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Abstract: FePt and FePtN thin films have been prepared on silicon substrates by the relatively new deposition technique known as High Target Utilisation Sputtering. Films were annealed post-deposition at temperatures up to 8000C in order to induce the high-anisotropy L10 phase. The FePt films initially showed an improvement in magnetic properties with annealing temperature, but for annealing above around 400 ◦C the magnetic properties deteriorated markedly. The magnetic properties of the FePtN films, however, continued to improve with increasing annealing temperature, right up to the maximum temperature applied of 800 ◦C. Analysis by x-ray diffraction revealed the formation of iron and platinum silicides in FePt films above 400 ◦C, but that such silicides are absent from the FePtN at all annealing temperatures except 800 ◦C. This behaviour is attributed to the nitrogen in FePtN films reacting preferentially with the silicon in the substrate to form silicon nitride, thus suppressing the formation of platinum and iron silicides. Thus, by introducing nitrogen during the deposition of FePt films on Si substrates th formation of deleterious silicides appears to be suppressed during thermal treatment, thereby offering protection against silicon pollution.

Suppressed silicide formation in FePt thin films by nitrogen addition

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Abstract

FePt and FePtN thin films have been prepared on silicon substrates by the relatively new deposition technique known as High Target Utilisation Sputtering. Films were annealed post-deposition at temperatures up to 800 °C in order to induce the high-anisotropy $L1_0$ phase. The FePt films initially showed an improvement in magnetic properties with annealing temperature, but for annealing above around 400 °C the magnetic properties deteriorated markedly. The magnetic properties of the FePtN films, however, continued to improve with increasing annealing temperature, right up to the maximum temperature applied of 800 °C. Analysis by x-ray diffraction revealed the formation of iron and platinum silicides in FePt films above 400 °C, but that such silicides are absent from the FePtN at all annealing temperatures except 800 °C. This behaviour is attributed to the nitrogen in FePtN films reacting preferentially with the silicon in the substrate to form silicon nitride, thus suppressing the formation of platinum and iron silicides. Thus, by introducing nitrogen during the deposition of FePt films on Si substrates the formation of deleterious silicides appears to be suppressed during thermal treatment, thereby offering protection against silicon pollution.

Keywords:
FePt thin films, FePtN thin films, Silicidation, HiTUS

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1. Introduction

FePt thin films have attracted considerable attention for their potential as high density magnetic recording materials. The $L1_0$ ordered phase of FePt alloy is one of the most promising candidates for perpendicular magnetic recording beyond 1 Tbit/in^2 because of its high magnetocrystalline anisotropy constant K_u of about $10^7 erg/cm^3$ [1] which leads to a high thermal stability against superparamagnetic effects [2]. However, as-deposited films fabricated by sputtering are of the disordered A1 phase. To obtain the $L1_0$ phase, post- deposition annealing above 500 °C is usually required (alternatively high deposition temperatures may be used [3]). High temperature processes are not however attractive for industrial production processes, and high temperature treatments also typically lead to both grain size and exchange coupling increases [4] which are unfavourable for obtaining high recording density and high signal-to-noise ratios. It is therefore important to examine ways in which the A1 to $L1_0$ transformation temperature might be reduced, and also ways in which inter-granular coupling might be alleviated.

Some approaches already investigated to reduce the ordering temperature have included the addition of additional materials to FePt (e.g. Cu [5] [6] , CrRu [7] and Al [8]), the use of underlayers (e.g. Bi [9], MgO [10] and Ag [11]) and the multilayering of Fe and Pt [12] [13]. It has also been shown that grain growth can be suppressed during heat treatment by isolating magnetic grains in a nonmagnetic matrix [14] [15] such as SiO_2 [16], Al_2O_3 [17], B_2O_3 [18], Si_3N_4 [19]. In this work we examine the efficacy of a novel thin film sputter deposition technique (the so-called High Target Utilisation Sputtering (HiTUS) approach) for the preparation of FePt alloy thin films, in particular when combined with an approach that incorporates nitrogen via the reactive sputtering of FePtN.

The HiTUS sputtering technique was originally developed by Thwaites et al [20] [21] and uses a remotely generated plasma ‘injected’ into the sputter chamber, rather than relying on a plasma generated directly above the sputter target. By this approach the ‘racetrack’ target erosion encountered in conventional magnetron sputtering is avoided, so enhancing target life and utilisation. In addition, it has been suggested that the remote plasma aids with densification of the growing thin film, avoiding process gas inclusion and poor intergrain boundaries or voids [20] [22]. Furthermore, the production of high-anisotropy FePt films by the incorporation of nitrogen via a FePtN starting composition may have beneficial effects; for example Wang et al found that introducing an appropriate amount of nitrogen during sputtering enhances the magnetic properties of $L1_0$ FePt films [23], while Newman et al [24] [25] found that high-anisotropy $L1_0$ CoPt thin films could be successfully produced by thermal annealing of as-deposited CoPtN material. In this paper we show that this method can also be used within the HiTUS system to produce FePt thin films and, importantly, that nitrogen introduced during sputtering appears to suppress the formation of Fe and Pt silicides during post-deposition thermal annealing, and the consequent deleterious effect on magnetic properties that such silicide formation introduces.

2. Experimental

Using the HiTUS system FePt samples of 25 nm thickness were sputtered directly onto naturally oxidised silicon wafers at ambient temperature. The base pressure of the sputter chamber was 1.0 x 10^{-6} mbar. The sputtering pressure remained at 2.2 x 10^{-3} mbar after introducing high purity argon gas (99.995 %). The RF power source was fixed at 1.10 kW and target voltage at 620 V. The deposition rate of FePt was 2.63 Å s^{-1}. For the FePtN samples, 10 sccm of nitrogen was introduced before deposition. The sputtering pressure remained at 2.2 x 10^{-3} mbar after the flow of nitrogen and argon had stabilised. The RF power source was fixed at 1.10 kW and target voltage at 620 V. The deposition rate of FePtN was 2.69 Å s^{-1}.

Following deposition, the samples were annealed in a sealed quartz tube in an argon/hydrogen mixed gas. The samples were annealed at various temperatures between 300 °C and 800 °C for 60 minutes. The different phases of the film were characterised by x-ray diffractometry (XRD) with $Cu - K\alpha$ radiation of wavelength 1.54060 Å. The magnetic properties were studied with vibrating sample magnetometry (VSM) at room temperature with maximum applied field of 20 kOe.

3. Results and discussion

The x-ray diffraction patterns for the as-deposited FePt samples and those annealed at different temperatures are shown in Fig. 1. The peaks in as-deposited films match those of $FePt_3$ of which the main $FePt_3$ (111) peak occurs at 40.3°. As the films are annealed through to 350°C, this peak is shifted slightly and becomes matched to FePt (111). This indicates a small loss in Pt during annealing up to this temperature. What were clearly the main peaks (for $FePt_3$ $2\theta = 40.3°$ and $46.9°$; for FePt $2\theta = 40.6°$ and $47.2°$) gradually become smaller as annealing temperatures increase and eventually disappear after 400 °C. At 400 °C a new set of peaks begin to take form at about 45° and become clearly 'separated' by 800 °C. One of these peaks is well matched to Fe (45.1°) and the other to $Fe_{0.75}Pt_{0.25}$ (43.1°). However, a strong peak at 45.3° could represent the presence of the iron silicide Fe_5Si_3 (211). This indicates that as the FePt film is annealed the Pt is continually being removed from the as deposited structures. Some Pt does remain bonded to the FePt as the $Fe_{0.75}Pt_{0.25}$ and $Fe_{0.5}Pt_{0.5}$ matches show, but a significant amount of Pt appears to have reacted with the silicon substrate forming platinum silicides. As annealing temperatures are increased to over 350 °C, a mixture of various iron silicides and platinum silicides are produced.

The x-ray diffraction results for the nitrided (FePtN) samples are shown in Fig. 2. Where in the FePt samples the disappearance of the main $FePt_3$ (111)/FePt (111) peaks is accompanied by the emergence of new peaks after 350 °C, this does not occur in FePtN. At 800 °C many new peaks do appear but the FePt (111) peak remains. FePt(100) and (110) peaks are shown to emerge at 500 °C and are clearly visible at 600 °C. This shows improved ordering of the film. When 800 °C is reached, many more new peaks appear. As well as peak matches to iron and platinum silicides, FePt(001) and (002) peaks are also present, which represent the phases for the high anisotropy $L1_0$ structure.

By comparison of Fig. 1 and Fig. 2 it can be seen that in FePtN starting films the presence of nitrogen appears to hinder silicide formation. This is most likely due to reaction of the nitrogen with the Si substrate, preferentially forming silicon nitride over platinum silicide. This is supported by considering thermodynamic data for the likely reactions expected for our films (Table 1). The enthalpy of formation for Si_3N_4 is much more negative than for the formation of the other products. This means that the formation of Si_3N_4 has a lower energy than the formation of the other products, indicating that Si would preferentially combine with nitrogen rather than with either Fe or Pt.

Silicide formation can play a detrimental role in the magnetic properties of FePt films. Fig. 3 shows the variation of the in-plane and perpendicular coercivity values for the FePt and FePtN films as a function of temperature. These values have been extracted from hysteresis loops measured by vibrating sample magnetometry.

It can be seen from Fig. 3 that the in-plane coercivity of FePt increases

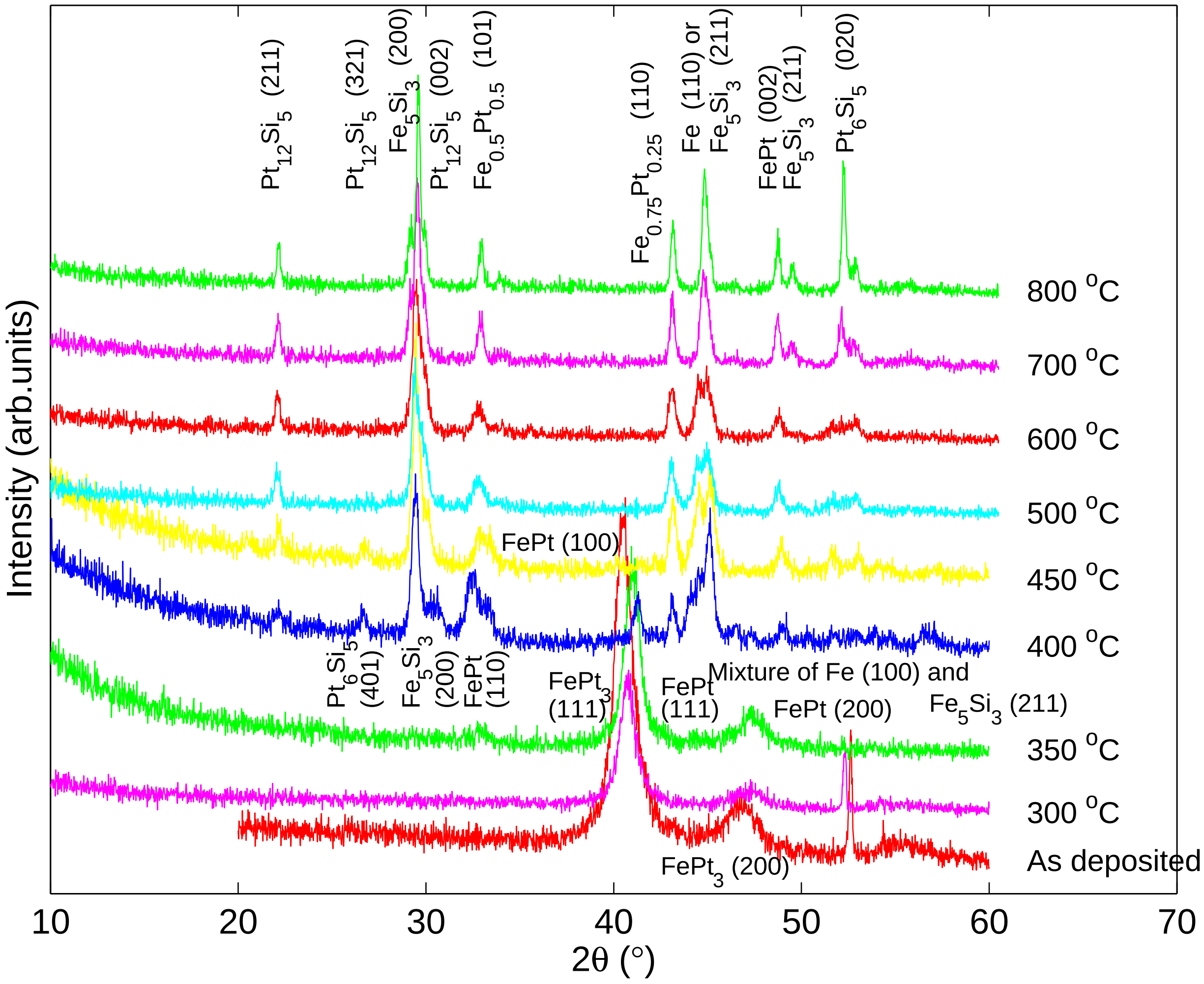


Figure 1: X-ray diffraction patterns of FePt for different anneal temperatures (deposited onto Silicon)

from 95 Oe to a maximum of 450 Oe after annealing at 450 °C. Increasing the temperature further results in a decrease of coercivity towards zero. The initial increase and subsequent decrease in coercivity is also observed in the perpendicular measurements, with an initial coercivity of 380 Oe rising to a maximum of 825 Oe at 350 °C before decreasing to zero at higher temperatures.

Both the in-plane and out-of-plane coercivities for the as-deposited ni-

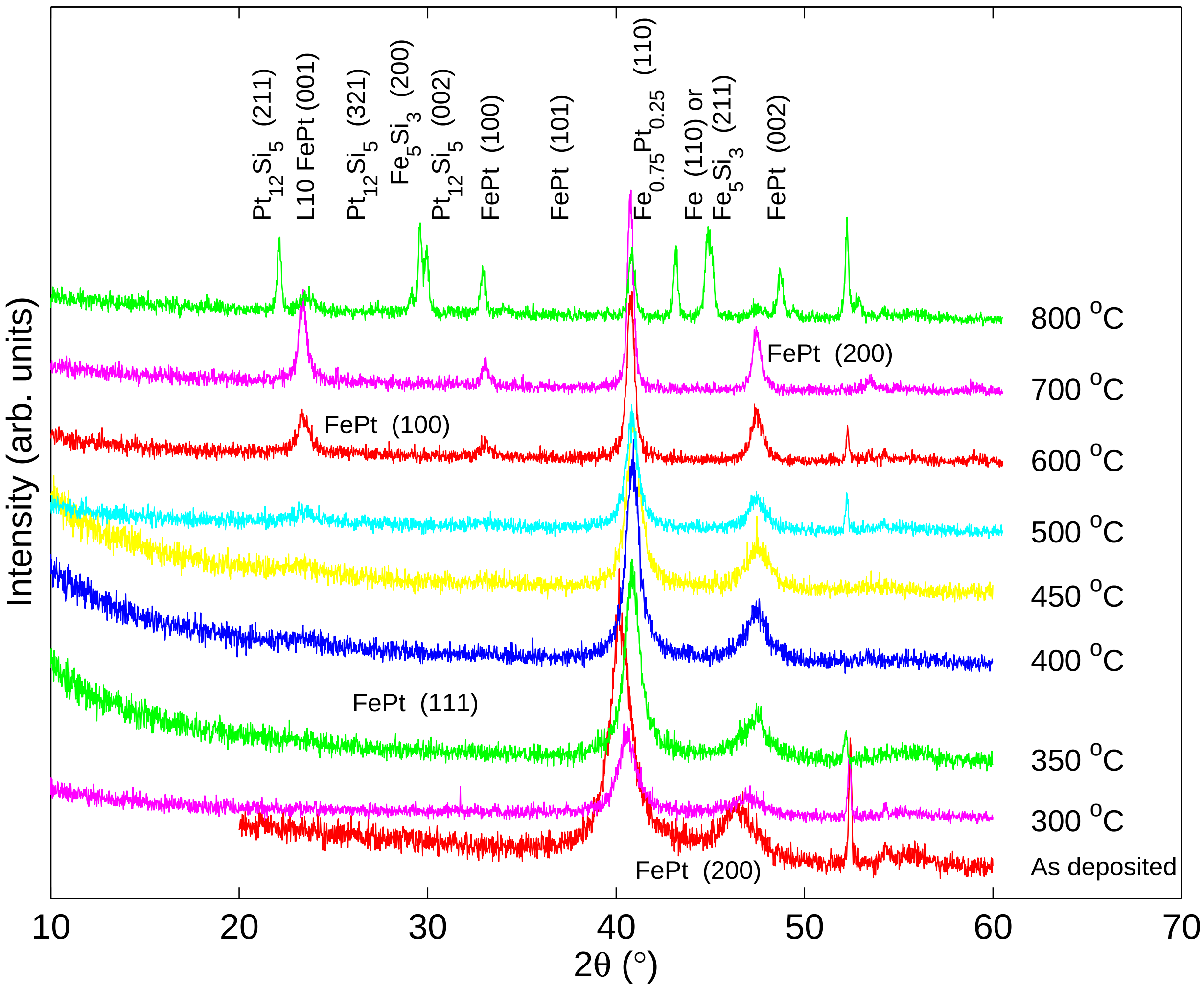
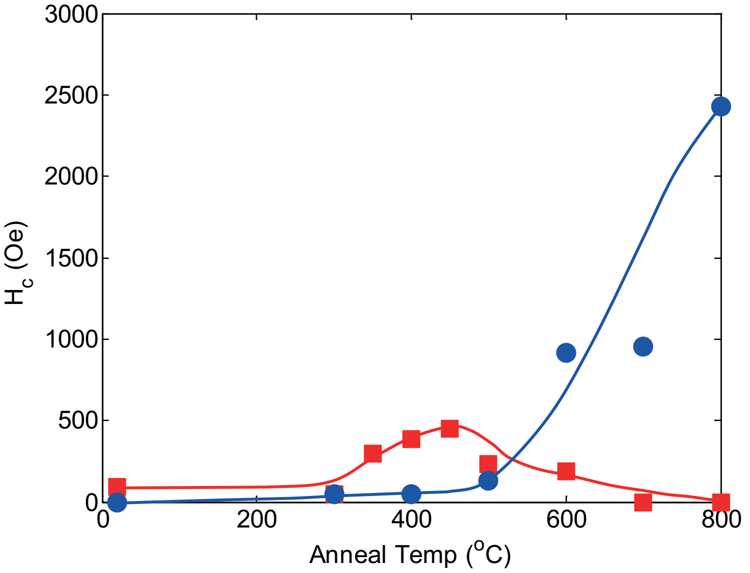
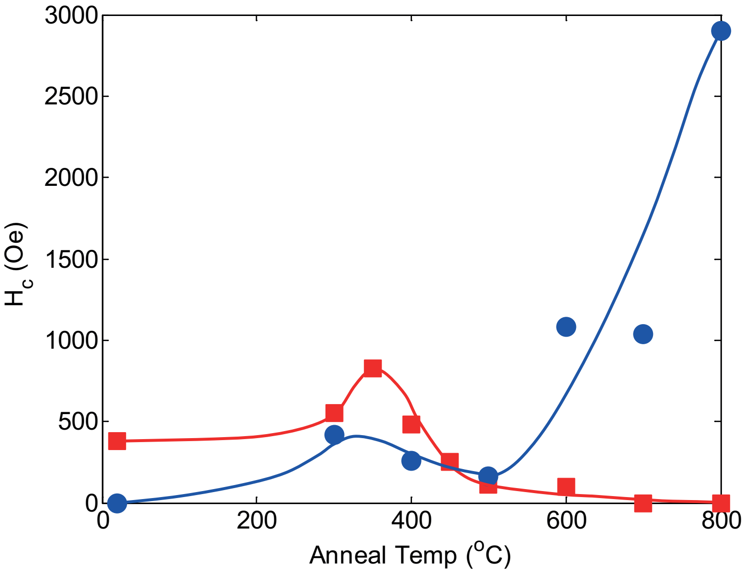


Figure 2: X-ray diffraction patterns of FePtN for different anneal temperatures (deposited onto Silicon)

trided samples have zero coercivity. This indicates successful inclusion of the nitrogen into the as-grown films, which are essentially non-magnetic (not ferromagnetic). The in-plane coercivity steadily increases with temperature, but from 500 to 600 °C there is a large increase from 130 to 900 Oe. Another major increase is apparent from 700 to 800 °C with a maximum in-plane coercivity of 2430 Oe at 800 °C. Interestingly, the out-of-plane coercivity increases to 420 Oe at 300 °C from zero (as deposited) but then decreases and



(a) In plane coercivity of FePt (red squares) and FePtN (blue circles)



(b) Perpendicular coercivity of FePt (red squares) and FePtN (blue circles)

Figure 3: In-plane and perpendicular coercivities of 250 Å FePt and FePtN films deposited on Si (solid lines are guides for the eye only)

Material	Enthalpy $(\Delta H_f^{\ominus})(KJmol^{-1})$	Reference
FePt	-27.2	[26]
FeSi	-99.8	[27]
PtSi	-59.6	[28]
Pt_2Si	-63.3	[28]
Si_3N_4	-828	[29]

Table 1: Enthalpy of formation values

increases again. These observations are likely to be due to the presence of iron nitride phases influencing the magnetic properties. Again major increases are observed between 500 and 600 °C and 700 to 800 °C with a maximum out-of-plane coercivity of 2900 Oe at 800 °C. Below about 500 °C FePt films have a higher coercivity (both in-plane and out-of-plane) than those containing nitrogen. Above annealing temperatures of 500 °C the coercivity of FePt diminishes but the coercivity (both in-plane and out-of-plane) of FePtN rises. These results suggest that the addition of nitrogen can offer FePt thin films protection against silicide pollution, therefore helping to preserve magnetic properties during thermal treatment and alleviating the requirements for inter-facial barrier layers between Si substrates and the magnetic film.

4. Conclusion

The high target utilisation sputtering (HiTUS) approach has been used to fabricate FePt and FePtN thin films onto silicon substrates. The effect of annealing on the structural phases and magnetic properties of the films were investigated. We show that during thermal annealing there is a degradation in magnetic properties of the FePt films at around 400 °C due to the formation of silicides, since the thermal processing promotes the reaction of the film with the substrate. However, in FePt films produced via a FePtN starting composition silicide formation is suppressed and the sample coercivity continued to rise with annealing temperatures up to the maximum temperature used of 800 °C.

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