \text{ nm} \) (\( E_{\text{ph}}=h\omega=1.55 \text{ eV} \)) with a duration of 150 fs were generated by a Ti:sapphire regenerative amplifier with 1 kHz repetition rate and 15 \( \mu \text{J} \) pulse power. The pulses were focused into a spot of 1 mm diameter on the sample surface. The SHG was measured in reflection at \( \lambda=400 \text{ nm} \) (\( E_{\text{ph}}=2h\omega=3.1 \text{ eV} \)). The angle of incidence was \( \theta=5° \). The polarization of the fundamental and frequency-doubled light was chosen by a proper orientation of polarizer and analyzer, allowing one to investigate the second harmonic generation (SHG) signal in four different polarization combinations (\( pp, ps, ss, sp \)). The fundamental light at \( \lambda=800 \text{ nm} \) was rejected by placing a blue filter (BG-39) into the reflected beam. The SHG signal was recorded using a photomultiplier and photon counting technique. The count-
ing time of each experimental point was 10–20 s. While the polarizer was rotated, the intensity of the excited light was kept constant (with an uncertainty of less than 5%) by means of a quarter-wave plate placed just before the polarizer. The sample was mounted on a motorized rotational stage providing azimuthal rotation around the surface normal with high precision (~0.001°).

For comparison purposes, together with the MSHG also the conventional longitudinal magneto-optical Kerr effect has been studied in the Fe/Cr films using a cw diode laser at λ = 670 nm for s polarization of the fundamental beam. The incidence angle in these measurements was kept at about 35°. A high sensitivity of the MOKE measurements was obtained by using a differential photodetector and lock-in technique.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. MSHG approach

In order to extract details of the interfacial magnetization behavior in Fe/Cr structures we have to describe the field dependencies of the MSHG response for different polarization combinations of the light. For this purpose we follow the formalism used in Ref. 9. The formalism is based on phenomenological expressions of the nonlinear response from the single surface or interface of \( C_{4v} \) symmetry.\(^{25–30} \) As shown in Ref. 9, these expressions can also be used to describe the nonlinear response of the system with several \( C_{4v} \) symmetry. In that case, the description of the system with more than one interface can be conducted in terms of effective nonlinear optical susceptibilities \( \chi^{(2)} \). The expressions for the SHG intensity at small incidence angles for \( pp \) and \( ss \) polarization combinations have the forms

\[
I_{2\omega} = A|\chi^{\text{eff}}_{\text{xxx}}|^2 M_x^2 + |\chi^{\text{eff}}_{\text{xxz}}|^2 M_x^2 + |\chi^{\text{eff}}_{\text{xyz}}|^2 M_x^2 + 2|\chi^{\text{eff}}_{\text{xxx}}| |\chi^{\text{eff}}_{\text{xxz}}| M_x \cos \theta^2 \quad \text{for } pp, \tag{1}
\]

\[
I_{2\omega} = A|\chi^{\text{eff}}_{\text{yyy}}|^2 M_x^2 \quad \text{for } ss, \tag{2}
\]

where \( \theta \) is the angle between the magnetization \( M \) and the field \( H \), \( \phi \) is the angle between the magnetization \( M \) and the hard axis \( H_1 \) (see Fig. 1). The magnetization value \( M = 1.71 \) kG for bulk Fe and the expression \( 2K / M = [0.55–2.5/\delta(ML)] \) kOe of Ref. 6 to define the anisotropy constant have been used in the calculations of \( \varepsilon_m \). The minimization of Eq. (3) gives the possibility of defining the orientation of the magnetization \( M \) in the film plane depending on the direction of the applied magnetic field \( H \). The parameters \( |\chi^{\text{eff}}|, |\alpha| \) and the phase shift \( \Delta \) are defined from the experimental values \( I_{2\omega}(H\rightarrow+0), \) \( I_{2\omega}(H\rightarrow-0), \) and \( I_{2\omega}(H_0) \) for corresponding polarization combinations \( (pp, ss) \).

It should be noted that the MSHG signal reflects the magnetization behavior only close to the interfaces within a depth of one or two atomic layers. In contrast to that, the conventional MOKE signal is mainly related to the volume magnetization. However, previous investigations of MSHG and MOKE in Fe/Cr/Fe structures showed that the volume and interfacial magnetization behave in a similar way.\(^9 \) In particular, the critical fields corresponding to jumps-like transformations of the magnetic structure are the same for both MOKE and MSHG.

B. Domain structure and estimation of relative domain volumes

In Figs. 2(a)–2(d) the field dependencies of \( I_{2\omega}(H) \) for \( pp \) and \( ss \) polarization combinations in Fe(10 nm)/Cr films at a deviation of the hard axis \( h_1 \) by \( \xi = \pm 2^\circ \) from the direction of the magnetic field \( H \) are shown. The solid lines correspond to calculations on the basis of Eqs. (1) and (2). In the range of positive fields \((H>0)\), the calculated curves correspond to a coherent rotation of the magnetization toward the nearest easy axis as the field decreases, i.e., \( \xi = -2^\circ \) and \( \xi = +2^\circ \) (see Fig. 1). The jumps of \( I_{2\omega} \) close to \( H \approx 0 \) are due to a change of sign of the component. In the range of negative fields \((H<0)\), the theoretical curves correspond to a magnetization rotation from the easy axis toward the direction of \( H \) as the field amplitude increases. For the \( pp \) polarization combination the field dependencies \( I_{2\omega}(H) \) measured at a deviation of the magnetic field from the hard axis by the same angle \( \xi = \pm 2^\circ \) display a mirror symmetry rela-
positive for insets of Figs. 2. The results of the decomposition are shown in the figure. The odd part of the loop is determined only by the parameter values as in Fig. 2. The thick arrows schematically show the magnetization state and orientation of the magnetization in domains.

In Fig. 3 for the same film the $I_{2\omega}(H)$ dependence for the $pp$ polarization combination and $\xi<0.1^\circ$ is presented. The dependence is very different from those shown in Figs. 2(a) and 2(b). In analogy to the dependence in Fig. 2(a), it starts from the monodomain state being conserved after the measurement cycle at $\xi=-2^\circ$. As the field increases, it achieves the saturation value and the magnetization aligns along the field. However, with a subsequent decrease of the field to $H=0$, the initial value of $I_{2\omega}(H)$ is not reached. This means that the magnitude of $M_s$ in this case is essentially smaller than the initial value corresponding to the monodomain state. After the field changes sign, $I_{2\omega}(H)$ approaches the saturation value at negative field $-H_s$. In the reversed field direction (from $-H_s$ to $0$) $I_{2\omega}(H)$ changes, corresponding to a coherent rotation of the magnetization in the monodomain state. In the area of $(0, +H_s)$ also a coherent rotation of the magnetization takes place. Note that, when the field changes sign in the $(H_s, H)$ area, the sign of the $M_s$ component does not change. The effective decrease of the $M_s$ component as the field decreases from positive values evidences the presence of a multidomain state in the film involving opposite signs of magnetization states and orientations of the magnetization in domains.

In Figs. 4(a)–4(f) the SHG field dependencies in an Fe(49 nm)/Cr(2 nm) system at different azimuthal angles of the magnetic field close to the hard axis are shown. The solid lines show the results of calculations for the case of coherent rotation of the magnetization to different easy axes. Thus, if a coherent rotation of M toward one of the easy axes in the film takes place, we expect the field dependence to agree with the calculated curves. However, if the system develops a multidomain structure, the experimental dependencies will fall inside the area formed by the calculated curves. In fact, a coherent rotation of the magnetization is observed in Figs. 4(a) and 4(b), while the existence of a multidomain structure in a wide range of field values (including zero magnetic field) is clearly seen in Figs. 4(c)–4(f). Note that the experimental dependence presented in Fig. 4(e) is nearly symmetrical—alogously to that observed in the Fe(10 nm)
...ing on the calculated curves decreases from positive as well as from negative fields. Bas-
ning on the calculated curves $I_{2\omega}(H)$ shown in Fig. 4 by solid lines, one may calculate the field dependencies of the relative volumes of domains $W_1(H)$ and $W_2(H)$ with opposite ori-
tations of the $M_y$ component. In the $pp$ polarization combina-
tion the dependence $I_{2\omega}(H)$ in the multidomain state can be written in the form

$$I_{2\omega}^{\text{calc}}(H) = W_1(H)I_{2\omega}^{\text{calc}}(\pm M_y(H)) + W_2(H)I_{2\omega}^{\text{calc}}(-M_y(H)),$$

$$W_1(H) + W_2(H) = 1,$$

where $I_{2\omega}^{\text{calc}}(\pm M_y(H))$ are the values of the SHG intensity calculated on the basis of Eq. (1). The solution of the system of Eqs. (4) and (5) allows us to calculate $W_1(H)$ and $W_2(H)$. As we can see in Fig. 4, the experimentally observed magni-
tude of $I_{2\omega}$ for positive and negative saturating field is slightly different. This small difference, which is absent in the theoretical curves, can be attributed to a small deflection of magnetic field from the incidence plane that results in small contributions of $M_z$ to $I_{2\omega}$. This contribution has been taken into account in the calculations and the results are shown in Fig. 5.

At a deviation of the magnetic field $H$ from the hard axis ($\xi = -0.9^\circ$) the $W_1(H)$ dependence represents a hysteresis loop of rectangular shape. In fields $|H| > 0.12$ kOe in the exposed area the monodomain states characterized by $M_y$ components of opposite sign are realized. At $\xi = -0.1^\circ$ the rectangular hysteresis is absent and a monodomain state is not established. Instead a quite complicated transformation of the domain structure accompanied by jumps of $W_1$ is observed. In particular, in the region $H = 0.3$ kOe [Fig. 5(b)] a sharp decrease of $W_1$ in increasing field is observed. In a sweep of the magnetic field in the reversed direction, an increase of $W_1$ is observed in the same field regime. At $\xi = 0^\circ$ the jumps are absent and the $W_1$ average values in the positive and negative field areas differ somewhat. This indicates that the volumes of different domains are still changing with the field even in this case. At $\xi = +0.1^\circ$ the rectangular...
loop starts to appear; however, in the negative field range the monodomain state is not yet achieved. Such an interpretation assumes that the lateral domain dimensions with positive and negative $M_z$ components may be comparable with the probed area. As follows from the theoretical arguments discussed in Ref. 17, in the case of a stripe domain structure with a period comparable to or exceeding the light wavelength, diffraction peaks should be observable in the angular distribution of the SHG signal. In addition, in the specular reflection geometry, the nonlinear susceptibilities odd in the magnetic field should not appear. The fact that in our measurements sizable linear-in-$M$ susceptibilities are observed means that diffraction effects do not play a significant role. This also suggests that the domain dimensions are much larger than the wavelength of the light. The magnetic contrast of the SHG signal from different domain areas is 60% at $H\sim 0.1$ kOe. This relatively large value should enable a visualization of these domain structures by means of a charge-coupled device camera.

In Fig. 6 the field dependencies of the longitudinal magneto-optical Kerr effect at different $\xi$ are shown. It should be noted that the polarization plane rotation observed in the experiment at a magnetic field applied close to the hard axis is defined not only by the longitudinal magnetization component $M_z$, but also by the transverse $M_x$ component through the terms quadratic in magnetization ($\sim M_x M_y$ type) in the tensor of the dielectric susceptibility. This leads to the magnetic field asymmetry of the measured field dependencies, and the observation that they not exactly reflect the behavior of the $M_x(H)$ component. The separation of contributions linear and quadratic in magnetic field can be performed with the help of a loop symmetrization procedure. The dependencies shown in Fig. 6 are the result of such a procedure and reflect the behavior of the longitudinal $M_z$ component. As follows from the Fig. 6, the MOKE weakly depends on $\xi$ at small $\xi$ values. That is, because the longitudinal MOKE is sensitive to the variation of the $M_z$ component and not sensitive to the formation of a domain structure with different orientations of $M_z$ components in the domains. After removing the magnetic field the magnetization in the domains is oriented along the easy axes ($e_1$) and ($e_2$), with the $M_z$ components being equal, but $M_x$ differing in sign at $\xi=0$. Therefore, the MOKE field dependence corresponding to a multidomain state at $H=0$ [Fig. 6(b)] is similar to that realized for the monodomain state [Figs. 6(a) and 6(c)].

C. Magnetic anisotropy fluctuations

It is known that for large values of the demagnetizing field the magnetization in Fe/Cr and Fe/Cr/Fe structures lies in the film plane. Such a situation is realized for film thicknesses larger than $d=8-10$ ML, if the perpendicular interfacial anisotropy energy is smaller than the volume one, i.e., its magnitude is insufficient to turn the magnetization out of plane. In this case the in-plane magnetization orientation at $H=0$ is defined by the in-plane magnetic anisotropy energy and its angular dependence. The Fe films grown on GaAs(100) substrates with Ag as a buffer layer display a fourfold (biaxial) in-plane magnetic anisotropy with the directions of two easy ($e_1$ and $e_2$) and two hard ($h_1$ and $h_2$) axes along [100]- and [110]-type crystallographic directions, respectively.

In principle, in the absence of an external magnetic field the magnetization $M$ might be directed along two mutually perpendicular directions $e_1$ or $e_2$ in the film plane, thereby forming a domain structure with four possible orientations of the magnetization vector in the domains. However, in an idealized perfect film, i.e., infinite extension in the film plane, such a structure is not favorable, because of the additional energy needed to nucleate domain walls. It should be noted, however, that in contrast to the bulk samples where the domain formation results in energy gain due to the compensation of demagnetizing fields, there is no energy gain due to domain formation in thin films, since the demagnetizing fields in the film plane are equal to zero (this holds, of course, only if the influence of the boundary can be neglected—otherwise there will be closure domains formed). Therefore, it is believed that after the external magnetic field is reduced to zero a monodomain state develops, with the direction of the magnetization pointing along one of the easy axes, namely, the one that is closer to the direction of magnetic field applied earlier.

However, if the magnetic field is applied exactly along a hard axis ($H\parallel h$), there are two equivalent easy axes ($e_1$ and $e_2$) oriented at angles of 45° to the direction of the magnetic field ($H$) and the hard axis ($h$). This results in a situation...
known as “Buridan’s donkey”: the magnetization has no way
to decide which direction is more favorable to rotate into as
the magnetic field slowly decreases from the saturation value
\(H=H_s\) to zero. Therefore, the question arises: In what man-
ner will the magnetization reversal take place in this particu-
lar case?

Two extreme variants of the magnetization reversal pro-
cess can be considered, related to either an ideal or a more
realistic picture of the thin film. (i) Even infinitely small
deflections of the magnetic field from the hard axis \(h\) result
in a symmetry breaking and thus a nonequivalency of the
easy axes. The magnetization will then coherently rotate to-
ward the nearest easy axis without a domain structure forma-
tion as the field decreases. This seems to be the energetically
most favorable variant, because the energy losses associated
with the formation of domain walls are absent in this case. In
practice, the realization of the situation \(H||h\) is extremely
difficult to achieve, because there may be always an arbi-
trarily small field component perpendicular to \(h\), leading to a
symmetry breaking. (ii) The uncertainty of the magnetization
rotation might be removed by assuming that the easy axes \(e_1\)
and \(e_2\) are varying across the film surface, i.e., locally
the easy axis fluctuates around the average direction of \(e_1\) or \(e_2\).
This fluctuation of the easy axis direction, which more
closely describes the situation in a realistic film, in turn, may
be caused by fluctuations of the magnetic anisotropy energy
\(\delta e_m(r)\) due to defects or strains, or to a locally varying ex-
term coupling between Fe and Cr magnetic moments at
the Fe/Cr interface. As a result, in different film areas the preferable
direction of the magnetization rotation will corre-
spond to differently oriented easy axes \(e_1\) and \(e_2\). This situa-
tion is a precondition for the formation of domain structure
at \(H=0\) after magnetizing along the hard axis.

In strong magnetic fields the Zeeman energy essentially
overcomes the magnetic anisotropy energy and the magneti-
zation aligns with the field, i.e., \(M||H\). As the magnetic field
decreases, the influence of the magnetic anisotropy becomes
stronger and the magnetization tends to rotate toward the
nearest easy axis. If in different film areas the directions of
the easy axes are fluctuating, the formation of nonuniform
distribution of the magnetizations in domains occurs. The
exchange coupling acts against this process, because it favors
parallel alignment of the magnetization in different areas.
However, the energy losses required for the formation of a
nonuniform structure are small, at least in relatively large
fields and can be compensated by a gain in magnetic aniso-
tropy energy. It seems that this situation is somewhat anal-
gous to the one considered in the “random anisotropy” model,\(^{41}\) used for the description of magnetic properties of
amorphous alloys or polycrystalline magnets. The experi-
ment shows that this nonuniform magnetization distribution
is kept down to \(H=0\), where a multidomain state with 90°
domains is realized.

The magnetic energy with lateral fluctuations taken into
account can then be written in the form

\[
e_m(r) = -HM_s \cos \psi + \frac{K_1}{4} \cos^2(2\varphi) + \delta e_m(r). \tag{6}
\]

The fluctuating part \(\delta e_m(r)\) that provides the local non-
equivalency of the easy axes can be written as follows:

\[
\delta e_m(r) = b(r)(u \cdot m)^2, \tag{7}
\]

where the coordinate function \(b(r)\) might have a different
sign in different areas of the film; \(m\) is the unit vector along
the magnetization direction; \(u\) is the unit vector along one of
the easy axes, e.g., \(e_1\). The addition of the fluctuating
uniaxial anisotropy to \(e_m\) results in a situation in which one
of the easy axes locally becomes an intermediate axis, while
the second easy axis is the only remaining one. For example,
assuming that \(b < |K_1|\), at \(b < 0\) (\(b > 0\)) the easy axis will be
\(e_1\) (\(e_2\)) and the middle axis will be \(e_2\) (\(e_1\)). If the magnetic
field is applied along the hard axis, \(H||h_1\), then as the mag-
netic field decreases from \(H_s\) to zero in the areas where \(b < 0\),
the magnetization will tend to rotate toward \(e_1\), while in
the \(b > 0\) areas to the \(e_2\) axis. It is important to note that, for
\(b < 0\) (\(b > 0\)), the magnetization will rotate toward \(e_1\) (\(e_2\))
not only at \(H||h_1\), but also in the case when the direction of
\(h_1\) slightly deviates from the field direction, so that the
middle axis \(e_2\) (\(e_1\)) lies closer to the field direction than to
the easy axis. The maximum value of this deflection (the
critical azimuth of the hard axis \(\xi_c\)) depends on the sign and
value of the parameter \(d = 2b/K_1\). The calculated dependen-
cies of the critical azimuthal angle \(\xi_c\) on \(d\) for positive and
negative values of \(d\) are presented in Fig. 7. In the calcu-
lations it was assumed that \(b(r) = \text{const}\), i.e., we assume that in
the area of each domain the function \(b(r)\) might be replaced
by its average (effective) value.

In particular, for \(d = +0.2\) (\(b > 0\)) as the field decreases
from \(+H_s\) to zero, the magnetization will rotate to \(e_1\) until the
azimuth of the magnetic field decreases down to \(-2.9^\circ\)
dashed red line approaching from the top the line with \(d > 0\).
At smaller azimuthal angles of the magnetic field the rotation
will happen in the other direction (green line). At \(d = -0.2\)
\((b < 0)\) the magnetization will rotate to \(e_2\) until the
azimuth of the magnetic field reaches \(2.9^\circ\) (green line ap-
proaching from the bottom line \(d < 0\)). At larger azimuth
values the magnetization rotates to \(e_2\) (dashed red line).
As seen in Fig. 7, for \(|d| \neq 0\) there is always a hard axis azi-
muthal region, where for positive and negative values of \(d\) a
magnetization rotation happens in opposite directions as the
field decreases. For \(|d| = 0.2\) that region is \(-2.9^\circ < \xi < 2.9^\circ\).
At \(\xi > 2.9^\circ\) and \(\xi < -2.9^\circ\) the rotation in all film areas will
happen in one direction (toward $\mathbf{e}_0$ or $\mathbf{e}_1$, respectively). As follows from the experiment (see Fig. 5) for the film with a thickness of 49 nm, the critical angle has the value $\xi_c \approx 0.3^\circ$, since for $|\xi| > 0.3^\circ$ the field dependencies $I_{2\alpha}(H)$ are well described by a coherent magnetization rotation for both positive and negative fields. For $|\xi| < 0.3^\circ$ the multidomain magnetic structure is observed in the film. That allows one to estimate the average value of the parameter $d$, which is $d = 0.01$. From that it follows that even small $\sim 1\%$ fluctuations of the magnetic anisotropy energy might result in essential changes of the magnetic structure in biaxially anisotropic films. These changes are manifested at a magnetic field direction near the hard axis when there are two equivalent magnetic states differing only in the sign of the $M_z$ component.

The fluctuations of the magnetic anisotropy energy might arise for different reasons. (i) It is known that at the Fe/Ag or Fe/Cr interfaces strained regions may form, because of the difference in unit cell parameters or due to interfacial alloying of Cr and Fe ions.\textsuperscript{6} The fluctuations of strain values in the film might result in lateral fluctuations of the magnetic anisotropy energy $\delta \varepsilon_m(r)$ described by Eq. (7). Analogous fluctuations of $\delta \varepsilon_m(r)$ may arise due to the existence of atomic scale steps and terraces, islands, and pits at the Ag/Fe and Fe/Cr interfaces oriented along fourfold axes.\textsuperscript{42} (ii) The fluctuations $\delta \varepsilon_m(r)$ may arise as a consequence of the antiferromagnetic exchange interaction between the interfacial magnetizations of Fe and Cr. It is known that in thin Cr films at $T < T_N$ there is a layer-by-layer antiferromagnetic ordering of the magnetic moments.\textsuperscript{5} In addition, in the Fe/Cr interface areas with enhanced concentration of atomic scale roughnesses biquadratic coupling might take place even in the case of one interface.\textsuperscript{22} If the antiferromagnetic state of the film is monodomain, then in the areas with a large amount of roughness the magnetization direction, which is orthogonal to that occurring in the areas with lower roughness, is more profitable. If in the Cr film the antiferromagnetic multidomain structure with orientation of the antiferromagnetic vector $\mathbf{L}$ along directions of [001] type is realized, then, even in the case of a uniform roughness distribution, the preferable magnetization direction is [010] in the areas with $\mathbf{L}||[001]$. A situation when the changes happen in magnetic structures of both Fe and Cr upon magnetizing is also possible. Clearly, separation of the mechanisms of (i) and (ii) should be possible by means of temperature-dependent studies. At a temperature remarkably higher than $T_N$ ($T > 1.8T_N$; Ref. 43) where Cr is in the paramagnetic state, the mechanism (ii) should disappear. The contribution resulting from the mechanism (i) may change with temperature, but should survive above $T_N$.

IV. CONCLUSIONS

Thus, the investigations of monocristalline Fe/Cr films with biaxial magnetic anisotropy show that, when the magnetic field is close to the hard axis direction, the remagnetization process is accompanied by multidomain structure formation. The multidomain structure appears not only in the narrow field region corresponding to the domain wall movement occurring during remagnetization, but in a much wider field range including also $H = 0$. The domains formed have equal magnetization components ($M_z$) along the magnetic field, but magnetization components ($M_x$) perpendicular to $H$ of opposite signs. It is shown that in the situation with equal domain volumes having opposite directions of $M_x$ components and, as a result, with zero sum, a transverse magnetization can be realized at orientations of the magnetic field close to the hard axis. The reason for the multidomain structure formation is proposed to lie in lateral fluctuations of the magnetic anisotropy energy arising in the film because of defects or strains or due to the presence of atomic scale roughness at the interface. The existence of multidomain structure can manifest itself in magnetization dynamics experiments and should be taken into account for the interpretation of experimental results (BLS, FMR).

ACKNOWLEDGMENTS

The authors would like to thank R. Schreiber for the sample preparation. For A.A.R. and B.B.K. this work was supported by RFFI Project No. N 05-02-16451-a.