

Magnetic domains and magnetostatic interactions of self-assembled Co dots

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Submicron quasi-hexagonal Co dots and dot chains have been grown epitaxially on Ru(0001) substrate with molecular beam epitaxy and investigated with magnetic force microscopy. All the dots exhibit in-plane single domains. The magnetization of the dots in a chain prefers to align along the chain direction due to inter-dot magnetostatic interactions. Micromagnetic calculations suggest that most observed single-domains are metastable energetically, but both vortex and single-domain can exist in zero-field. The inter-dot magnetostatic coupling, modeled with a dot-pair, introduces a uniaxial anisotropy with easy axis along the pair/chain direction. The induced anisotropy field decreases with increasing inter-dot distance and approaches the dipolar limit.

Small magnetic structures have been attracting increased attention due to their impacts on applications. Magnetic dot arrays, for example, have been seriously discussed^{1,2,3,4,5} as an alternative approach to ultra-high-density storage media. When lateral sizes of the magnetic objects approach sub-microns to nanometers, the ground states and their stability⁶ in magnetic structure, as well as their interaction⁷ become crucial issues in constructing discrete media.⁸ In addition, metastable states may exist depending on magnetization history, which has not been discussed in detail. Previously, we have reported the formation⁹ and linear alignment¹⁰ of self-assembled Co dots on a Ru (0001) single crystal with molecular beam epitaxy (MBE). Truncated quasi-hexagonal Co dots, ~ 90 -600 nm in diameter, depending on film coverage, have been observed to have a relatively tight size distribution. Such self-assembly is attributed⁹ to strain relaxation in epitaxial growth, which is being extensively utilized in self-assembled quantum dots of semiconductor.¹¹ In addition, these sub-micron

dots can grow into dot-chains and continuous stripes on grooved substrates.¹⁰ These systems provide a test ground for investigating magnetic properties of sub-micron structures. In this work, magnetic properties of these self-assembled Co dots and dot pairs/chains have been studied with magnetic force microscopy (MFM) and micromagnetic calculations. In-plane single domains were observed on all dots. After calculating the ground states of the dots, which indicate the experimental single-domain state as metastable, our micromagnetic calculations mainly focus on the *stability* of both the ground and metastable states by adding magnetic field to convert from each other. The existence of such a nearly bi-stable region has, to our best knowledge, not been modeled previously. The closely situated dots in dot-chains tend to have their magnetizations lined up with each other, which is understood as the result of a sizable dipolar-like magnetostatic interaction among dots.

Self-assembled Co dots have been grown with MBE on Ru(0001) single crystal at elevated temperature in an ultra-high-vacuum (UHV) system. The experimental details on substrate preparation and sample growth can be found in previous publications^{9,10}. The morphology of the film and its magnetic structures were examined *ex situ* with atomic force microscopy (AFM) and MFM (Nanoscope III, digital instruments) in the as-grown virgin state. A lift-scan mode was used for MFM measurements with scan height ~60-120nm. Standard commercial Co-Cr tips with medium moment ($\sim 3 \times 10^{-12}$ em μ) and coercivity (~ 400 Oe)¹² were used to map the magnetic domain structures of the dots. The tip was magnetized perpendicular to the film plane prior to measurement. No magnetization direction switch caused by the tip local field was observed on fresh sample.

The micromagnetic simulations were performed with the public code OOMMF.¹³ The program employs a two-dimensional (2D) grid of square cells with 3D magnetic moments, which are constant in value at the centers of the cells. Magnetization M in a given field is determined by integrating the Landau-Lifshitz equation

$$\frac{dM}{dt} = \gamma M \times H_{\text{eff}} - \frac{\gamma\alpha}{M_s} M \times (M \times H_{\text{eff}}), \quad H_{\text{eff}} = -\mu_0^{-1} \frac{\partial E}{\partial M}$$

where M_s is the saturation magnetization, γ is the gyro magnetic ratio, and α the damping coefficient. The average energy density E includes the magnetocrystalline anisotropy, exchange, demagnetization (magnetostatic), and Zeeman energy terms. Further technical details of the program can be found in the literature¹⁴ and in the OOMMF User's Guide¹³. The dots are assumed to have a hexagonal shape with diameters, defined as the distance between the opposite corners, of $\sim 90 - 900$ nm and thickness of $\sim 0.2 - 12$ nm. Parameters from bulk Co were used in the calculations. Specifically, $M_s = 1400 \times 10^3$ A/m, $A = 30 \times 10^{-12}$ J/m,

$K_1=520 \times 10^3 \text{ J/m}^3$, and $\alpha=0.5$. The cell size was chosen as 6nm for most of the calculations, though other cell sizes were also examined to estimate the errors in such modeling.

Figure 1 shows the corresponding (a) AFM and (b) MFM images of typical Co dots formed by strain-induced self-assembly during epitaxial growth.^{9,10} The horizontal grooves on the substrate cause the dots to roughly align into dot chains. The MFM image (Fig.1b) clearly reveals magnetic contrast on each dot. The arrows in (a) illustrate the inferred magnetization orientations in each dot. All the dots exhibit in-plane single domain in their virgin state, regardless of the spacing among them. The magnetizations of the closely situated neighboring dots tend to line up along a chain direction. No globally preferred magnetization direction, however, is observed. This magnetic correlation along the chains, therefore, is not due to any external field but indicates a significant role of the dipolar-like magnetostatic coupling among the dots. A dipolar coupling is known to align the magnetic moments, i.e., the dipoles, along the chain. The fact that no moment is perpendicular to the chain also indicates a uniaxial magnetic anisotropy with its easy axis along the chain direction.

To investigate the ground state of single dots, Fig. 2 plots the total energy density of dots in either vortex or single domain states as a function of (a) dot lateral size and (b) thickness. As seen from Fig.1a, for dots with a thickness of 6nm, single-domain configuration is the ground state with the dot diameter $< 120\text{nm}$, while vortex becomes energetically favorable for dot diameter $>120\text{ nm}$. Similarly, there is also a crossover point, i.e., $\sim 2.2\text{ nm}$ at a lateral size of 288 nm, as a function of thickness (see Fig.2b). This is basically in agreement with previous results on other magnetic dots^{15,16,17} and are qualitatively understood with a simple physical picture: for small dots, exchange energy dominates, which favors single domain states. For relatively large dots, however, demagnetization energy can increase significantly in an in-plane single domain configuration, but can be minimized in a vortex state. The experimentally observed single-domains in larger and thicker Co dots are, therefore, metastable states. We believe that during the growth process, the dots initially are small and naturally stable in single-domain states. When the dots grow bigger, they may become trapped in these metastable states due to an energy barrier to convert into the ground state.

To evaluate the stability of the magnetic state, we calculate the magnetic field needed to convert between the two states in a dot with vortex as the ground state, as seen in Fig. 3. Exchange energy turns out to be convenient in distinguishing quasi-single-domains and quasi-vortex states, since the former possess less exchange energy than the latter. With vortex as the initial state, Fig. 3(a) shows the exchange energy as a

function of external field, which is gradually increased and then decreased along the y -direction. The sudden drop in exchange energy indicates the annihilation of the vortex, which is driven out of the dot and results in a sharp decrease in average angle among the moments. Interestingly, even though the vortex state is the ground state, once the vortex annihilation occurs, the system relaxes into a metastable single domain state when the field is decreased back to zero. The system, therefore, is bi-stable, depending on magnetization history. The annihilation field of a vortex, derived from Fig.3a, is about 36 mT for a typical Co dot with ~ 288 nm in diameter and ~ 6 nm in thickness. Such a behavior was earlier seen in the calculations of circular permalloy dots, though not discussed in terms of bi-stable states.¹⁷ The annihilation and nucleation of the vortex state in thicker films are discussed in Ref. 18 and 19, where, unlike the bi-stable states we predict for thinner films, the saturated single-domain states always reverse back to the vortex ground state at zero field.

Fig.3b plots the exchange energy as a function of the external field normal to the surface for an initially single-domain metastable state. When the maximum field is kept below a critical value, the system always relaxes back to the original single-domain state, as seen in Cycle I. When the field exceeds this critical value, a jump in exchange energy indicates the sudden reshuffling of magnetization and therefore the formation of a potential vortex. This critical field, i.e. a creation field for a vortex state, is ~ 770 mT for the same dot as in Fig. 3a. It suggests a sizable energy barrier to convert a single-domain state into a vortex state. Further calculations show that this field / energy barrier is very sensitive to the shape, lateral size, and thickness of the dot and decreases away from the phase boundary between the vortex and single-domain states.

To discuss inter-dot interactions, Fig. 4a shows the angular dependence of demagnetization energy at saturation for dot pairs with different inter-dot spacing. The energy values are normalized by that of a single dot to correct for any artificial effect from the coarseness of the dot edges. The two-fold symmetry of the curve is indicative of a uniaxial anisotropy with the easy axis along the pair direction, i.e. 90° in the figure. The magnitude of in-plane magnetic anisotropy can be measured from the difference in total energy, and therefore demagnetization energy, between easy (90°) and hard (0°) axis. As seen in Fig. 4a, such an anisotropy decreases with increasing dot spacing. Fig. 4b plots the anisotropy energy as a function of inter-dot distance. The anisotropy energy from pure dipole-dipole interaction with the same total moment was also drawn in the figure for comparison. The results of the micromagnetic calculation approach to the dipolar limit when the dots are far apart, but deviate from the dipolar $\sim 1/r^3$ decay when they are close (i.e., the center-to-center distance is less than

2 times of the dot size). This uniaxial anisotropy energy can also be expressed as anisotropy field as seen on the right side of the figure. The anisotropy field for two contacting dots is about 270 Oe, and decreases to ~ 5 Oe for dots at distance of ~ 640 nm. This uniaxial anisotropy is consistent with the one observed in Fig. 1. It should be noted, however, that additional anisotropy may also exist due to the growth on grooved substrates.

We have observed in-plane single domains in all the Co dots. Micromagnetic simulations on annihilation and creation of a vortex in a Co dot reveals that both single-domain and vortex states can be preserved at zero field, even when the vortex state is the ground state. Angular dependence of the demagnetization energy in dot-pairs shows that the magnetostatic coupling among the dots leads to a sizable two-fold anisotropy with easy axes along dot pair (chain) direction. The anisotropy energy decreases with inter-dot distance rapidly to approach the dipolar limit.

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Figure 1. a) AFM and b) MFM image of self-assembled Co dots, grown on Ru (0001) substrate at nominal thickness of ~ 1.7 nm. The dots have an average lateral size of ~ 300 nm with mean height ~ 4 nm. The arrows in the AFM image indicate the magnetization orientation in each dot as derived from the MFM image.

Figure 2. a) Total energy of a hexagonal Co dot as a function of lateral size for single-domain and vortex configurations. The equilibrium single-domain and vortex states at zero fields are shown as insets. b) Total energy of a hexagonal dot as a function of thickness for vortex and single-domain state.

Figure 3. Simulation of the conversion between the vortex and single-domain states under external fields, as monitored with exchange energy, for a Co dot of 288 nm diameter and 6 nm thickness with a vortex ground state. a) vortex to single-domain under an in-plane field; b) single-domain to vortex under a perpendicular field.

Figure 4. a) Angular dependence of saturation demagnetization energy in dot-pairs with different inter-dot distance S . The dot pair, with each dot 288nm in diameter and 6nm in thickness, is shown schematically in the inset. b) Magnetostatic anisotropy energy and the corresponding anisotropy field of the dot pair as a function of inter-dot center-to-center distance. The dotted line indicates a pure dipolar interaction for comparison.

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