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## Magneto-optical indicator film study of the hybrid exchange spring formation and evolution processes

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## Abstract

The elementary events of the remagnetization processes in nanocomposite magnetic bilayers were investigated using iron-garnet indicator films with in-plane anisotropy. We have observed hybrid domain walls consisting of both ferromagnetic and antiferromagnetic sections perpendicular to the interface. The external magnetic field shifts only the ferromagnetic part of the domain walls. This leads to the formation of a hybrid exchange spin spring parallel to the interface. The processes of spring nucleation and untwisting occur at different locations. With the field oriented antiparallel to the macroscopic unidirectional anisotropy, remagnetization of the soft ferromagnet layer in the hard/soft nanocomposite starts by the formation of an exchange spring consisting of micrometer-scale sub-domains with opposite direction spin twisting. A rotating magnetic field (smaller than some critical value) creates firstly a single-chiral spin spiral; this spiral then loses stability, incoherently untwists and gradually inverts its chirality with increasing field rotation. Untwisting of the hybrid exchange spring at higher fields leads to the creation of unusual hybrid non-180° domain walls. The initial (ground) state of the bilayer with such noncollinear magnetized domains is not restored after stopping the field rotation and returning it to zero. The revealed phenomena are attributed to the influence of the dispersion in the unidirectional anisotropy induced by magnetization frustration in the interface and bilayer crystal lattice defects.

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The science and technology of heterophase thin film magnetic materials are currently attracting worldwide attention because of the unique physical properties and important practical applications of these materials [1,2]. Spin coupling across the interface between thin layers possessing different magnetic order leads to a broken magnetic symmetry. Ferromagnets of this type are characterized by having a unidirectional anisotropy: wherein opposite spin directions are energetically inequivalent. The magnetization reversal of such a nanocomposite ferromagnetic bilayer is determined by the nucleation and evolution of hybrid spin spirals (exchange springs) that differ dramatically from topologically stable domain walls (DWs) in bulk conventional ferromagnets.

For bulk materials, the sample size is greater than the domain dimensions and much greater than the domain wall width. Large regions of uniform spin orientation in conventional bulk ferromagnets are separated by narrow topologically stable spin spirals–domain bound-aries [3]. In conventional single-phase ferromagnets, the remagnetization process proceeds primarily by the motion of DWs as quasi-particles of classical nature.

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They were nucleated and formed in a very small part of the sample. In contrast, the thickness of each ferromagnetic layer in a nanocomposite bilayer is usually the same order of magnitude (or less) as the domain wall width. The remagnetization of the nanocomposite ferromagnet in a heterophase bilayer proceeds primarily through the formation process of hybrid spin spirals.

Experimental observation of the nucleation and development process of DW-type spin spirals is an extremely important but difficult problem. Even in uniform bulk ferromagnets, until now, there has not been a direct experimental study of the formation process. In this article we present the results of an investigation of the formation and evolution of hybrid spin spirals in two types of heterophase nanocomposite bilayers: (1) in exchange coupled ferromagnet/antiferromagnet (FM/AF) thin films and (2) in magnetically soft and hard ferromagnets (sF/hF). It was possible to do this using the advanced magneto-optical indicator film (MOIF) technique [4–6]. This technique is based on the use of the Faraday rotation of linearly polarized light in an indicator film with in-plane anisotropy, placed on the sample. Stray fields emanating from its edges, DWs, and other magnetic inhomogeneities deflect an indicator film magnetization out of its plane. As a result, domain structure of the sample is imaged via Faraday effect in the indicator film.

We studied a 160 Å thick Ni<sub>81</sub>Fe<sub>19</sub> permalloy film that was grown on an epitaxial Fe<sub>50</sub>Mn<sub>50</sub>(300 Å)/Cu(300 Å)/ Si structure [7]. It was cooled in zero magnetic field after FM film demagnetization in a decreasing AC magnetic field at T = 400 K, a temperature which is higher than the MnFe blocking temperature and lower than the Curie temperature of the permalloy. The magnetization reversal of the FM in the magnetic field  $\mu_0 H = 20$  mT was characterized by two separate hysteresis loops, each shifted by the same amount but in opposite direction along the *H*-axis (Fig. 1a). This means that the FM film contains regions with oppositely oriented axes of unidirectional anisotropy. Using the MOIF technique, we visualized for the first time the domain wall in the AF layer coupled to an FM DW, and studied the evolution of such a hybrid domain wall (HDW) in a magnetic field.

Fig. 2 shows the domain structure for the ACdemagnetized AF/FM bilayer as the external field is cycled. One can clearly see the  $180^{\circ}$  domains in the initial ground state (Fig. 2a). They are revealed due to the well-defined black–white contrast on their boundaries as well as on the sample edges. The component of the magnetization, M, perpendicular to the wall (as well as to the sample surfaces), gives rise to magneto-static poles, the type of which depends on the relative orientation of the magnetic moments in the domains. In the described case, a head-to-head M position leads to the black tint in the MOIF image of the wall. The light tint appears in case of a tail-to-tail orientation of the M in the region adjacent to the DW.

The FM layer magnetization changes with the Happlication not only because of displacement of the initial ground state DWs, but also due to nucleation of new walls (and their subsequent motion) in domains originally magnetized in the direction opposite to the field. The domain structure corresponding to the halfloop to the right of the origin in Fig. 1a is presented in Fig. 2b-e. The corresponding pictures for the half-loop on the left side of Fig. 1a are shown in Fig. 2f-j. Note that the approach to saturation under positive fields (Fig. 2b and c) and remagnetization back into the demagnetized state (Fig. 2c-e) occur in a domain depicted in the central part of the picture, while under negative fields (Fig. 2f-j) the remagnetization process proceeded only in the adjacent domains. Fig. 2 clearly evidences the asymmetry of the nucleation centers activity [8,9] during field cycling of this sample.

Note also in Fig. 2b that only one of the two adjacent walls separating domains in the FM in the demagnetized state moves upon application of positive fields. The DW configuration is drastically different during application of negative fields (Fig. 2g). The most unexpected result



Fig. 1. Minor hysteresis loops of Py(160 Å)/FeMn(300 Å) at 300 K (a) after demagnetizing at 400 K, and zero field cooling and (b) after field cooling in a 20 mT field.



Fig. 2. MOIF images of domain structure taken during unidirectional-axis remagnetization of the zero field cooling Py/FeMn bilayer. (a)–(e) correspond to the right loop and (a), (f)–(j) to the left hysteresis loop at Fig. 1a. (a)  $\mu_0 H = 0$ , (b) +1.8, (c) +6.0, (d) +0.6, (e) +0.6 after 20 s, (f) -1.15, (g) -1.2, (h) -6.0, (I) -0.4, (j) -0.35 mT, The top band perpendicular to the unidirectional axis is the edge of the bilayer.

was the appearance of DW contrast even when the FM was magnetized to saturation (Fig. 2c and h), and this contrast was located in the same place where the DW had been located in the ground state (Fig. 2a). It is obvious that this contrast is associated with the stationary HDW section, which is situated in the AF layer. In this sample cooled in a constant saturating field (characterized by one unidirectional anisotropy,

Fig. 1b), such faint DWs were not observed at saturation.

Thus, the presented results clearly show that the MOIF technique permits one to investigate the domain structure in an AF thin film exchange coupled with an FM layer. AF domains arise under the influence of 180° domains in the FM layer during cooling of the bilayer at temperatures below the blocking temperature point. The FM layer also plays the role of a sensitive sensor in which stray field along the intersection of the interface and the AF DW is created. The ferrimagnet indicator film can visualize magneto-optically this ultra-weak stray field and reveal the evolution of the HDW under the application of external magnetic fields. Stationary DWs in an AF stabilize the domain structure of an FM layer near H = 0. They become visible due to specific black and white contrast on grav background when the movable section of the HDW in the FM moves away from them. A parallel to the interface exchange spin spring forms in this place connecting the shifted FM and the stationary AF sections of the HDW.

The bilayer domain structure and its evolution in the magnetic field are shown schematically in Fig. 3. In the ground state, the crystal contains a threading HDW (those penetrating the layers and lying normal to the magneto-coherent phase interface) that consist of FM and AF sections (Fig. 3a). They are formed near the blocking temperature (during the cooling process of the bilayer) when the magnetization vector directions in FM



Fig. 3. Schematic diagram of the spin structures in the FM/AFM bilayer of Fig. 1 during remagnetization.

domains have determined the local orientation of the antiferromagnetism vectors.

The external magnetic field exerts pressure only on the FM part of the HDW. Consequently, only the FM section moves while the AF section of the HDW remains stationary as the field is applied. However, the rotating spins in the ferromagnetic section of the HDW twist the AF spins near the intersection of the interface and the HDW. This latter spin twisting leads to the formation and growth of the novel HDW segment that is located in the AF/FM interface perpendicular to the stationary AF and shifting FM sections of the HDW. This remagnetized interface area is a specific exchange spin spring, consisting of FM and AF spins (Fig. 3b and c). Such a "partial" DW was considered in Ref. [10]. The intersection of this partial DW with the DW in the ferromagnet is a high-energy boundary line which possesses a unique spin configuration determined also by the "coherent" spin configuration between the unchanged AF/FM interface on the other side of this line. This boundary line (node) plays a key role in determining the resistance to motion of the FM domain wall. In order for the FM DW to move, a hybrid exchange spring must be grown or eliminated on one side of this node depending on the direction of the wall motion. During growth of the exchange spring (i.e. as H is increased) the ground state is firstly overcome in regions with low values of the AF anisotropy  $K_a$  and exchange  $A_a$  energies:  $E \approx \sqrt{A_a K_a}$ . When the magnetic field decreases, its pressure on the FM spins weakens so that at some critical field the amount of energy stored in the spin spring becomes enough to drive its untwisting. This process starts in the regions where  $K_a$  and  $A_a$  are high (but not as low as it was during the spring nucleation). The heterogeneous FM/AF exchange spring subsequently begins leaving the AF and gradually transforming into a pseudo-monophase FM spin spring (a quasi-180° DW in the FM). It leads to the nucleation and growth of domains in the FM layer and the sample's remagnetization back into the ground state (Fig. 3d).

In a soft ferromagnet/hard ferromagnet bilayer, the spins in the soft film start gradual rotation at reverse external magnetic fields larger than some critical value. The angle of rotation increases with increasing distance from the interface, resulting in a spin spiral similar to that in a conventional Bloch DW. At the beginning of the remagnetization process, the exchange spring is localized mainly in the soft ferromagnet of sF/hF bilayer. But unlike in the AF/FM bilayer, its motion can lead to domain formation in the second constituent of the composite. We have continued the MOIF investigation [11-13] of sF/hF nanocomposite remagnetization to study features of the hybrid spin spiral formation and motion in exchange spring magnets. In order to examine such a system, we applied the MOIF technique to an Fe(500A)/Sm<sub>2</sub>Co<sub>7</sub> (350 A) film prepared



Fig. 4. Hysteresis loop of SmCo/Fe bilayer.



Fig. 5. MO images of the SmCo/Fe bilayer region near the 300 mM hole at different reverse fields. The field is applied at angle  $\alpha = 0^{\circ}$  (a-c) and  $10^{\circ}$  (d-f).

by DC magnetron sputtering onto a Cr(200A)-buffered single-crystalline MgO(110) substrate (see Refs. [11,14] for details). In this material, the epitaxial growth of SmCo gives rise to an in-plane uniaxial magnetic anisotropy and a high coercivity. The hysteresis loop of the sample (Fig. 4) is typical for exchange spring magnets.

In order to analyze the magnetization processes of the exchange spring sample, a 300 µm hole was bored through the magnetic layers and leakage fields around the hole were monitored. The location and intensity of the bright and dark areas around the hole (Fig. 5) are determined by the direction and magnitude of the leakage field and provide a means for estimating the orientation and value of the average magnetization M[13]. A set of MOIF images in Fig. 5 shows the remagnetization process of the Fe layer when the reverse field H is either aligned with the unidirectional anisotropy axis (a–c) or deviates from it  $10^{\circ}$  in the clockwise (d-f) direction. If the applied field is perfectly aligned with the easy axis (Fig. 5a-c) the average magnetization direction remains collinear with the H, but there is a dramatic change in MO contrast during

the reversal. It decreases, practically disappears and reappears with a new sign as the reversed H increases in magnitude.

Unlike theoretical predictions, in this case, as in the case of (100)-oriented MgO [12,13], the magnetization reversal process of a uniaxial sF/hF bilayer starts and proceeds by the formation and evolution of a twodimensional ES. This is a natural consequence of incoherent spin rotation with opposite chiralities in adjacent local areas upon increasing the field. The smallangle dispersion in the sF anisotropy direction caused by weak random deviations of the easy axis in the grains of the bilayer system from the mean direction has a crucial effect on the thin structure of the exchange spring. A single-chiral spin spiral can be formed when the deviation of the remagnetizing field from the direction of the macroscopical unidirectional anisotropy exceeds the maximal easy axis dispersion (Fig. 5d-f) or in a rotating field, as was revealed [13] in Fe/SmCo bilayer with an in-plane four-fold magnetic anisotropy grown on MgO (100) substrate.

MOIF studies of the exchange-spring behavior in SmCo/Fe films with uniaxial in-plane anisotropy in a rotating magnetic field uncovered a new unusual phenomenon. Fig. 6 shows the response of the Fe layer when the applied field is fixed in magnitude  $(\mu_0 H = 0.06 \text{ T})$  but is rotated by angle  $\alpha$  in the film plane. From the easy magnetization direction (Fig. 6a), when  $\alpha$  increases, the average magnetization smoothly rotates with the field with some phase delay in the same way as in the sample with four-fold symmetry [13]. This is easily revealed by the rotation of the symmetry of the hole magneto-optical contrast (Fig. 6b and c). After the field reaches a critical value of the angle  $\alpha$ , the local magnetization in some places begin to rotate in the opposite direction while the field direction continues to change in the original direction. This leads to nucleation and growth of macroscopical domains with noncollinear magnetizations that are bounded by non-180° walls (Figs. 6b-e). The subsequent rotation of the total magnetization, after completion of this stage of DW motion and annihilation, is again in synchronization with the field. In this final stage of magnetization reversal, M takes the phase lead over H up to the point where they both coincide with the unidirectional anisotropy axis and the MO contrast in the sample again becomes homogeneous (Fig. 6f). The single domain ground state is fully restored at this point. That there is hysteresis in the magnetization rotation is shown by the data in Fig. 6g.

Fig. 6 shows DWs, which are formed because of untwisting of the exchange spring in different locations and subsequent adjacent changes in chirality. In a rotating field these specific domain walls shift, annihilate and leave the sample, leading it again in the singledomain state. In this case, the remagnetization is limited



Fig. 6. MO images of the SmCo/Fe bylayer region near the hole during an in-plane field rotation for  $\mu_0 H = 60$  mT. The white and black arrows indicate the directions of H and M, respectively. The graph shows the rotation angle  $\varphi$  of M vs. the magnetic field rotation angle ( $\alpha$ ) for different field amplitudes measured from the MOIF images.

by the motion of the line singularity connected to the intersection of the DW and the exchange spring. However, the ground state does not restore after interrupting the field rotation and decreasing it to zero if noncollinear DWs have arisen in the specimen (Fig. 7). Upon decreasing the field there is no visible domain wall motion, only a weakening of its MO contrast is observed. At small values of the magnetic field a clear contrast of micro-scale domains elongated along the uniaxial anisotropy direction arises (Fig. 7b–d). This finding provides an evidence that the exchange spring has penetrated into the hard ferromagnet and led to the formation of micro-domains. These micro-domains



Fig. 7. MOIF images of the SmCo/Fe bilayer region near the hole during decreasing field  $\mu_0 H = 90 \text{ mT}$  after interrupting its rotation.

appear as magnetization frustrations on the interface and cause noncollinear spin orientations in exchangecoupled hard and soft ferromagnet layers [15,16].

In summary, using the MOIF technique, we have revealed the new features of spin spiral nucleation and motion in soft ferromagnet films exchange coupled either with a hard ferromagnet or antiferromagnet layer that provide insights into the microscopical magnetization reversal mechanisms of these nanocomposite materials.

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