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Fabrication and magnetization reversal of $L1_0$ FeMnPt dots surrounded by paramagnetic $A1$ phase formed by ion irradiation

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ABSTRACT

A nanopatterning method involving a ferromagnetic (FM)–paramagnetic (PM) phase transformation caused by ion irradiation has been developed. A $(Fe_{0.56}Mn_{0.44})_{50}Pt_{50}$ film with the ordered $L1_0$ structure and uniaxial magnetocrystalline anisotropy constant (K_u) of 2.1×10^6 J m^{-3} was fabricated. Mn ion irradiation produced a smooth FM dot array with dot diameters of 100 nm surrounded by the PM phase, which was obtained from the induced FM–PM phase transition caused by the structural transformation from the $L1_0$ structure to the disordered $A1$ structure and a slight increase in the Mn content. The magnetization reversal process was studied by investigating the angle dependence of the dot reversal field.

Recently, nano-magnetic thin films with perpendicular anisotropy have been actively investigated due to their potential applications in high-density magnetic data storage [1,2], spintronic devices [3], and drug delivery [4]. In particular, bit-patterned media (BPM) is a candidate for the creation of ultra-high-density magnetic data storage devices [1,2]. Therefore, a proper understanding of the magnetization reversal process of individual magnetic dots in BPM is important for achieving a lower bit error rate.

Previous studies have shown that continuous magnetic films undergo magnetization reversal via the nucleation of low-anisotropy regions followed by the propagation of domain walls [5,6]. In the ideal case, single-domain dot reversal occurs due to coherent magnetization rotation, where the coercivity of the dots is determined by their magnetic anisotropy [7] and temperature [8,9]. This magnetization reversal process can be analyzed by investigating the angle dependence of the reversal field H_r, which is observed under the action of the magnetic field H applied at angle θ with respect to the magnetic easy axis. While continuous films usually obey the $1 / \cos\theta$ dependence described by the Kondorsky model (indicating domain wall propagation), ideal small dots exhibit a Stoner–Wohlfarth (S–W) dependence with the minimum value of H_r obtained at an angle of 45° [10–12].

Typically, a method involving ion milling, which produces physically separated ferromagnetic (FM) regions, is used for BPM fabrication. After ion milling, backfilling and polishing stages are required to obtain BPM with a smooth surface, which is a prerequisite for read/write heads to be able to fly at a distance of few nanometers above the medium surface. One technique that allows BPM to be fabricated without introducing topological damage uses ion irradiation [13–21], which can replace the ion milling, backfilling, and polishing steps. The resulting BPM consist of multiple phases, which include the FM phase (dots) characterized by a high uniaxial magnetocrystalline anisotropy constant K_u along with the diamagnetic, antiferromagnetic, and paramagnetic (PM) phases. The utilized ion irradiation procedure results in structural disordering and the related FM–PM transformation, which has been previously observed for the irradiated Co/Pt multilayers [13,14] and $L1_0$ $CrPt_3$ films [15]. In these materials, the magnitude of K_u obtained for the FM phase is approximately 10^5 J m^{-3}, which is not sufficient for the realization of high magnetic data storage density of over 2 Tb in^{-2} because the thermal fluctuations of magnetic grains or dots are governed by the magnetic anisotropy energy defined as K_uV (here V is the volume of the isolated magnetic grain or dot). As the dots become smaller, the thermal fluctuation energy equal to k_BT (where k_B is the Boltzmann constant, and T is the temperature) becomes greater than the magnetic anisotropy energy, which allows spontaneous magnetization reversal to occur. The observed thermal fluctuation can be reduced by using materials with high values of K_u, which increase the thermal stability factor ($K_u V / k_B T$) to values >60 (this metric was derived using the Sharrock equation [22]) that is typically considered a minimum requirement for magnetic data storage [2,23].

The face-centered tetragonal $L1_0$ FePt films of the CuAu-I type with lattice constants $a = b > c$ are possible candidates for BPM due to their high values of K_u of 7.0×10^6 J m^{-3} and saturation magnetization M_s of 1.4 Wb m^{-2} [24]. However, it is not easy to modify the magnetic

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properties of FePt films via ion irradiation due to the very low sensitivity of M_s to this process [16–19]. Generally, the high M_s of the inter-dot spaces in the storage media produces noise spikes. In our previous study, substituting Mn for Fe in $L1_0$ FePt films dramatically improved the sensitivity of M_s to ion irradiation [21]. The $L1_0$ $(Fe_{1-x}Mn_x)_{50}Pt_{50}$ films with $x \leq 0.44$ exhibit FM properties corresponding to $K_u > 2.1 \times 10^6$ J m^{-3}, whereas the disordered face-centered cubic (fcc) $A1$ films with $x \geq 0.44$ and $a = b = c$ possess PM properties at room temperature. These films experience a transition from the FM to PM state as the $L1_0$ structure transforms into the $A1$ phase during ion irradiation. In this study, the observed FM–PM phase transformation in FeMnPt films was utilized for the fabrication of a magnetic dot pattern, and the corresponding magnetization reversal process was characterized by magnetometry and magnetic force microscopy (MFM) techniques.

$(Fe_{1-x}Mn_x)_{50}Pt_{50}$ films with a thickness of 3.0 nm were deposited onto single crystalline MgO (100) substrates at a temperature of 298 K via magnetron co-sputtering. The base pressure of the sputtering chamber was 10^{-5} Pa, and an Ar gas pressure of 0.5 Pa was used during deposition. The Mn content x was controlled by varying the sputtering rates for the Fe and Mn targets, while the transformation process was studied using an electron probe X-Ray microanalyzer. Nanoscale composition profiles were obtained using an energy dispersive X-Ray detector (EDX) attached to a transmission electron microscope (TEM).

The produced films were subjected to rapid thermal annealing (RTA) at a heating rate of 300 K s^{-1} with a background pressure of 2×10^{-4} Pa to promote both epitaxial growth and atomic ordering. The resulting crystalline structure was investigated by out-of-plane X-ray diffraction (XRD) measurements with CuKα radiation ($\lambda = 1.540593$ Å). A vibrating sample magnetometer (VSM) with a maximum field ($\mu_0 H$, where μ_0 is the vacuum permeability) of 1.8 T and a superconducting quantum interference device (SQUID) magnetometer with a maximum field of 5.0 T were used to evaluate the magnetic properties of the films. The surface topography and magnetic domain structure were investigated via atomic force microscopy (AFM) and MFM, respectively. The magnetization curves of the dot pattern sample were obtained by recording the corresponding element-specific Pt L_3-edge (11.566 keV) X-ray magnetic circular dichroism (XMCD) hysteresis curves using the BL39XU beamline located at the SPring-8 synchrotron facility with an X-ray spot diameter of 100 μm and maximum field of 10.0 T applied at angle θ to the normal of the film plane.

A positive-type resist (ZEP520A, ZEON Corp., Japan) was used for the film nanopatterning via electron beam lithography. First, a positive resist layer with a thickness of 50 nm was spin-coated onto the [001]-oriented FM $L1_0$ $(Fe_{0.56}Mn_{0.44})_{50}Pt_{50}$ film. A square dot array was created via electron beam lithography using a resist mask with side length and spacing of 100 nm, which was subsequently irradiated with Mn ions and then removed. The total area of the dot pattern produced via ion irradiation was approximately 120 μm × 120 μm. The irradiation energy was 4 keV, and the dose was 3.0×10^{15} ions cm^{-2}. The base vacuum pressure prior to irradiation was approximately 10^{-5} Pa. A water-cooled metal sample holder was used to mitigate the heating effect observed during irradiation, maintaining a temperature close to 298 K.

Prior to nanopatterning, the crystal structure and magnetic properties of the sputtered $(Fe_{0.56}Mn_{0.44})_{50}Pt_{50}$ continuous films were evaluated. Fig. 1(a) shows their out-of-plane XRD patterns obtained before and after ion irradiation. The unirradiated film previously subjected to RTA at a temperature of 973 K for 10 min exhibited only the superlattice (001) and fundamental (002) reflection peaks, but not the fundamental (111) reflection, indicating that the (001) crystalline texture was oriented perpendicularly to the film plane, which corresponded to the ordered $L1_0$ structure (the estimated degree of the long-range chemical order determined from the ratio between the (001) and (002) peak intensities was approximately 0.90 [25]). The films irradiated with Mn ions exhibited the fundamental (200) reflection peaks without the superlattice (001) reflections, which suggested that the (200) crystalline texture was oriented perpendicularly to the film plane, corresponding to the disordered $A1$ (fcc) structure. Fig. 1(b) and (c) show the magnetization curves obtained from VSM measurements for the unirradiated and irradiated films, respectively, at a temperature of 298 K with the field H applied perpendicularly (⊥, solid line) and parallel (∥, dashed line) to the film plane. The magnetization easy axis of the unirradiated film (Fig. 1(b)) was perpendicular to the film plane characterized by the magnitude of M_s equal to 0.75 Wb m^{-2}, coercivity of 0.65 T, and K_u of 2.1×10^6 J m^{-3}. The value of K_u was evaluated using the relation $K_u = (M_s \times H_k / 2) + K_{shape}$, where H_k was the magnetic anisotropy field, and K_{shape} was the shape anisotropy calculated from the demagnetization factors N of the film shape ($N_\perp = 1, N_\parallel = 0$). H_k was determined from the intersection point of the extrapolated parallel hysteresis loop with the perpendicular loop, which yielded a value of $H_k = 5.0$ T perpendicular to the film plane. Using a thermal stability factor of $(K_u V / k_B T) > 60$, the minimum diameter of spherical dots estimated for this film was 6.1 nm, indicating that the system had sufficient thermal stability to be used for magnetic storage at densities of around 10 Tb in^{-2}. After ion irradiation, the films exhibited essentially PM properties (Fig. 1(c)) with a maximum saturation magnetization of around 0.12 Wb m^{-2}. The Mn ion irradiation produced two effects that decreased the spontaneous magnetization (M_0): disordering and a slight increase in the Mn content. In our previous study, the magnitude of M_0 measured for the disordered $A1$ $(Fe_{0.56}Mn_{0.44})_{50}Pt_{50}$ film was almost zero, while its Curie temperature was below the room temperature [21]. As a result, a phase transformation was successfully induced, and the FM properties were replaced with PM properties.

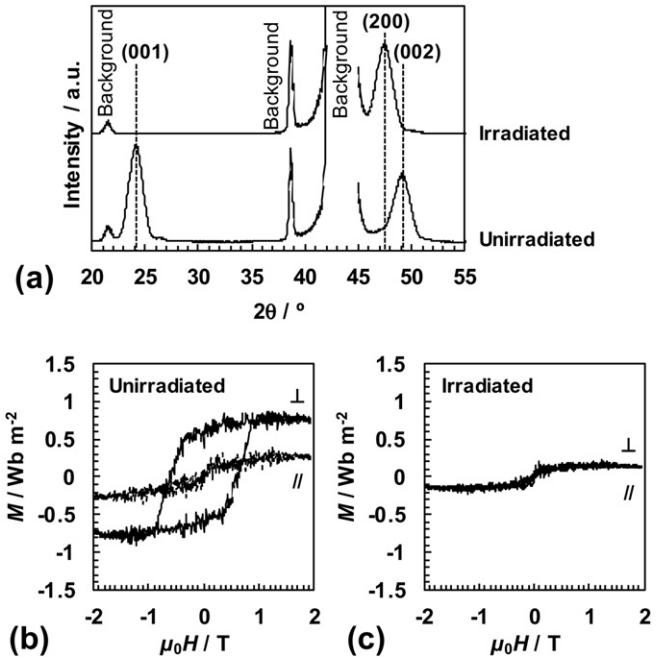

Fig. 1. A 3.0 nm thick $L1_0$ $(Fe_{0.56}Mn_{0.44})_{50}Pt_{50}$ film was irradiated with Mn ions at an energy of 4 keV and dose of 3.0×10^{15} ions cm^{-2}. (a) Out-of-plane XRD patterns obtained for the studied film before (unirradiated) and after (irradiated) ion irradiation. Magnetization curves (⊥: perpendicular, ∥: parallel) recorded at a temperature of 298 K (b) before and (c) after ion irradiation.

Fig. 2(a) schematically illustrates the nanopatterning process. The resist mask thickness was 50 nm, which was sufficient for preventing Mn ions from penetrating the FeMnPt films during irradiation (it was assumed that after nanopatterning followed by Mn ion irradiation, only the spaces between the dots underwent the structural transformation from the FM $L1_0$ to the PM $A1$ phase, whereas the dots themselves retained the $L1_0$ structure with the FM properties). Fig. 2(b) and (c) show the cross-sectional TEM image and lateral Mn distribution,