

## Low-temperature fabrication of $L1_0$ ordered FePt alloy by alternate monatomic layer deposition

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$L1_0$  ordered FePt alloy films with large magnetic anisotropy have been successfully prepared by alternating Fe(001) and Pt(001) monatomic layers on MgO (001) substrates at low temperatures below 230 °C. In addition to the fundamental (002) peak, (001) and (003) superlattice peaks have clearly been observed in the x-ray diffraction patterns for all the samples, indicating the formation of  $L1_0$  ordered structure. The magnetization measurements show that all the samples are perpendicularly magnetized. Large uniaxial magnetic anisotropy ( $K_u = 3.0 \times 10^7$  erg/cc) and high chemical ordering (long-range order parameter  $S = 0.7 \pm 0.1$ ) have been obtained even at the substrate temperature  $T_s = 200$  °C. The magnetization curves show good magnetic squareness ( $M_r/M_s \sim 0.9$ ) for the samples grown at  $T_s \geq 160$  °C. © 2002 American Institute of Physics. [DOI: 10.1063/1.1432446]

$L1_0$  ordered FePt alloy with large magnetic anisotropy<sup>1</sup> has attracted much attention in recent years, since it is a potential material for extensive applications such as ultrahigh density recording media and bias magnets in monolithic microwave integrated circuits. A lot of studies were reported for Fe–Pt alloy films obtained by conventional deposition techniques such as sputtering and molecular beam epitaxy.<sup>2–7</sup> It is well known, however, that the substrate temperature during deposition and/or the post annealing temperature are required to be high (usually more than 500 °C) for the preparation of highly ordered FePt alloy films. For practical use, it is essential to reduce the growth temperature. Recently, several works on lowering of the ordering temperature were reported by multilayering<sup>8,9</sup> or ion irradiation.<sup>10</sup> However, the temperatures necessary for high chemical order were still quite high ( $\sim 300$  °C). In previous papers, we reported the artificial fabrication of ordered alloys by alternate monatomic layer deposition.<sup>11–13</sup> For example, the  $L1_0$  ordered structure consists of alternating monatomic layers of two different metal elements, and consequently,  $L1_0$  ordered FeAu alloy that does not exist in thermal equilibrium was successfully produced by alternating Fe(001) and Au(001) monatomic layers.<sup>12</sup> In this paper, it is proposed that the alternate monatomic layer deposition of Fe and Pt should be one of powerful methods for preparing  $L1_0$  ordered FePt alloy films at reduced temperatures.

Fe(1 ML)/Pt(1 ML) (ML: monatomic layer) multilayer films were prepared using an UHV deposition system with two independent  $e$ -guns. An Fe seed layer of 10 Å was deposited on a MgO (001) substrate, and consecutively an epitaxial Pt(001) buffer layer of 400 Å was grown at 70 °C. Monatomic layers of Fe (1.4 Å) and Pt (2.0 Å) were deposited alternately at various substrate temperatures ( $T_s$ ) in the range of 120–230 °C. The substrate temperature was measured by a tungsten-rhenium thermocouple on the back side of the substrate holder. It was, in advance, calibrated by the

comparison with the value measured using another tungsten-rhenium thermocouple wrapped up in a copper foil, which was contacted directly on a MgO substrate. The repetition of both Fe and Pt layers was 50 times. The typical deposition rate was 0.1 Å/s. The thickness was controlled based on the values monitored by a quartz crystal oscillator. The calibration of the oscillator was carefully made by the observation of reflection high-energy electron diffraction (RHEED) intensity oscillations for Fe on Fe and Pt on Pt before the preparation of multilayer films. X-ray diffraction with  $CuK\alpha$  radiation was performed for structural characterization. Magnetic properties were measured by a superconducting quantum interference device (SQUID) magnetometer in the field up to 5.5 T at room temperature, and the uniaxial magnetic anisotropy was determined from the area enclosed between the magnetization curves in applied fields parallel ( $H_{\parallel}$ ) and perpendicular ( $H_{\perp}$ ) to the film plane. The film composition was determined by Rutherford backscattering spectrometry (RBS) and all the samples showed the equiatomic ratio within the accuracy of  $\pm 3$  at. %.

The epitaxial growth of Fe and Pt layers was confirmed by RHEED. The RHEED patterns for the uppermost Pt layer of a Fe(1 ML)/Pt(1 ML) multilayer film grown at 200 °C is shown in Fig. 1. The sharp streak pattern confirms a relatively flat film surface and epitaxial growth. X-ray diffraction patterns for Fe(1 ML)/Pt(1 ML) multilayer films grown at 120 °C, 160 °C, 200 °C, and 230 °C are shown in Figs. 2(a), (b), (c), and (d), respectively. In addition to the fundamental (002) peak, (001) and (003) superlattice peaks of the face centered tetragonal phase have clearly been observed for all the samples, indicating the formation of  $L1_0$  ordered structure. No peaks from the other planes of  $L1_0$  ordered structure are seen. The sharp and intense superlattice peaks are observed for  $T_s = 230$  °C. However, the intensities of superlattice peaks decrease with decreasing  $T_s$ , indicating the reduction in the degree of chemical ordering.

Integration of the FePt (00L) ( $L = 1, 2, 3,$  and  $4$ ) peak areas yields a one-dimensional chemical ordering parameter  $S$  which can be defined as

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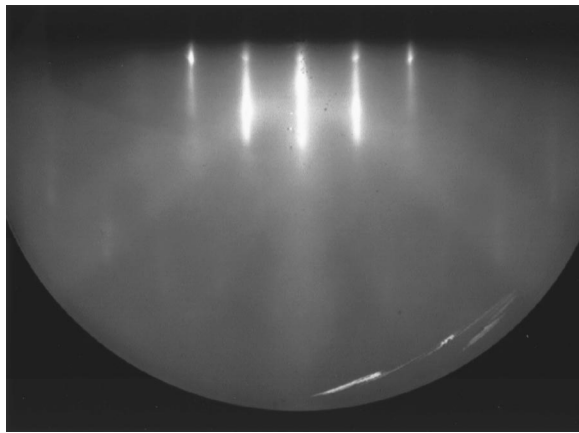


FIG. 1. The RHEED pattern ([100] azimuth) of the uppermost Pt layer for a Fe (1 ML)/Pt (1 ML) multilayer film grown at 200 °C.

$$S^2 = \frac{[I_{\text{super}}/I_{\text{fund}}]_{\text{obs}}}{[I_{\text{super}}/I_{\text{fund}}]_{\text{calc}}^2} \quad (1)$$

Here  $I_{\text{fund}}$  and  $I_{\text{super}}$  are the intensities of fundamental peaks and superlattice peaks, respectively. Calculated intensities of both  $I_{\text{fund}}$  and  $I_{\text{super}}$  were estimated by using following parameters; atomic fractions, atomic scattering factors, Debye–

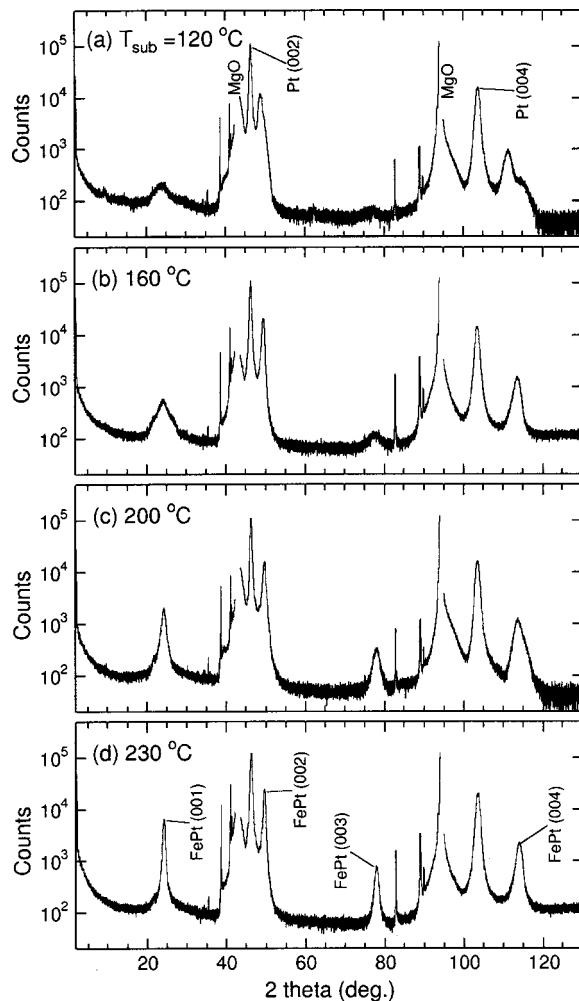


FIG. 2. X-ray diffraction patterns for Fe (1 ML)/Pt (1 ML) multilayer films grown at  $T_s$  = (a) 120 °C, (b) 160 °C, (c) 200 °C, and (d) 230 °C. In addition to the peaks associated with FePt ordered structure, the sharp peaks representative of Pt buffer layer and MgO (100) substrate are also observed.

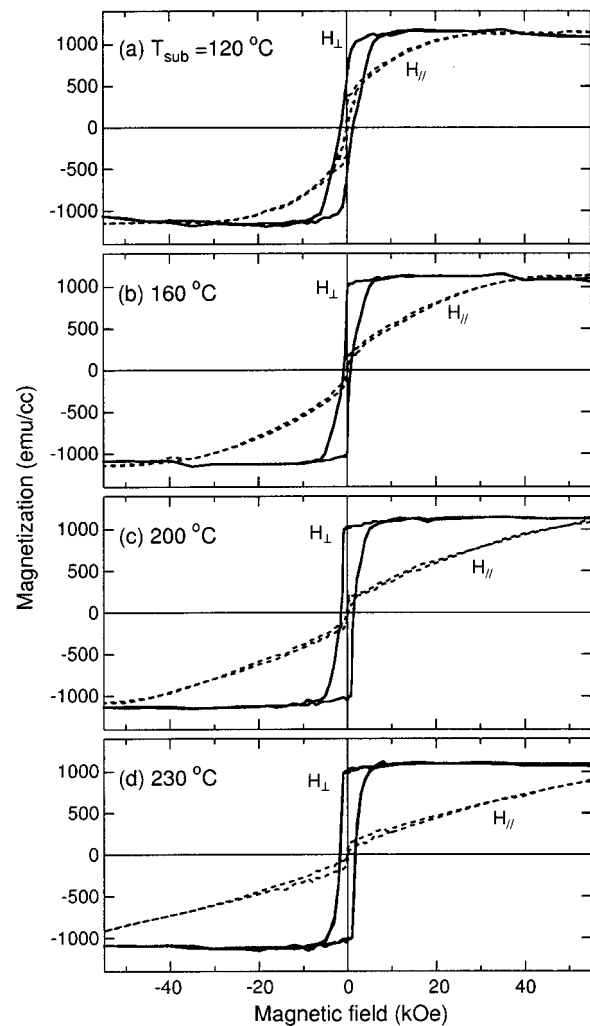


FIG. 3. Magnetization curves for Fe(1 ML)/Pt(1 ML) multilayer films grown at  $T_s$  = (a) 120 °C, (b) 160 °C, (c) 200 °C and (d) 230 °C measured in applied fields parallel ( $H_{\parallel}$ ) and perpendicular ( $H_{\perp}$ ) to the film plane.

Waller corrections, Lorentz polarization factors and structure factors.<sup>5,13</sup> The values of  $S$  for different  $T_s$  are listed in Table I.  $S$  ranges from 0.3 to 0.8, depending on  $T_s$ . High chemical ordering can be achieved for  $T_s \geq 200$  °C.

Magnetization curves for Fe(1 ML)/Pt(1 ML) multilayer films are shown in Fig. 3. The magnetic field was applied in parallel ( $H_{\parallel}$ ) and perpendicular ( $H_{\perp}$ ) directions to the film plane. For all the samples, the easy axis is perpendicularly to the film plane, since the [001] orientation of tetragonal  $L1_0$  ordered structure is perpendicular to the film plane. The magnetic field of 55 k Oe is not sufficient to saturate the magnetization in the in-plane direction for  $T_s \geq 200$  °C, indicating

TABLE I. Atomic fractions of Fe ( $x_{\text{Fe}}$ ) and Pt ( $x_{\text{Pt}}$ ) determined by the RBS analysis, chemical ordering parameter ( $S$ ), uniaxial magnetic anisotropy energy ( $K_u$ ), magnetic squareness ( $Mr_{\perp}/Ms_{\perp}$ ) and coercive force ( $Hc_{\perp}$ ) in the applied field perpendicular to the film plane for Fe(1 ML)/Pt(1 ML) multilayers for different substrate temperatures ( $T_s$ ).

$T_s$ (°C)	$x_{\text{Fe}}$ (at. %)	$x_{\text{Pt}}$ (at. %)	$S$	$K_u$ (erg/cc)	$Mr_{\perp}/Ms_{\perp}$	$Hc_{\perp}$ (Oe)
120	49.0	51.0	$0.3 \pm 0.1$	$1.4 \times 10^7$	0.51	1400
160	51.6	48.4	$0.5 \pm 0.1$	$2.3 \times 10^7$	0.87	800
200	52.4	47.6	$0.7 \pm 0.1$	$3.0 \times 10^7$	0.88	1500
230	49.3	50.7	$0.8 \pm 0.1$	$4.1 \times 10^7$	0.92	1700

the existence of large uniaxial magnetic anisotropy. Uniaxial magnetic anisotropy energy ( $K_u$ ), magnetic squareness ( $Mr_{\perp}/Ms_{\perp}$ ) and coercive force ( $Hc_{\perp}$ ) in the applied field perpendicular to the film plane for the Fe(1 ML)/Pt(1 ML) multilayer films as a function of  $T_s$  are listed in Table I. The uniaxial anisotropy energy  $K_u$  increases with  $T_s$ , and consequently with  $S$ , and it ranges from  $1.4 \times 10^7$  to  $4.1 \times 10^7$  erg/cc. This is quite a large value comparable to that of bulk L1<sub>0</sub> ordered FePt alloy ( $7 \times 10^7$  erg/cc).<sup>1</sup> A large  $K_u$  of  $3.0 \times 10^7$  erg/cc and a high coercive force  $Hc_{\perp}$  of 1500 Oe has been obtained even at  $T_s = 200$  °C. The magnetization curves also show good magnetic squareness ( $Mr_{\perp}/Ms_{\perp} \sim 0.9$ ) for the samples grown at  $T_s \geq 160$  °C. However,  $Hc_{\perp}$  decreases to 800 Oe for  $T_s = 160$  °C, corresponding to the reduction in  $S$ .  $Hc_{\perp}$  increases again for  $T_s = 120$  °C, possibly because of the change in domain structure due to the further reduction in  $S$ .

In conclusion, L1<sub>0</sub> ordered FePt alloy films with perpendicular magnetization have been successfully prepared by alternating Fe (001) and Pt (001) monatomic layers at temperatures below 230 °C, which are lower than the process temperatures reported so far for getting high chemical ordering. Large uniaxial anisotropy energy, high coercivity and good magnetic squareness were also obtained even for  $T_s \sim 200$  °C. Thus, the alternate monatomic layer deposition technique is very effective to fabricate the FePt ordered alloy at low temperatures.

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