Model of Fe nanostripes on Cu(111)

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Magnetization as a function of temperature calculated with Monte Carlo techniques is compared to experimental results of Fe stripes grown on vicinal Cu(111) surfaces. The stripes are step decorations grown with molecular beam epitaxy (MBE), are 1–2 monolayers thick, and display perpendicular magnetization. The atomic parameters are determined from fully relativistic electronic structure calculations. The moments are found to be 2.57 μ_B, with some variation due to film thickness, and uniaxial anisotropy of 40 μRy/atom for Fe atoms facing vacuum. The Heisenberg model extended to include crystalline anisotropy as well as dipole–dipole interactions is considered for two different values of the exchange constant: $J=20$ and 2 meV. Under a large applied field (4000 G), the calculated saturation magnetization falls slowly from 507 emu/cm^3 with an increase in temperature until it falls rapidly around 600 K, after which a more modest falloff with an increase in temperature is observed. For larger J the rapid change occurs for higher temperatures. The importance of disorder in the height and width of the stripes is investigated by generating stripe geometries with a model that incorporates nucleation and growth of Fe particles at step edges under the constraint of constant deposition from MBE. The primary effect of disorder in the stripes is to reduce the saturated magnetization at lower temperatures.
[DOI: 10.1063/1.1452252]

Recently, magnetic measurements have been performed on monolayer-high Fe stripes grown on a vicinal Cu(111) surface.[1,2] The nearest step–step distance on the Cu(111) surface is ~10 nm. Parallel, aligned Fe stripes were formed via step decoration of Fe atoms on the upper terrace of the step edges. The height and width of the Fe stripes vary depending on the nominal thickness. At a nominal thickness of 0.8 monolayer (ML), the Fe stripes are 1–2 atomic layers in height and ~5 nm in width. Due to the nature of step decoration, the Fe stripes have a certain edge roughness that is estimated to be about 2 nm. A scanning tunneling microscopy (STM) image of the stripes is shown in Fig. 1.

The magnetic measurements performed by the *in situ* magneto-optical Kerr effect (MOKE) indicate that the easy magnetization axis is along the surface normal. The saturation magnetization along this axis is shown versus the temperature in Fig. 2. The remanent magnetization of the stripes is substantially smaller than the saturation magnetization, and has a distinct time-dependent behavior in the absence of an external field. At temperatures higher than 160 K, the magnetization decays to zero in less than 1 s after switching off the external field.

Here we present numerical estimates of the properties of Fe nanostripes using a combination of first-principles calculations and a Heisenberg model. The iron nanostripes are modeled as a set of rectangular cells. Each uniformly mag-

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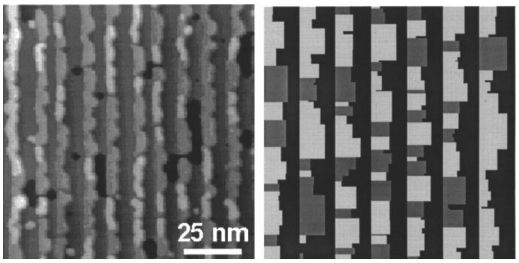

FIG. 1. Left: STM image of a 120 nm×120 nm section of Fe nanostripes constructed on step edges on vicinal Cu(111). Right: 21 nm×21 nm sections of model nanostripes constructed via Eq. (2). The black regions are the substrate, the dark gray regions stripes that are 1 ML thick, and the white regions stripes that are 2 ML thick.

netized cell has a three-dimensional total magnetization vector $\mathbf{S}_i$ whose magnitude is fixed. The energy of the system is given by a two-dimensional classical Heisenberg Hamiltonian,

$$H=\sum_i K_i[1-(\hat{\mathbf{s}}_i\hat{\mathbf{K}}_i)^2]-\sum_{\langle i,j\rangle} J_{i,j}\hat{\mathbf{s}}_i\hat{\mathbf{s}}_j-\frac{1}{2}\sum_{i,j}\hat{\mathbf{s}}_i\mathbf{D}\hat{\mathbf{s}}_j, \quad (1)$$

extended to include crystalline anisotropy[3] and dipole–dipole interactions. The exchange due to magnetization in adjoining cells is $J_{i,j}$, while the contribution from the crystalline anisotropy of the surface atoms in each cell is K_i. The matrix $\mathbf{D}=-m_im_j(3\hat{\mathbf{r}}_{i,j}\hat{\mathbf{r}}_{i,j}-1)/r_{i,j}^3$ is the dipole–dipole interaction energy between ith and jth cells, with m_i and m_j the total magnetization of those cells.

Fully relativistic first-principles calculations provide the following parameters for Fe on Cu(111). The magnetic moment for a Fe atom in a monolayer film is found to be 2.57 μ_B, while in a two-monolayer film the magnetic moment is 2.56 μ_B and 2.46 μ_B for atoms in the top and bottom layers, respectively. To simplify the model, we always use the value for atoms in the monolayer which corresponds to a magnetization density of 2.384×10^{-20} erg/G. For Fe only a few monolayers thick, the magnetization is directed perpendicular to the substrate as a consequence of strong uniaxial anisotropy. The calculations show this anisotropy to be significant only for atoms facing the vacuum, with strength of 5.4 $\times10^{-4}$ eV/atom. Thus the anisotropy constant for the ith model spin, K_i, is taken to be proportional to the number of surface atoms in the cell. The value of the exchange constant J has not yet been determined through first-principles calculations.

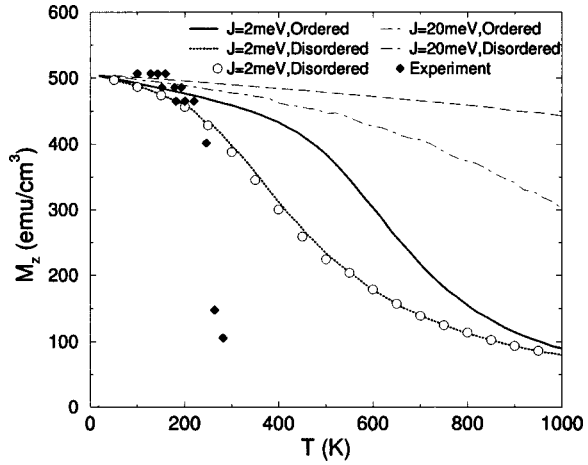

FIG. 2. Calculated saturation magnetization perpendicular to the substrate, M_z, at an externally applied field of 4000 Oe. All calculations neglected dipole–dipole interactions except the one shown by ○. The experimental data were normalized to 507 emu/cm^3 as $T\rightarrow0$ K.

The nanostripes fabricated, seen in the left-hand panel of Fig. 1, display variations in width (perpendicular to the step edge) that have the potential to significantly impact the magnetic properties. In order to construct individual model stripes that are spatially disordered, we use a model of nucleation and growth of new islands of Fe under the assumption of constant deposition. Consider a step edge oriented along the $\hat{y}$ axis. Nucleation is assumed to occur along the edge with a rate I events per unit time per unit length, and only where no island already exists. The material deposited on the step is assumed to accumulate at the nearest island. Assuming the rectangular island is fixed at its left edge (where $x=0$), it is defined by y_t, y_b, and x, the position of the top, the bottom, and the right edges, respectively. The growth of the surface area, A, of an island is

$$\frac{dA}{dt}=\frac{dx}{dt}(y_t-y_b)+x\left(\frac{dy_t}{dt}-\frac{dy_b}{dt}\right)+\frac{dx}{dt}\left(\frac{dy_t}{dt}-\frac{dy_b}{dt}\right), \quad (2)$$

with the constraint that edges in contact with other islands do not grow. Assuming the unconstrained edges grow at the same rate, the growth for a short time interval is found by solving

$$n(\Delta x)^2+\Delta x[nx+(y_t-y_b)]-\Delta A=0, \quad (3)$$

where edges move Δx during the interval, n is the number of unconstrained edges perpendicular to the step, and ΔA is the change in interfacial area during the interval.

The model system for the Fe stripes at step edges on vicinal Cu(111) is 42 nm×42 nm with a separation between step edges of 3 nm. Matching the experimental system, 0.8 ML of iron is assumed to cover one half of the copper surface. Two types of model systems are considered: (1) those with well-ordered stripes discretized into 1.5 nm×1.5 nm cells of uniform thickness and (2) those with disordered stripes generated via Eq. (2) with cells randomly chosen to be 1 or 2 ML thick. For both cases each cell is represented by one spin. The exchange constant $J_{i,j}$ between cells is based on the exchange J between atoms and the contact area between the cells. Each calculation started with uniformly oriented spins perpendicular to the substrate and involved 5000 relaxation Monte Carlo steps per spin (MCSS) followed by approximately 10^5 MCSS used to sample the magnetization. The Metropolis acceptance criterion[4] was used, and each proposed step consisted of rotating a spin within a solid angle adjusted to keep the acceptance ratio[5] near 0.42.

The resulting saturation magnetization with an external field of 4000 Oe applied perpendicular to the substrate is shown versus the temperature for $J=2$ meV, the solid curve, and $J=20$ meV, the dashed curve, in Fig. 2. For $J=2$ meV, the crossover from large to small saturation magnetization occurs at approximately 600 K, while for $J=20$ meV the crossover temperature exceeds 1000 K. Com-

parison to the experimentally observed crossover temperature of about 250 K supports the fact that exchange within the Fe stripes is more than an order of magnitude smaller than in bulk Fe. However, the model still overestimates the crossover temperature. Disorder in the stripes may be responsible for the lower crossover temperature.

The crossover temperature associated with disordered samples is indeed lower. For the system with an edge-decoration nucleation rate $I=1.5\ \mathrm{nm}^{-1}\ \mathrm{ns}^{-1}$ and deposition rate of 0.5 ML/ns, shown in Fig. 2, the crossover temperature for $J=2$ meV is 400 K. Different realizations of disorder for this I lie within 5% of the curve shown. However, changing I changes the crossover temperature more significantly, and details of the disorder in the stripes may be important in determining the crossover temperature in the saturation magnetization curve. As a check that dipole–dipole interactions are not important, one calculation with these included, and the system treated as a supercell repeated infinitely in the x and y directions, is shown by the circles in Fig. 2. Here it can be seen that dipole–dipole interactions do not significantly affect the results.

The exchange constant J between two nearest neighbor Fe atoms adsorbed onto the Cu(111) surface has also been estimated by an atomistic Monte Carlo calculation[6,7] using Eq. (1). Using the crystalline anisotropy found by the first-principles calculations and ignoring dipole–dipole effects, the magnetization was calculated for a 64×64 system of Fe atoms arranged on a triangular lattice with the same atomic separation as that found in bulk Cu. By matching the temperature at which the equilibrium magnetization perpendicular to the substrate vanished to experimental results[2] for a monolayer of Fe on Cu(111), a value of $J=7.5$ meV is found. This result indicates that the exchange coupling between Fe atoms in the film is much smaller than that observed in bulk Fe. Effects other than those discussed here could also reduce the crossover temperature by reducing exchange coupling between the segments of a stripe. One example would be between adjoining segments, each grown on one of the two degenerate sublattices of threefold hollow sites on the Cu(111) surface. This changes the interatomic distance and reduces the number of neighboring atoms across this mismatch.

The small remanence seen in the Fe stripes on time scales of seconds is consistent with the small activation volumes associated with these stripes. First, shape anisotropy is not a stabilizing influence because the magnetization is perpendicular to the long axis of the stripes. In addition, since reversal via spatially inhomogeneous magnetization configurations is possible for lengths above a few exchange lengths,[8] the activation volume for the stripes is on the order of 5×10^2 atoms. The activation energy corresponds to creating two Bloch walls, and using the parameters calculated here it corresponds to about $6k_BT$ at room temperature.

This work was supported by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory and DOE-OS through OASCR-MICS under Contract No. DE-AC05-00OR22725 with UT-Battelle LLC, as well as DOE-OS through BES-DMSE under Contract No. W-7405-82 with Iowa State University.

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