Accelerating disorder–order transitions of FePt by preforming a metastable AgPt phase

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Abstract

Many approaches had been reported to successfully reduce the transition temperature of FePt from A1 to L10 phase, though without detailed knowledge. In this work, we deposited the metastable AgPt layer adjacent to the Fe layer and addressed the importance of vacancies in the disorder–order transition of FePt at reduced temperatures on the basis of a kinetic diffusion model. The decomposition of the metastable AgPt phase, creating excess vacancies during the post-deposition annealing process, accelerated the intermixing between Fe and Pt and the nucleation of L10 FePt. The evolution of phase transformation from AgPt–Fe to L10 FePt–Ag was monitored by in situ high temperature X-ray diffractometry and was also validated by first-principles calculations. The intermixing between Fe and Pt and the nucleation of L10 FePt after annealing at 230 °C were directly observed by transmission electron microscopy and grazing incidence X-ray diffractometry, respectively. With the assistance of the decomposition of AgPt, we obtained a (001)-dominated L10 FePt film with an out-of-plane coercivity as large as 13.3 kOe after annealing at a temperature as low as 350 °C. The principles of the proposed method can be applied for versatile disorder–order phase transitions.

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1. Introduction

The rearrangement of atoms through a disorder–order transition causes a superstructure to be formed that significantly alters the properties of a material, in particular the magnetic properties. The L10 FePt shows extraordinarily large magnetocrystalline anisotropy (MCA), which makes it a promising material for ultrahigh-density perpendicular magnetic recording media. Such large MCA is attributed to the strong spin–orbit coupling in Pt and 5d Pt–3d Fe hybridization through the alternate repetition of atomic Fe and Pt lamellas in a stack [1]. However, the nucleation of ordered crystallites requires atoms with greater energy than is required for activation so that they can move to the right positions to form the superstructure. Accordingly, to achieve the L10 FePt, a high-temperature annealing treatment after deposition or a heated substrate during deposition is typically required, which may induce undesirable interlayer diffusion and grain growth.

Many approaches had been reported to reduce the ordering temperature required for L10 FePt (typically higher than 500 °C) from various aspects. The introduction of in-plane tensile stress had been widely reported to reduce the ordering temperature of FePt. The stress was induced by inserting an underlayer (Ag, PtMn, CrRu and MgO) [2–5]. In addition to the static stress originating from the lattice mismatch between FePt films and underlayers, stress was also introduced dynamically. Formation of a Cu3Si phase by Cu and Si underlayers caused dynamic in-plane tensile stress due to the abrupt expansion in volume caused...
by the formation of Cu₃Si at a temperature below 300 °C, which thus induced the ordering of FePt at 300 °C [6]. Low-temperature ordering was also achieved by coherent ordering between an AuCu or PtMn underlayer and a FePt film because of the lower ordering temperature of the underlayer [3,7]. Accelerated diffusion with the aid of vacancies was also effective in reducing the ordering temperature. He-ion irradiation on FePt films was demonstrated to directly transfer energy to as well as introducing point defects in the FePt lattice; therefore, the diffusion was significantly enhanced and the formation of the ordered phase was achieved after the irradiation [8]. A multilayer structure of [Fe/Pt], was proposed to enhance the diffusion at the Fe/Pt interface [9]. Furthermore, the alternate monatomic layer deposition of Fe and Pt was manipulated to prepare L₁₀-ordered FePt alloy films at reduced temperatures [10]. The addition of a third element (Au and Ag) with high mobility was shown to be quite effective in reducing the ordering temperature by increasing the mobility of Fe and Pt atoms during the heating process [11]. Cu was added to form a FePt/Cu ternary ordered alloy, in which the Cu atoms was located in the Fe sites of the FePt ordered alloy, and this ordering occurred at reduced temperatures [12]. Furthermore, the addition of Ag has been demonstrated not only to reduce the ordering temperature of FePt, but also to be applicable for thermally assisted recording media [13,14]. Various approaches for adding Ag to the FePt system have been reported to reduce the ordering temperature: Ag was incorporated into FePt films through co-sputtering [11,15], insertion of an ultrathin layer [16] or diffusion from an underlayer or top layer [2,17]. These works claimed that the rapid movements of Ag atoms enhanced the mobility of Fe and Pt atoms and kinetically lowered the ordering temperature of FePt. However, according to Rellinghaus et al. [18], the mean diffusion length for the volume diffusion of Pt in FePt at 530 °C for 1 h is only 0.3 nm, and this vanishes exponentially down to 350 °C.

In this work, we take into account the extremely low diffusivity of Pt and propose a new method to reduce the disorder–order transition temperature by preforming a metastable AgPt phase adjacent to the Fe layer. We assume that the metastable AgPt phase decomposes at low temperatures, so that Ag atoms leave the AgPt layer and exchange their positions with Fe atoms. The mechanism improves the Fe diffusivity and, by creating vacancies inside the AgPt layer, accelerates the low-temperature ordering of L₁₀ FePt. Here, we prove the proposed mechanism by both theoretical (including equilibrium thermodynamics and kinetics) and experimental means.

### 2. Theory/calculation

The L₁₁ and L₁₀ structures have been observed experimentally and confirmed as ground states by ab-initio calculations in the Ag–Pt and Fe–Pt binary systems, respectively [19–22]. First-principles calculations allow us to verify which of two possible phase separations, Fe – AgPt₉ or Ag – FePt₉, is the most thermodynamically stable in an equiatomic AgFePt. Let us consider the enthalpies (per atom) of a phase separation of pure Fe with L₁₁–AgPt:

\[
H^\text{at}(\text{Fe} – \text{AgPt}_{L_{11}}) = x_{\text{Fe}}E_{\text{Fe}}^{\text{at}} + 2x_{\text{Pt}}E_{\text{AgPt}_{L_{11}}}^{\text{at}}
\]

and pure Ag with L₁₀–FePt:

\[
H^\text{at}(\text{Ag} – \text{FePt}_{L_{10}}) = x_{\text{Ag}}E_{\text{Ag}}^{\text{at}} + 2x_{\text{Pt}}E_{\text{FePt}_{L_{10}}}^{\text{at}}
\]

In both Eqs. (1) and (2), \(x_A(\text{A} = \text{Ag}, \text{Fe}, \text{and Pt})\) are the total compositions and \(E^{\text{at}}\) are the energies (per atom) of the corresponding pure solids and binary compounds. The difference between these two enthalpies at the equiatomic composition C:

\[
C = \text{AgFePt}(x_{\text{Ag}} = x_{\text{Fe}} = x_{\text{Pt}})
\]

is:

\[
\Delta H_C \equiv H^\text{at}(\text{Fe} – \text{AgPt}_{L_{11}}) – H^\text{at}(\text{Ag} – \text{FePt}_{L_{10}}) = \frac{2}{3} \left( \Delta H_{\text{AgPt}_{L_{11}}}^{\text{Ag}} - \Delta H_{\text{FePt}_{L_{10}}}^{\text{Fe}} \right)
\]

where \(\Delta H_{\text{AgPt}_{L_{11}}}^{\text{Ag}}\) and \(\Delta H_{\text{FePt}_{L_{10}}}^{\text{Fe}}\) are the formation enthalpies of the binary compounds:

\[
\begin{align*}
\Delta H_{\text{AgPt}_{L_{11}}}^{\text{Ag}} &= E_{\text{AgPt}_{L_{11}}}^{\text{at}} - 0.5(E_{\text{Ag}}^{\text{at}} + E_{\text{Pt}}^{\text{at}}) \\
\Delta H_{\text{FePt}_{L_{10}}}^{\text{Fe}} &= E_{\text{FePt}_{L_{10}}}^{\text{at}} - 0.5(E_{\text{Fe}}^{\text{at}} + E_{\text{Pt}}^{\text{at}})
\end{align*}
\]

We calculated \(\Delta H_{\text{AgPt}_{L_{11}}}^{\text{Ag}}\) and \(\Delta H_{\text{FePt}_{L_{10}}}^{\text{Fe}}\) from first principles, and these are listed in Table 1. Note that we consider the formation enthalpies as referring to both body-centered cubic (bcc) and face-centered cubic (fcc) Fe because the low-temperature bcc Fe transforms into fcc at high temperatures [19]. The two corresponding enthalpies may describe the low- and high-temperature regions of the phase diagram in an approximation of zero magnetic entropy. The values of \(\Delta H_C\) obtained by substituting the calculated \(\Delta H_{\text{AgPt}_{L_{11}}}^{\text{Ag}}\) and \(\Delta H_{\text{FePt}_{L_{10}}}^{\text{Fe}}\) into Eq. (4) are 131 and 181 (meV), with respect to bcc and fcc Fe, respectively. The strongly positive values of \(\Delta H_C\) establish that the phase separation into pure Ag plus L₁₀–FePt is more thermodynamically stable than pure Fe plus L₁₁–AgPt in equiatomic AgFePt. This tendency should be even stronger at higher temperatures (in an approximation of zero magnetic entropy), as \(\Delta H_C\) is larger with reference to fcc Fe than to bcc Fe.

On the basis of the formation enthalpies of all known ground state compounds in the Fe–Pt, Ag–Pt and Fe–Ag binary systems calculated from first-principles listed in Table 1, we constructed the tentative ternary phase diagram of Ag–Fe–Pt shown in Fig. 1. We used the generalized gradient approximation with projector augmented wave pseudopotentials [23], as implemented in the VASP package [24], at zero temperature and without zero-point motion. The calculations are performed within the high-throughput computational framework AFLow [25]. The effect of lattice vibrations is omitted. Numerical convergence to within about ~1 meV atom⁻¹ is ensured.
of Fe (2.67 nm)/Ag 35Pt65 (3.61 nm)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1

<table>
<thead>
<tr>
<th>x_i</th>
<th>Structure</th>
<th>( \Delta H_{X}^{at} ) (meV atom(^{-1}))</th>
<th>( \Delta H_{Fe-fcc}^{at} ) (meV atom(^{-1}))</th>
</tr>
</thead>
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<tr>
<td>Fe</td>
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<td>152</td>
<td>0</td>
</tr>
<tr>
<td>Fe</td>
<td>bcc</td>
<td>0</td>
<td>-152</td>
</tr>
<tr>
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<td>-44</td>
</tr>
<tr>
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<td>-193</td>
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<tr>
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<td>L1_0</td>
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<td>-316</td>
</tr>
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<td>-237</td>
</tr>
<tr>
<td>FePt</td>
<td>L1_1-AM</td>
<td>-216</td>
<td>-254</td>
</tr>
<tr>
<td>FePt</td>
<td>( \beta )</td>
<td>-226</td>
<td>-277</td>
</tr>
<tr>
<td>FePt</td>
<td>Pt6Ti</td>
<td>-122</td>
<td>-139</td>
</tr>
</tbody>
</table>

Fig. 1. The ternary phase diagram of Ag–Fe–Pt built by using of ab-initio formation enthalpies of all known ground state compounds in Fe–Pt, Ag–Pt and Fe–Ag listed in Table 1. The central point C of the triangle denotes the equiatomic AgFePt. The dashed blue line and corresponding L1_2–FePt compound also appear in the high-temperature phase diagram, where fcc Fe (rather than bcc Fe) is taken as the reference state. The red star denotes the experimentally studied ternary Ag_{53}Fe_{45}Pt_{12} (reflecting the bilayer film of Fe (2.67 nm)/AgPt (3.61 nm)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

by enforcing a high energy cut-off (350 eV), and dense Monkhorst–Pack \( k \)-point meshes, equivalent to a \((12 \times 12 \times 12)\) mesh, for cubic conventional unit cells. All structures are fully relaxed (cell shape/volume, collinear spins and atom positions).

Because the theoretical model is based on ab-initio calculations at zero temperature, the free energy contributions from configurational (substitutional), phonon and magnetic entropies may become significant at higher temperatures. In particular, high-temperature phase transition of Fe is a consequence of interplay between phonon and magnetic effects at elevated temperatures [26]. We introduce this approximate transformation into our model by replacing bcc Fe with fcc Fe as a reference state for formation enthalpies. Further development of the model should also include the possibility of forming Ag–Fe–Pt ternary compounds.

Based on the ternary phase diagram of Ag–Fe–Pt shown in Fig. 1, we experimentally designed the bilayer structure (Fe/AgPt) with the composition marked by a red star in Fig. 1, at which the equilibrium configuration consists mostly of decomposed pure Ag and L1_0 FePt. In fact, Ag has been reported in experimental [10] and theoretical works [27,28] to have low solubility in L1_0–FePt. Using the sputtering technique, we can further fabricate disordered metastable AgPt as the initial state. This disordered state makes the occurrence of the phase transformation even more thermodynamically favorable due to its higher formation energy compared to the L1_1 AgPt.

In addition to the thermodynamic validity, we apply a simple diffusion model to construct the kinetic process, which represents the phase transformation mechanism as illustrated in Fig. 2. We consider the high and extremely low mobilities of Ag and Pt, respectively. For the bilayer film composed of Fe/AgPt, the AgPt layer serves as a platform for exchanging Ag and Fe during post-annealing: Ag is expelled and segregates, while Fe is included to form FePt. After a series of position exchanges, an intermixed FePt is achieved even though the Pt atoms have never left their original sites. Based on a kinetic diffusion model [29], the self-diffusivity \( D \) through the vacancy mechanism can be expressed as: \( D = \frac{1}{V} \cdot \Gamma_{V} \cdot \alpha \cdot \ell_{V} \cdot \Delta E_{V} \), where \( \Gamma_{V} \) is the jump frequency, \( a \) is the lattice constant, \( f \) is the correlation factor close to unity and \( V \) is the number of fcc nearest neighbors. At 350 °C, \( D \) (Ag) = 2.1 \times 10^{-10} and \( D \) (Fe) = 3.0 \times 10^{-18} [30,31], whereas \( D \) (Pt) = 1.3 \times 10^{-23} (all in cm\(^2\) s\(^{-1}\)) as extrapolated from the experimental data \( D = 0.05 \exp(-2.67eV/kT) \) cm\(^2\) s\(^{-1}\) [32]. The high self-diffusivity of Ag indicates a much higher jump frequency, and thus a shorter average migration time. Consequently, Ag atoms move out of the AgPt layer much faster than the Fe atoms move in, leading to the formation of vacancies inside the remained Pt layer. The vacancies created in the AgPt layer promote Fe diffusion, which accelerates the Fe–Pt intermixing and the ordering process. The created vacancies may also slightly adjust the positions of Pt atoms to complete the disorder–order transitions. Based on our ab-initio calculations and self-diffusivity models, we conclude that L1_0 FePt–Ag is more thermodynamically stable than AgPt–Fe and the fast diffusion of Ag creates excess vacancies in the AgPt layer, accelerating the ordering of L1_0 FePt.

3. Experimental details

Bilayer films of Fe (2.67 nm)/Pt (2.34 nm) and Fe (2.67 nm)/AgPt (3.61 nm) deposited onto thermally oxidized Si (001) substrates were prepared at ambient temperature under a working pressure of 3 mtorr by a DC
magnetron-controlled sputtering system; the base pressure was $1 \times 10^{-7}$ torr. The layer of AgPt was co-sputtered from Pt and Ag elemental targets, and was a metastable phase in a wide composition range. The resultant composition of Fe$_{55}$Pt$_{45}$ and Ag$_{35}$Pt$_{65}$ was fixed for all films, confirmed by inductively coupled plasma mass spectrometry. The film thickness was estimated with the aid of atomic force microscopy. After film depositions, an annealing process was carried out in a vacuumed furnace ($1 \times 10^{-6}$ torr) at 230 °C for 2 h. The film structure was characterized by X-ray diffractometry (XRD): θ–2θ scans XRD, grazing incidence XRD (GIXRD) and in situ high temperature XRD (HTXRD). All X-ray sources were Cu $K_\alpha$-radiation. The magnetic properties were measured with a vibration sample magnetometer (VSM) and superconducting quantum interference device (SQUID) with applied fields of 7 T. The film microstructure was analyzed by transmission electron microscopy (TEM).

4. Results and discussion

To experimentally realize the proposed mechanism, we first fabricated the metastable AgPt phase. Although Ag and Pt are immiscible at the designated composition (Ag$_{35}$Pt$_{65}$) at low temperatures according to the Ag–Pt binary phase diagram [19], they can be deliberately alloyed together in thin-film form by the sputtering technique, inspired by the work of Ma [33].

The GIXRD pattern of the relaxed co-sputtered Ag$_{35}$Pt$_{65}$ (3.61 nm) film is shown in Fig. 3; three diffraction peaks, which shift coherently to lower angles with reference to those of pure Pt, are denoted as AgPt (111), AgPt (200) and AgPt (220). This Ag$_{35}$Pt$_{65}$ phase is verified to be an fcc structure with a lattice parameter larger than that of Pt. It is worth noting that the composition ratio of Ag:Pt is not equiatomic as used in first-principles calculations. The off-equatomic composition of Ag$_{35}$Pt$_{65}$ allows us to fabricate a metastable phase instead of the stable $L1_1$ AgPt phase, which makes the proposed phase transition more thermodynamically favorable. We also chose a ratio of Ag/Pt smaller than 1 because a large Ag content in FePt promotes (111)-oriented $L1_0$ FePt(Ag). A smaller, insufficient Ag content does not affect the tendency of the phase transition from Fe–AgPt to Ag–FePt$^{L1_0}$ because $L1_1$ AgPt is the only stable compound in both the Fe–Ag and Ag–Pt binary systems.

To verify the phase stability of the Ag$_{65}$Pt$_{35}$ film, we annealed the film at the elevated temperature for 1 h. The resultant XRD patterns, shown in Fig. 4, reveal that the sputtered AgPt film possesses good stability even after annealing at 400 °C. The peak shift in the pattern obtained from 400 °C annealed films compared to that of the as-deposited one may result from the strain relaxation. The sputtered AgPt film decomposes after annealing at 700 °C, as evidenced by the peak splitting. Although the single AgPt film can maintain its fcc structure even at
400 °C, when the Fe layer is deposited adjacent to the AgPt layer, the phase stability of AgPt is altered.

Fig. 5 shows the XRD 0–20 patterns of as-deposited [Fe/AgPt]_{x} films, where x = 1, 2, 4 and 8; the total thickness is fixed at 13 nm to all films. The AgPt (111) peak vanishes with increasing number of interfaces of Fe/AgPt, and is accompanied by the occurrence of the disordered FePt (111) peak. This result coincides well with the much lower formation energy of FePt than of AgPt. Consequently, the interfacial bonding between Fe and Pt is easily formed and the metastable AgPt phase is no longer stable even at room temperature.

To validate how the presence of the metastable AgPt accelerates the ordering of FePt, we fabricated bilayer films composed of Fe (2.67 nm)/Ag_{35}Pt_{65} (3.61 nm) and Fe (2.67 nm)/Pt (2.34 nm) for comparison. Both samples were annealed at 230 °C in a vacuumed furnace for 2 h. Fig. 6(a) shows the out-of-plane hysteresis loops of the annealed Fe/Ag_{35}Pt_{65} and Fe/Pt films. The out-of-plane coercive fields \(H_{c,\perp}\) of annealed Fe/Ag_{35}Pt_{65} and Fe/Pt films are around 750 and 180 Oe, respectively. The GIXRD patterns, shown in Fig. 6(b), reveal the presence of the \(L_{10}\) FePt phase, confirmed by the existence of the FePt (001) peak, in the annealed sample of Fe/Ag_{35}Pt_{65}; the larger \(H_{c,\perp}\) obtained from annealed Fe/AgPt film is due to a fraction of \(L_{10}\) FePt nucleated. Both the increase in \(H_{c,\perp}\) and the nucleation of \(L_{10}\) FePt obtained by annealing the Fe/Ag_{35}Pt_{65} bilayer indicate that the metastable AgPt plays an important role in reducing the ordering temperature of FePt.

To further verify the function of the metastable AgPt phase, cross-sectional TEM (XTEM) images of the as-deposited and annealed films were obtained to...
investigate the microstructure evolution, as shown in Fig. 7 (a–d). Prior to the annealing treatment, both as-deposited Fe/Pt and Fe/AgPt films are composed of two distinct layers. After the annealing treatment, the Fe/Pt is still composed of two distinct layers but the interface is shifted slightly upwards, suggesting that the A1 FePt formation occurs in the vicinity of the interface. In contrast, the interface of Fe/AgPt disappears after annealing and Fe/AgPt becomes a single layer, the Fe and Pt intermixing throughout the whole film, together with the nucleation of a fraction of L10 FePt, consistent with our VSM and GIXRD results. Here, we have successfully lowered the nucleation temperature of L10 FePt down to 230 °C with the aid of a preformed metastable AgPt phase.

To understand how the L10 phase of FePt is formed with the aid of the Ag35Pt65 layer during post-annealing, we monitored the structure transition of the Fe/AgPt film in situ by HTXRD. The HTXRD patterns were acquired at room temperature and at 100, 200 and 300 °C, and are shown in Fig. 8. The patterns obtained at room temperature and 100 °C are composed of a single peak denoted as AgPt (111). This peak vanishes at 200 °C with the emergence of an asymmetric peak, which is a combination of AgPt (111) and A1 FePt (111) peaks. In other words, the AgPt phase is stable at 100 °C but decomposes at 200 °C, accompanied by the formation of disordered FePt. With further elevation of the temperature to 300 °C and above, an FePt (001) peak appears, indicating the presence of the L10 phase. This result is consistent with our findings for films annealed at 230 °C; the ordering process does not take place until the temperature reaches 230 °C and proceeds at higher temperatures. These results are explained well by our ab-initio calculations and by the kinetic self-diffusion model described in Section 2: the phase diagram shown in Fig. 1 indicates the thermodynamic driving force for phase transformation from AgPt–Fe to L10 FePt–Ag and the in-plane tensile stress induced by RTA [35]. We therefore changed the bilayer structure of Fe/AgPt to multilayers to improve magnetic properties. [Fe (0.45 nm)/AgPt (0.60 nm)]6 multilayers with the same total thickness as the Fe/AgPt bilayer were fabricated. After annealing at temperatures as low as 350 °C by RTA (at a ramping rate of 35 °C s⁻¹) for 1 min, the FePt (001)-dominated structure was obtained and Ag (or Ag with limited Pt alloy) segregation was evidenced by the presence of an Ag (111) peak, as shown in Fig. 9(a). An \( H_{c,\perp} \) of 13.3 kOe and an \( M_r/M_s \) of 0.92 were obtained from the hysteresis loop in Fig. 9(b); these values are much higher than those (\( H_{c,\perp} = 6.9 \) kOe) observed in the samples of Fe/Pt multilayers annealed under the same annealing conditions [34].

5. Conclusions

The ordering process of FePt is significantly improved by preforming a metastable phase of AgPt. We demonstrate the consistency between experiments and theoretical models (first-principles calculations and a diffusion model) of transformation from Fe–AgPt to L10 FePt–Ag. XTEM images, HTXRD patterns and VSM results suggest that the AgPt phase decomposes at low temperature, which leads to
the nucleation of $L_{10}$ FePt occurring around 230 °C. Based on this concept, we fabricated a (001)-dominated $L_{10}$ FePt:Ag film with $H_{cH}$ equal to 13.3 kOe after a 350 °C annealing process. A well-developed underlayer, e.g. of MgO, can further improve all the magnetic properties. We also observe the segregation of Ag from the XRD pattern. Because of the plasmon resonance heating effect, Ag segregation in FePt films may be potentially beneficial for the applications of heat-assisted magnetic recording media [36]. The principles of the proposed mechanism can be applied for versatile order–disorder phase transformations, especially for nanoscale thin films.

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