Atomic-Level Control of the Domain Wall Velocity in Ultrathin Magnets by Tuning of Exchange Interactions

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We demonstrate that the propagation velocity of field driven magnetic domain walls in ultrathin Au/Co/Au films with perpendicular anisotropy on vicinal substrates is anisotropic and strongly depends on the step density of the substrate. The velocity of walls oriented perpendicular to the steps drastically increases with increasing local step density while being unchanged or only weakly decreased for the walls oriented parallel to the steps. We develop an analytical model revealing the step-modified exchange interactions as the main driving force for this anisotropic behavior. The enhancement of the domain wall velocity at low magnetic fields far below the Walker instability threshold makes this phenomenon interesting for magnetic nanodevices.

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One of the obstacles impeding the development of devices based on field- or current-induced motion of magnetic domain walls (DWs) [1] is the open problem of tuning of wall damping and speed [2]. To overcome this difficulty, several promising concepts have been discussed in the literature. The main effort has been concentrated on the remarkable reduction in the velocity of domain walls above the Walker threshold. It has been proposed that the velocity breakdown can be suppressed by introducing a certain degree of roughness [3] and by using a ferro- [4] or antiferromagnetic underlayer [5] inducing additional magnetic anisotropy. As a way of reducing the wall velocity to a nearly full stop, an introduction of elastic linear defects has been recently proposed [6].

In this Letter, we demonstrate experimentally and theoretically that DW motion in ferromagnetic films with perpendicular anisotropy grown on a stepped substrate can be tuned by modifying the underlying step density of the supporting substrate. We were able to produce ultrathin Au/Co/Au films with different step density but identical magnetic anisotropy. The dynamics of magnetic domain walls in such films appears to be spatially anisotropic and strongly dependent on the step density. Our theoretical analysis reveals the exchange interaction as a driving force for the observed highly anisotropic dynamics of magnetic domain walls. The proposed analytical model, however, is universal and permits one to take the effect of magnetic anisotropy into account. In the present investigation an increase of domain wall velocity in very weak fields (far below the Walker threshold) by 2 orders of magnitude has been reached. Our theoretical analysis predicts even larger effects for other film thicknesses and step densities.

In order to obtain surfaces with different step distributions, a Si(111) substrate, known for its preparation flexibility [7], has been chosen. The samples were prepared under ultrahigh vacuum conditions with a base pressure of 10^{-8} Pa utilizing direct current heating up to 1250 °C. We used three types of Si(111) substrates: nominally flat with large step-free areas, vicinal with a 2° miscut towards the $[-1\ -12]$ direction giving single- and some triple-layer-height steps [8], and vicinal with a 2° miscut towards the $[11\ -2]$ direction exhibiting a periodic array of step bunches [7]. Figure 1 shows the typical stepped morphologies after substrate preparation. The steps are comprised of a sequence of atomically flat 7×7 reconstructed terraces and single steps [Fig. 1(a)] or step bunches of 8–11 single steps for a total height of about 3 nm [Fig. 1(b)]. The mean terrace widths are 9 and 80 nm for single steps and step bunches arrays, respectively.

The following layers have been deposited on the three substrates: (i) a buffer layer of 4 monolayer (ML) Cu at $T = 100$ °C—subsequent layers have been deposited at

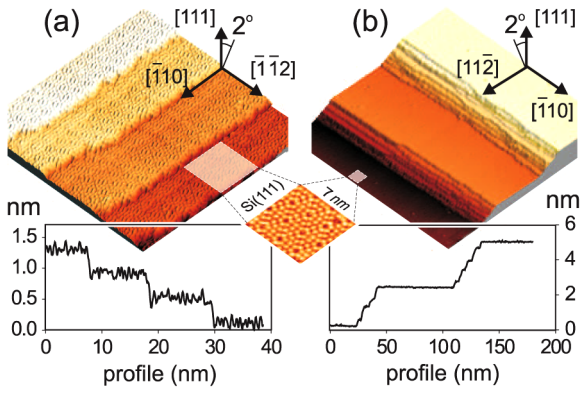

FIG. 1 (color online). *In situ* STM topographs: (a) 40×40 nm^2 image of the surface with single steps and (b) 180×180 nm^2 image of the surface with step bunches. Insets show crystallographic orientations and the cross sections of steps.

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room temperature; (ii) a 30 ML thick Au(111) underlayer; (iii) a 3 ML thick Co layer; and (iv) a 30 ML thick Au protective layer enabling an *ex situ* study. For the Au/Co/Au(111) system, it has been demonstrated that Co films have an hcp Co(0001) morphology [9]. We checked with STM that each successive fill adheres to the step density of the Si substrate. In all cases the step-edge orientation was parallel to the $[-110]$ direction of the Si.

Magnetic properties and magnetic anisotropy were studied *ex situ* at room temperature using a magneto-optical Kerr effect (MOKE) based magnetometer [10]. Magnetization reversal has been determined using the Kerr rotation in the polar (P-MOKE) configuration where the angle of incidence of the laser light is close to the sample normal and the external magnetic field H perpendicular to the sample surface. The rectangular shape of the P-MOKE hysteresis loops observed for the 3 ML Co film grown on the different substrates indicates a perpendicular magnetization which is typical for Au/Co/Au systems. Magnetic domain imaging has been performed by means of optical polarizing microscopy in the P-MOKE configuration. The microscope was equipped with a digitally cooled CCD camera. The magnetic contrast of each domain structure has been improved by image processing including subtraction of a reference image after saturation of the sample. DW displacements ΔL have been determined from comparison of remanent domain structure images in zero field and after the application of a 0.01–1 s long magnetic field pulse t. The DW velocity v has been determined from the slope of $\Delta L = f(t)$ for different fields close to coercivity. The DW displacements have been measured in two in-plane directions [parallel ($\parallel$) and perpendicular ($\perp$) to Si$[-110]$] as shown in Fig. 2. The experiments reveal a strong increase (up to 2 orders of magnitude) of the velocity ratio $v_\parallel/v_\perp$ for samples with higher step density.

The clear exponential character of $v(H)$ strongly suggests that our system belongs to the class of weakly disordered media [11] and is in the low field regime where the propagation of the wall is determined by the overcome of microscopic energy barriers E_B. The field independence of the velocity ratio $[v_\parallel/v_\perp](H) = \text{const}$ for all samples reveals an identical mechanism of DW motion in the explored field regime. The height of the barrier E_B depends on the magnetic interactions and on the so-called activation volume V_A, which is a phenomenological measure of the volume of material which is swept out as the energy barrier is overcome. To understand the remarkable increase of the observed velocity and the anisotropy during the DW propagation found, we analyze E_B and V_A analytically.

The discussed energy barrier is determined by the following energy contributions:

$$E_B = \Delta E_{\mathrm{dw}} + \Delta E_m + HM_sV_A + K_{\mathrm{eff}}M_s^2V_A, \qquad (1)$$

where H is the applied field, M_s the saturation magnetization, K_{eff} the anisotropy energy density, ΔE_{dw} the change in the domain wall energy, and ΔE_m the change in the magnetostatic energy due to the elongation of the domain. In Eq. (1) all terms, except ΔE_{dw} and K_{eff}, are direction-independent and can be safely neglected.

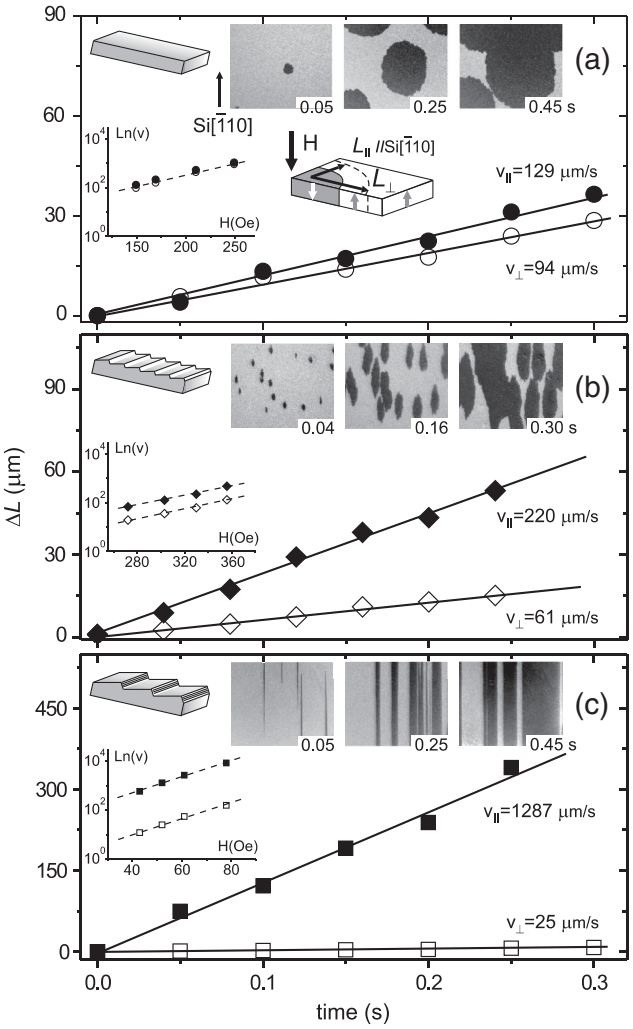

FIG. 2. DW displacement parallel and perpendicular to the Si$[-110]$ direction for: (a) an atomically flat surface, (b) monatomic steps, and (c) an array of step bunches. Insets show the field dependence of DW velocity and a selection of remanent domain structure images (220×170 $\mu\mathrm{m}^2$) recorded at different times.

The effective anisotropy consists of the dipolar K_d, the magnetocrystalline K_{ma}, and the magnetoelastic K_{me} contributions. While the first term does not show any significant directional variation, K_{ma} includes a weak in-plane step-induced $[-110]$ uniaxial anisotropy K_{vic}, and K_{me} may change for step bunches, which can be regarded as very rough facets [12]. Significant changes in K_{me} would result in the variation of the in-plane K_{vic} and out-of-plane K_{u1} contributions [10] of K_{eff} on a vicinal surface with respect to those of an isotropic Co film. Our recent MOKE measurements [13], however, do not show any significant changes either in K_{vic} or in K_{u1} for Au/Co/Au films investigated here. To cross-check these results, we have

performed additional measurements of K_{eff} by means of a magnetic resonance technique showing that K_{eff} is out-of-plane and has an identical magnitude for both isotropic Co(0001) flat and stepped surfaces within experimental accuracy. From this we conclude that, while K_{me} has to be taken into account for certain materials or substrate geometries, our epitaxial Co films on stepped Si(111) substrates cannot be approximated by an isotropic bent magnetic film and, therefore, strained or stressed in the area of step bunches, and one has to look for other mechanisms affecting the propagation of magnetic domain walls.

To tackle this problem, we assume that the epitaxial growth of Co on a stepped surface may be represented by a sequence of ideal Co layers with partially broken bonds as shown in Fig. 3. Figure 3(a) gives side views of 3 ML of Co on a flat (111) surface, on vicinal planes with monatomic steps, and on a step bunch. Comparing the three cases, one recognizes that two atoms at each step on the vicinal surfaces lose part of their nearest neighbors as outlined in Fig. 3. Another remarkable feature is the replacement of *ABA* by *BAB* stacking which leads to additional defects and mutual compensation of magnetoelastic *ABA* and *BAB* contributions on bunched surfaces. The loss of nearest neighbors leads to a weaker magnetic binding of the moments at the step edges and, hence, to the anisotropy in the exchange coupling along and perpendicular to the steps or bunches [14]. To elucidate this point, top views of

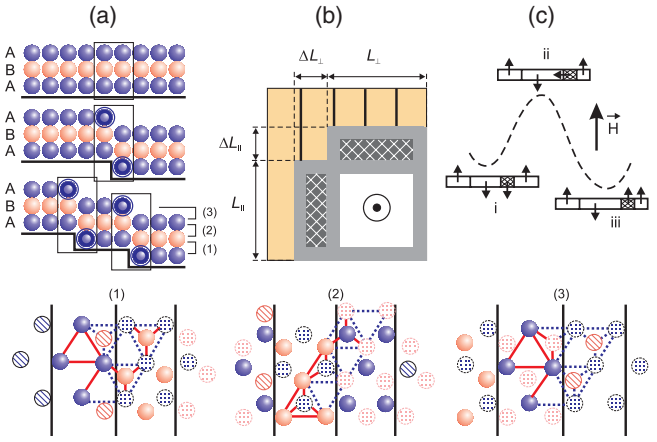

FIG. 3 (color online). Analytical model. (a) Side view of a continuous film, a monatomic step, and a step bunch consisting of three monatomic steps. The atoms belonging to the *A*-hcp planes are dark gray (blue), while those of *B*-hcp planes are light gray (red). The top views of bottom (3), middle (2), and top (1) atomic layers are shown at the bottom of the figure. Hatched circles denote missing atoms; dotted balls represent the atoms lying in the underlayer. Three typical unit cells with broken bonds are outlined by parallelograms. Solid and dashed lines denote intact and broken lines, respectively. (b) Schematic representation of a nucleated domain (white square) with domain walls (gray bars) and activation volume V_A (hatched areas). (c) Schematic representation of the thermal energy barrier and corresponding orientation of magnetization in the activation volume.

three typical unit cells at three levels of a step of the hcp(111) vicinals are shown at the bottom of Fig. 3. The intact nearest neighboring bonds are depicted by solid lines, while the broken ones by dashed segments.

The decreased exchange energy per atom at steps substantiate the assumption that the domains nucleate mainly at these defects. Moreover, since the exchange energy is lost only in the domain walls, it is energetically preferential to place the walls on the top of the steps or step bunches as visualized in Fig. 3(b). As we are mainly interested in the comparison of domain wall growth in two perpendicular directions, we analyze two basic situations—elongation of the nucleated domain along the steps and perpendicular to them. The activated volume in Fig. 3(c) depicted by the hatched rectangles in Fig. 3(b) is $V_A^{\parallel} = \Delta L_{\parallel} \times L_{\perp} \times d$ in the first case, while $V_A^{\perp} = \Delta L_{\perp} \times L_{\parallel} \times d$ in the second, with d denoting the Co thickness.

For $E_B \geq kT$ the waiting time τ before a barrier is overcome at a temperature T is described by an Arrhenius law of the form

$$\tau^{-1} = \nu_0 e^{(-\Delta E_B/kT)}, \tag{2}$$

where ν_0 is an attempt frequency and k the Boltzmann factor. In our case τ is the time needed for the walls to elongate by $\Delta L_{\parallel}$ or $\Delta L_{\perp}$. Hence, in a steady domain wall motion regime, the domain wall velocity is just $v_{\parallel(\perp)} = \Delta L_{\parallel(\perp)}/\tau_{\parallel(\perp)}$. The velocity ratio is inversely proportional to the ratio of corresponding waiting times. According to the model of Fig. 3, the strongest anisotropic contribution in Eq. (1) is given by $E_{\mathrm{dw}}^{\parallel(\perp)}$. Therefore, the velocity ratio becomes

$$\frac{v_{\parallel}}{v_{\perp}} = \frac{\Delta L_{\parallel}}{\Delta L_{\perp}} e^{[(E_{\mathrm{dw}}^{\perp} - E_{\mathrm{dw}}^{\parallel})/kT]}. \tag{3}$$

The exponent of Eq. (3) can be rewritten as

$$E_{\mathrm{dw}}^{\perp} - E_{\mathrm{dw}}^{\parallel} = 4d(\Delta L_{\parallel}\sqrt{\bar{A}_{\parallel} K_{\mathrm{eff}}} - \Delta L_{\perp}\sqrt{\bar{A}_{\perp} K_{\mathrm{eff}}}), \tag{4}$$

where $d\Delta L_{\perp(\parallel)}$ is the area of the domain wall and $\bar{A}_{\perp(\parallel)}$ are components of the exchange stiffness tensor along two directions. It is natural to postulate that the elementary elongation of a domain wall is on the order of the exchange length $\Delta L_{\parallel(\perp)} \approx \sqrt{A_{\parallel(\perp)}/K_{\mathrm{eff}}}$. Notice that for Bloch walls assumed here the elongation along the steps ($L_{\parallel}$) costs energy perpendicular ($A_{\perp}$) to the steps [15]. Therefore Eq. (3) reduces to

$$\frac{v_{\parallel}}{v_{\perp}} = \sqrt{\frac{\bar{A}_{\parallel}}{\bar{A}_{\perp}}} e^{[4d(\bar{A}_{\parallel} - \bar{A}_{\perp})/kT]}. \tag{5}$$

To determine the effective parameter $\bar{A}_{\perp(\parallel)}$, one has to first calculate the components of the exchange stiffness tensor at a site i using $A_{\perp(\parallel)}^{i} = nJS^2 \sum_{j=1}^{z} \omega_{ij}$ [16]. Here J is the exchange coupling constant, n the number of atoms in a unit cell, z the coordination number, and ω_{ij} the

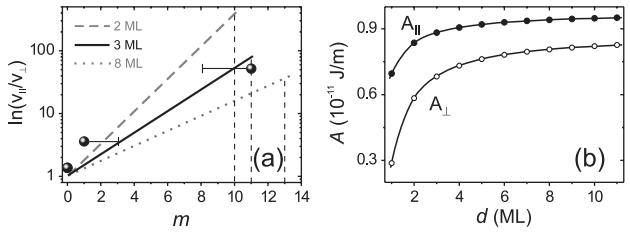

FIG. 4. (a) Theoretical prediction for $v_{\parallel}/v_{\perp}$ as a function of the number of steps m belonging to the domain wall for three different film thicknesses and experimental data for 3 ML Co films. Vertical dashed lines show the width of domain walls. (b) Theoretically calculated exchange stiffness at the step as a function of film thickness.

expansion of direction cosines in powers of bond projections on the chosen direction [15,16]. Taking $J = 1.2 \times 10^{-21}$ J from Ref. [17], we find $A_{\perp}^{\mathrm{flat}} = A_{\parallel}^{\mathrm{flat}} = 1.6 \times 10^{-11}$ J/m for 3 Co ML on Au/Cu/Si(111), in good agreement with the literature [18]. At step edges the exchange stiffness is reduced to $A_{\perp}^{\mathrm{step}} = 0.7 \times 10^{-11}$ J/m and $A_{\parallel}^{\mathrm{step}} = 1.3 \times 10^{-11}$ J/m. These values are valid for steps with terraces of width $b \geq 3a$, with a being the lattice constant [see Fig. 4(a)].

Next, the exchange stiffness has to be averaged over the width of the domain wall $l \approx \sqrt{A/K_{\mathrm{eff}}}$. As the reduction of exchange energy takes place only at steps, the averaging can be done by $\bar{A} = [mA^{\mathrm{step}} + (N - m)A^{\mathrm{flat}}]/N$, where m is the number of steps in a wall ($m = 0$ for a flat film) and N is the width of the domain wall measured in steps $N \approx l/(da)$. The logarithmic representation of velocity ratios predicted for different numbers of steps in the bunches is given in Fig. 4(a). The dependence [Eq. (5)] shows exponential growth of $v_{\parallel}$ with respect to $v_{\perp}$ which is in good agreement with the experimental data. The anisotropy in the domain wall velocity ($v_{\parallel}/v_{\perp} \approx 50$) for a bunched surface is enormous in comparison with the asymmetry of forward and backward domain wall mobility in Co/Pt multilayers reported recently [19].

In the following, we analyze the thickness dependence of the velocity ratio in the framework of our model. For monatomic steps depicted in Fig. 3, the thickness increase corresponds to the increasing number of ''bulk'' unit cells of type (2) (see Fig. 3), while only one surface cell of type (1) and one of type (3) exist. Although the exchange stiffness of the cell (2) is the largest among the three variants, it is still almost 2 times smaller than that of a continuous film because of mixed *ABA-BAB* stacking. Therefore, a thickness averaged exchange stiffness of a step with an hcp stacking converges towards $A_{\perp}^{\mathrm{step}} = 0.83 \times 10^{-11}$ J/m$^{-1} < A_{\parallel}^{\mathrm{step}} < A^{\mathrm{flat}}$ [see Fig. 4(b)]. After introducing these data into the relationship for $\bar{A}_{\perp}$, we get the velocity ratios for stepped Co films of different thickness. Three of them are exemplified in Fig. 4(a). Hence, assuming the topography of Fig. 3, the anisotropy between $v_{\parallel}$ and $v_{\perp}$ will persist for all thicknesses below the spin reorientation transition at ≈ 8 ML of Co [13]. The prediction given in Fig. 4 is general for any material with an hcp stacking. In order to apply this model to systems with different values of anisotropy, one has to determine the width of the domain wall N as described above. In the case of materials where K_{me} is important, the value of corresponding K_{eff} has to be introduced into Eq. (5).

In summary, we establish the importance of the density of steps on vicinal substrates for determining exchange interactions at the atomic scale in ultrathin magnetic films. We show for ultrathin Au/Co/Au films with perpendicular magnetic anisotropy that the exchange stiffness at monatomic steps becomes anisotropic. By means of analytical calculation combined with experimental studies, we demonstrate that the change of the exchange stiffness tensor turns out to be a driving force for highly anisotropic dynamics of magnetic domain walls. In particular, the velocity of domain walls oriented perpendicularly to the steps can be increased by 2 orders of magnitude for bunched surfaces.

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