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Full Length Article

The microstructural evolution of chemical disorder and ferromagnetism in He^+ irradiated $FePt_3$ films

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ABSTRACT

This paper investigates the role of ion-induced disorder on the morphology and magnetic properties of chemically ordered $FePt_3$ films. The effects are studied for 15 keV He^+ ions as a function of the ion fluence for 0, 2×10^{16} and 2×10^{17} ions cm^{-2}. Substitutional mixing of the $L1_2$-type Fe-Pt sites takes place within the region of the chemically ordered $FePt_3$ film affected by the irradiation. This accompanies a paramagnetic-to-ferromagnetic transition, as determined by room-temperature magnetometry. Dark-field transmission electron microscopy (TEM) measurements confirm that the 15 keV He^+ ions induce a 120 nm-thick chemically disordered layer into the sub-surface region of the nominally 280 nm-thick ordered $FePt_3$ film. The average domain size *and* the fractional density of the chemically ordered domains within the irradiated $FePt_3$ microstructure are found to mutually decrease with increasing ion fluence. Selected-area electron diffraction results demonstrate that the film's single crystallinity is preserved after irradiation, irrespective of the ion fluence. High-resolution TEM elucidates the coexistence of ordered domains and precipitate disordered domains in the near-surface, low-ion impacted regions of the $FePt_3$ film. Collectively, this work provides detailed insights into the material-science relationship between ion-induced disorder and ferromagnetism in $FePt_3$, as a step towards creating fully customisable, ion-beam-synthesised magnetic nano-elements.

1. Introduction

Bit-patterned media (BPM) is recognised as one of the candidate schemes [1–5] capable of postponing the thermal stability problem of magnetically stored data [6], which arises due to finite-size effects in conventional hard disks. Bit-patterned structures are traditionally fabricated by lithography from continuous ferromagnetic (FM) films into individually addressable, non-interacting, single-domain islands. However, as lithographically engineered structures are susceptible to etch damage, long-range non-uniformity, as well as disturbed crystallinity and topography, alternative nano-fabrication methods are currently being pursued. Increasingly, ion beams – employed to either irradiate, implant or mill template material – are gaining recognition as viable substitutes. Focussed ion irradiations are adept to machine superior, three-dimensional, yet planar, spatial arrays of FM components ideally suited to the tribology of BPM devices [7–9]. While ion implantations could be used to prepare novel logic and spintronic devices [10], or similarly employed to sculpt the conducting edge states of topological insulators [11].

$FePt_3$ is an ideal framework for investigating BPM structures formed by ion irradiation, because it features tailorable antiferromagnetic (AFM) and FM characteristics depending on the degree of long-range chemical order occupied by its constituent atoms [12,13]. For example, purely chemically ordered $FePt_3$ is AFM below $T_N = 160$ K, while purely chemically disordered $FePt_3$ displays ferromagnetism until $T_C = 425$ K. The AFM-to-FM transition is mediated in $FePt_3$ by the conversion of the atomic lattice from a $L1_2$-type arrangement in the chemically ordered state (Fig. 1; top-right inset), to a face-centered-cubic (fcc) ensemble in the chemically disordered state (Fig. 1: bottom-right inset) where each site is represented by a ¼ Fe and ¾ Pt occupancy. Consequently, diffraction from the chemically ordered $FePt_3$ lattice will produce both fundamental (where (*hkl*) = all odd or all even integers) and superstructure (where (*hkl*) = a mixture of odd and even integers) Bragg reflections, whereas the latter reflections will be

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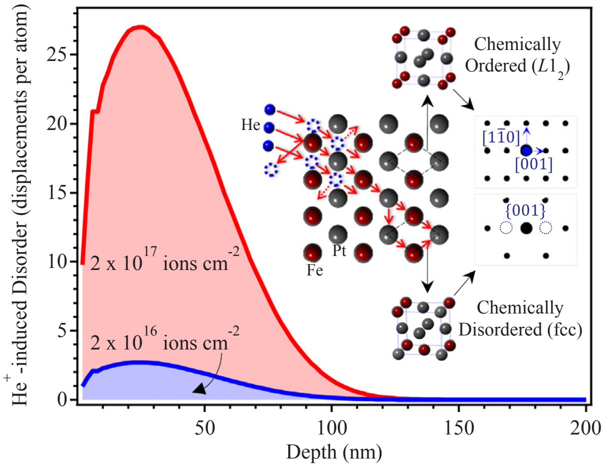

Fig. 1. Simulated disorder profiles for 15 keV He$^+$ ions of 2×10^{16} and 2×10^{17} ions cm^{-2} incident upon a FePt$_3$ target. Inset: Schematic of the expected change in position among the $L1_2$ chemically ordered lattice atoms of FePt$_3$ during low-energy He$^+$ irradiation, showing the formation of ion-induced chemically disordered (fcc) FePt$_3$ domains. The diffraction pattern of each lattice arrangement is correspondingly shown.

forbidden by the chemically disordered fcc lattice. Furthermore, because the order-to-disorder transformation occurs *via* the random-local propagation of disordered domains, the characteristics of any given FePt$_3$ sample will be a mixture of the two aforementioned crystal structures and their respective magnetic properties.

Recently, we exploited the unique quantum chemical feature of FePt$_3$, in tandem with ion-irradiation, to induce lattice atom mixing in a chemically well-ordered FePt$_3$ film. As a result, we crafted a room-temperature *chemically disordered* FM/*chemically ordered* paramagnetic (PM) bilayer which showed evidence of an abrupt magnetic interface [14]. Furthermore, the crystallinity and topography of the film were preserved, and no exotic ion species were implanted into the structure post-irradiation, in line with previous literature [15,16]. While these results are encouraging for ion-beam designed BPM devices, they do not provide a detailed microstructural analysis of FePt$_3$, nor do they explain how the microstructure and magnetic characteristics of FePt$_3$ change as a function of chemical disorder. Therefore, in this article we comparatively investigate the morphology and magnetism of un-irradiated, low- and high-fluence 15 keV He$^+$ irradiated FePt$_3$ films. In our study, we combine transmission electron microscopy (TEM) methods – including dark-field imaging, selected-area electron diffraction, high-resolution TEM and simulation – with magnetometry. Detailed knowledge about the evolution of magnetic, atomic and microstructural parameters upon He$^+$ irradiation of FePt$_3$ is gained, as a prerequisite step towards implementing the ion-beam technique as an effective tool for producing planar patterned magnetic structures suitable for future BPM technologies.

2. Materials and methods

An epitaxial and chemically well-ordered FePt$_3$ film was grown by magnetron sputter deposition in an argon gas (99.999% pure) atmosphere. Sputtering took place from a composite Fe$_{0.25}$Pt$_{0.75}$ target which predetermined the stoichiometric composition of the deposited film. During deposition the (0 0 1)-oriented MgO substrate was heated to 1023 K to promote epitaxial film growth, as well as to obtain the chemically ordered phase of FePt$_3$. *In situ* reflection high-energy electron diffraction was used to monitor the surface epitaxy of the film throughout the deposition process. The nominal thickness of the FePt$_3$ film, estimated from the sputtering rate, is 280 nm. Further details describing the deposition set-up can be found in Lu et al. [17] From X-ray diffraction (XRD) measurement the FePt$_3$ film is found to be (0 0 1) epitaxially oriented. By analysing the integrated intensity ratio of the film's fundamental and superstructure XRD peaks, using formalism described in previous works [17], a film-averaged chemical-order parameter (S) of 0.82 is calculated (where $S = 1$ denotes perfect chemical order). Irradiations were performed at room temperature on the LEII-Surface Engineering beamline of the 6 MV SIRIUS Tandem Accelerator at the Australian Nuclear Science and Technology Organisation (ANSTO) [18]. The FePt$_3$ film was cut into three equal pieces and irradiated with 15 keV He$^+$ ions with fluences of 0 (un-irradiated), 2×10^{16} (low-fluence) and 2×10^{17} (high-fluence) ions cm^{-2}, respectively. Simulations of the disorder profiles generated by 15 keV He$^+$ ions incident upon an amorphous FePt$_3$ target with fluences of 2×10^{16} ions cm^{-2} and 2×10^{17} ions cm^{-2} were performed using Transport of Ions in Matter software [19]. Full-damage cascade simulations using a total of 10^6 He$^+$ ions were performed. The FePt$_3$ number density was set to 6.56×10^{28} atoms m^{-3} ($\approx$5% reduction from bulk density), which was prior determined from fitting the film's critical edge of neutron reflectivity [14]. The magnetic response of each irradiated and un-irradiated film was measured by a vibrating sample magnetometer (VSM) physical properties measurement system (Ever-Cool II by Quantum Design). The films were field-cooled from room temperature to 5 K in a 1 T in-plane magnetic field, and magnetic hysteresis loops were measured at several temperature steps (T = 5, 10, 25, 50, 100, 150, 300 K) upon warming across a ± 20 kOe field range. Magnetisation values (i.e. emu cm^{-3}) were obtained by normalising the measured moment to the film volume. Transmission electron microscopy (TEM) samples were fabricated using a Zeiss Auriga 60 crossbeam focussed-ion beam. TEM images were recorded using a JEOL 2200FS TEM device operating at 200 kV. FePt$_3$ crystal structures were modelled using CrystalMaker™, and selected-area electron diffraction (SAED) simulations were generated using SingleCrystal software [20]. JEMS software was used to carry out high-resolution (HR) TEM image-map simulations of the FePt$_3$ film irradiated with 2×10^{17} He$^+$ ions cm^{-2} [21].

3. Results and discussion

3.1. He$^+$ disorder simulations

Fig. 1 displays the simulated disorder profiles calculated for 15 keV He$^+$ ions of low (2×10^{16} ions cm^{-2}) and high (2×10^{17} ions cm^{-2}) fluence incident upon a FePt$_3$ target. The inset of Fig. 1 models the expected change in position among chemically ordered FePt$_3$ lattice atoms during a low-energy He$^+$ irradiation. The incident He$^+$ ions modify the site occupancy of the chemically ordered $L1_2$-type Fe-Pt nearest neighbours, resulting in the nucleation and growth of chemically disordered domains (which display the conventional fcc structure) throughout the ion-impacted area. Due to the low momentum of the impinging He$^+$ ions, the irradiation leads exclusively to the substitution of Fe and Pt atoms within a few atomic distances of the ion-path, without the formation of interstitial defects [8]. The ions undergo nuclear stopping in the target material where they preferentially migrate to the grain boundaries and later diffuse from the FePt$_3$ lattice.

Regardless of the ion fluence, it is computed that the disorder profiles follow Gaussian distributions with peak centroids located 28 nm beneath the target surface. Increasing the ion fluence by an order of magnitude (from 2×10^{16} to 2×10^{17} ions cm^{-2}), however, results in a comparative enhancement to the disorder of the FePt$_3$ target at each corresponding depth. This trend suggests that disorder can be tailored in FePt$_3$ by modifying the fluence of the irradiating ion. This is because higher ion fluences support a greater probability of interaction between the ion and target atom, giving rise to a greater density of chemical order-to-disorder transformations throughout the FePt$_3$ domain structure.

For a given target material and irradiating ion, it is known that the ion penetration depth is dependent upon the energy of the irradiating