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The formation of FePt nanoclusters via gas condensation has attracted a great deal of attention. The clusters normally form with the magnetically soft $A1$ structure rather than the desired $L1_0$ structure with high magnetocrystalline anisotropy. This work has examined the effects of plasma characteristics on the early stages of order in the formation $L1_0$ FePt nanoclusters via inert gas condensation. The plasma characteristics have been modified to control ion density in the nanocluster condensation region. Increased ion density results in more cluster-ion collisions. The energy imparted to the clusters as a result of these collisions allows atomic rearrangements to form the ordered structure. The results indicate that controlled ion density directly impacts the early stages of FePt nanocluster ordering, according to high-resolution electron microscopy structure observations and coercivity measurements. © *2010 American Vacuum Society.*
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I. INTRODUCTION

Due to their large magnetocrystalline anisotropy, $L1_0$-structure FePt nanoclusters are one of the more promising candidates for high density recording media.[1,2] However, most preparation methods produce the disordered, magnetically soft $A1$ phase and require additional processing which can introduce contaminants or result in particle agglomeration.[3] Additionally, experiments and simulations indicate that phase *transformation* from $A1$ to $L1_0$ may be kinetically inhibited in nanoclusters smaller than 4 nm.[4] A method that allows formation of nanoclusters already in the $L1_0$ state, without the need for postformation thermal treatments, would not only save significant processing cost, it would increase information storage density for recording media, beyond what is currently projected.

Recently, formation of FePt nanoclusters via inert gas condensation has yielded direct formation of the partially ordered, magnetically hard $L1_0$ structure.[5,6] This method promises a high degree of cluster property control. To date, efforts have not focused on control of specific plasma parameters, such as ion density and electron temperature. Rather, control efforts have relied upon external "knobs" (e.g., target power) that alter several unmeasured plasma conditions simultaneously (e.g., electron temperature, collision probability, and ion density). While it is not possible to alter only one plasma parameter without affecting others, it is possible to measure the effects of the alteration, with the goal of minimizing them. Previous Langmuir probe measurements[5] indicated a two-order magnitude increase in ion density by alter-

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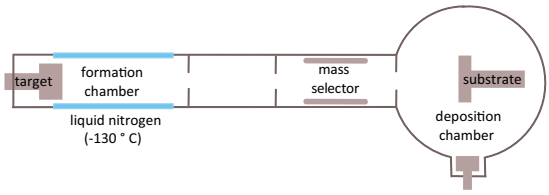

FIG. 1. (Color online) Schematic picture of nanocluster fabrication system. The formation chamber is on the left, inside the cooled region; the deposition chamber is on the right, near the rotatable substrate. The FePt target is the leftmost object in the chamber.

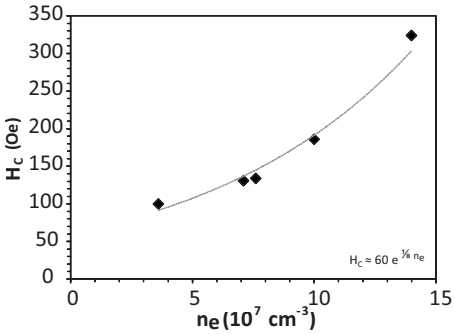

FIG. 2. (H_C) as a function of ion density (n_e). Confidence is $\pm 1 \times 10^7$ cm^{-3} and ± 5 Oe.

ing plasma processing parameters in a recipe known to produce $L1_0$ precursors successfully. The higher ion density created FePt clusters with a coercivity of 1.1 kOe compared to a coercivity of a few oersted in clusters formed in the low-density conditions. The higher coercivity suggests that the clusters formed with at least partial $L1_0$ ordering in high ion density conditions. A new experiment was needed to investigate early stage $L1_0$ structure formation as a function of plasma ion density. The eventual goal of this research is to discern causal mechanisms of direct $L1_0$ FePt nanocluster formation so that it may be systematically controlled in manufacturing applications.

II. EXPERIMENT

Inspired by the previous results, the current work was performed to better understand the relationship between plasma characteristics and FePt cluster formation. It is a step toward specific plasma process control over direct FePt $L1_0$ cluster preparation. In particular, we have systematically controlled ion density, while minimizing and measuring other plasma parameters' resulting changes. We were thus able to determine the effect of ion density on cluster properties, early in the direct ordering process.

FePt clusters were created by inert gas condensation[1–3,7–13] from a composite sputtering target consisting of Pt pieces in an Fe target, biased at 360 V with 150 W, drawing 330 mA of plasma current (Fig. 1). Argon introduced at 144–700 SCCM (SCCM denotes cubic centimeter per minute at STP) into the source chamber created source pressures of between 130 and 540 mTorr. The deposition chamber pressures of between 2 and 7 mTorr were controlled via argon flow rates between 25 and 200 SCCM. A quarter inch, double-sided Langmuir probe,[14] approximately 2 in. from the target, collected electron temperature, current, and ion density information. Electron temperatures remained between 0.1 and 1.1 eV during parameter variations. Samples were analyzed for coercivity using a Micromag 2900 alternating gradient force magnetometer at room temperature and for structure with high-resolution transmission electron microscopy (HRTEM). Imaging was accomplished on clusters directly deposited onto carbon support TEM grids using a Tecnai G2F20 operating at 200 kV.

III. RESULTS

Investigation of the causal mechanism for ordered structure evolution *in situ* led to the confirmation of the expected trend: cluster coercivity (H_C) varied proportionally with ion density (n_e) (Fig. 2). An exponential relationship was observed. Pressure differences ($\Delta p = p_f - p_d$) between the formation (p_f) and deposition (p_d) chambers were used to alter the measured ion density. Lower pressure differences create longer dwell times for particles in the formation chamber, thereby increasing the ion density. Higher ion densities in the formation chamber increase cluster-ion collision probability. The mechanism for direct heating (and ordering) of clusters is likely the cluster-ion collision. Cluster impact studies show that cluster-ion collisions are responsible for charging clusters and transferring heat.[9] Therefore, increased ion density should promote ordering in forming FePt clusters. Because the $L1_0$ structure has a large magnetocrystalline anisotropy compared to the $A1$ structure, coercivity is a good indicator of the presence of $L1_0$ order. Thus, the observed increases in coercivity with ion density suggest an increase in $L1_0$ order.

HRTEM images revealed structural and morphological differences as a function of plasma parameters, as has been seen elsewhere. (A transition from nominally spherical clusters to highly faceted clusters was observed as ion density increased.) The HRTEM results suggest that the $L1_0$ structure may begin to form after initial cluster formation in the $A1$ structure, as evidenced by twinning that arises from the formation of multiple ordered variants (MOVs) from the parent $A1$ structure. The images alone cannot distinguish the $L1_0$ from the $A1$ structure, however, as they show early, not final, stages of direct $L1_0$ structure formation. The MOV structure was absent from the clusters that were formed with lower ion density, where the HRTEM revealed essentially single-crystalline clusters (Fig. 3). Multiply twinned particles of similar icosohedral structure have been seen elsewhere in annealed FePt nanoclusters.[15] Fourier transforms of the images revealed identical (100) plane spacings for all visible plane orientations. Calculation of the cluster-cluster collision frequency has been performed and compared to the cluster-ion collision frequency to lend credence to the hypothesis that the MOV structure is more likely formed from the $A1$ structure. Calculations estimate 10^3 cluster-cluster

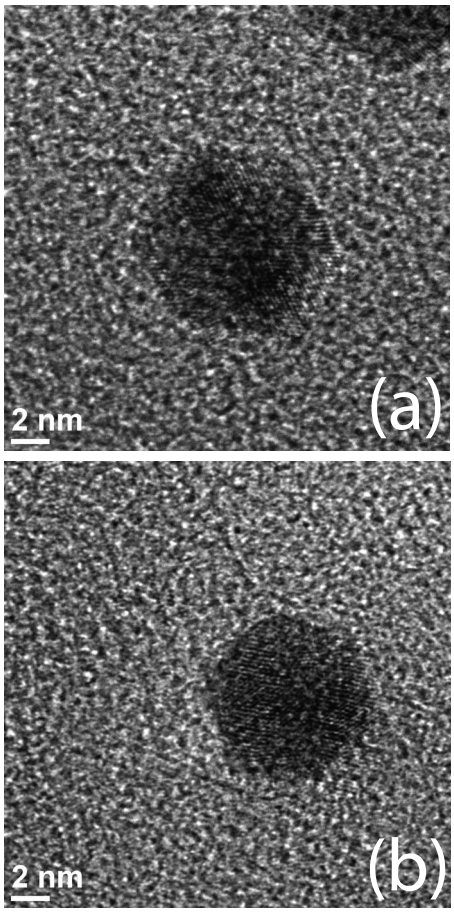

FIG. 3. HRTEM images of (top) variants on a single crystal from higher density ($\approx 20\times 10^7$ cm^{-3}) plasma conditions and (bottom) single crystal structure from lower density ($\approx 5\times 10^7$ cm^{-3}) plasma conditions.

collisions/s and 10^{20} cluster-ion collisions/s. Since cluster-cluster collisions are many orders of magnitude less frequent, this suggests that the MOV structures are grown during agglomeration. Since cluster-ion collisions are more frequent, it suggests the MOV structures grew from disordered $A1$ structures, heated by collisions with energetic ions. Because of the extreme difference in collision frequency, even if estimates were off by a few orders of magnitude, we can still confidently state that the MOV structure more likely forms from ion heating. Thus ion density is an early indicator of cluster ordering, and directly correlates with cluster coercivity.

IV. SUPPLEMENTAL INFORMATION: DETAILS OF COLLISION FREQUENCY CALCULATION

An estimate of cluster density was necessary to complete this calculation. The estimate was taken from the Langmuir probe current measurements at high bias voltages, using the same methods as those for calculating ion density from the same curve at lower bias voltages.[5,14] Using this method, the cluster density was measured at roughly 10^{14} cm^{-3}. To validate the estimate, cluster density was calculated another way. A two-dimensional TEM image of a 4 nm cluster film deposited at a rate of 5 nm/s for 80 s was used to gain an upper bound on the cluster density. The upper bound is on the order of 10^{18} clusters cm^{-3}. Since the cluster film represents the most densely packed cluster scenario, and since it is used in the calculation of both collision frequencies (and thus will not alter the relative collision frequencies of ion-cluster versus cluster-ion collisions), the lower estimate was taken.

The frequency (ν), with which an object collides with a cluster of density n_{cl}, is given by

$$\nu = n_{\mathrm{cl}}\sigma u,$$

where σ is the hard-sphere collision cross section (estimated with the 3 nm radius of an FePt cluster) and u is the velocity of the object colliding with the cluster. The average velocities for clusters (u_{cl}) and argon ions (u_{Ar}) are estimated at 9×10^{15} and 2×10^{18} cm/s, respectively. This gives the estimated 10^3 cluster-cluster collisions/s and 10^{20} cluster-ion collisions/s. Thus, the MOV structure more likely forms from ion heating, thus an early indicator of cluster ordering, and corresponds to low but increasing values of cluster coercivity.

V. SUMMARY

We present two early indicators of ordering during direct preparation of $L1_0$ FePt nanoclusters: (1) ion density and (2) MOV structure observations. These findings demonstrate ion density cluster property control during the early stages of direct $L1_0$ FePt nanocluster formation. Ion heating is a controllable mechanism to promote the evolution of FePt clusters toward the $L1_0$ phase.

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