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MAGNETIC EXCHANGE-COUPLING IN CoPt/Co BILAYER THIN FILMS

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ABSTRACT

Thin film CoPt/Co bilayers have been prepared as a model system to investigate the relationship between microstructure and exchange coupling in two-phase hard/soft composite magnets. CoPt films, with a thickness of 25 nm, were sputter-deposited from a nearly equiatomic alloy target onto oxidized Si wafers. The films were subsequently annealed at 700°C and fully transformed from the FCC phase to the magnetically hard, ordered $L1_0$ phase. The coercivity of the films increased rapidly with annealing time until it reached a plateau at approximately 9.5 kOe. Fully-ordered CoPt films were then used as substrates for deposition of Co layers, with thicknesses in the range of 2.8-225 nm, in order to produce the hard/soft composite bilayers. As predicted by theory, the magnetic coherency between the soft Co phase and the hard, ordered CoPt phase decreased as the thickness of the soft phase increased. This decrease in coupling was clearly seen in the magnetic hysteresis loops of the bilayers. At small thicknesses of Co (a few nanometers), the shape of the loop was one of a uniform material showing no indication of the presence of two phases with extremely different coercivities. At larger Co thicknesses, constricted loops, *i.e.*, ones showing the presence of a mixture of two ferromagnetic phases of different hardnesses, were obtained. The magnetic exchange present in the bilayer samples was qualitatively analyzed using magnetic recoil curves and the dependence of exchange coupling on the soft phase dimension in the bilayer hard/soft composite magnet films is discussed.

INTRODUCTION

The development of high-performance permanent magnet materials for energy-related applications is motivated by the need for increased maximum energy product, which describes the ability to store magnetostatic energy. The energy product increases with coercivity and remanence but can never exceed the intrinsic theoretical limit of $\mu_o M_s^2/4$. This limit corresponds to an ideal rectangular hysteresis loop, where the saturation magnetization (M_s) is equal to the remanence (M_r) and μ_o is the permeability in vacuum [1]. Thus, permanent magnet development has been focused on achieving this theoretical limit by increasing both coercivity and remanence. However, further increase of the energy product in single phase permanent magnets such as $SmCo_5$, and $Nd_2Fe_{14}B$ is intrinsically limited by their relatively low saturation magnetization.

The "exchange-spring magnets", which are nano-dispersed composites of hard and soft magnets, have been explored in attempts to approach the theoretical limit of the energy product [1,2]. It is expected that a significantly improved energy product may be obtained by exchange-coupling a soft phase with a high saturation magnetization to a hard phase with a high coercivity. However, appropriate control of the nano-scale microstructure is required to take full advantage of the attributes of both phases, since the exchange coupling depends strongly on microstructural factors.

To this end, fundamental studies of the role of microstructural attributes - grain size, spatial dimension of the phases, and structural coherency - on the interphase magnetic exchange coupling and nature of the domains and domain walls in the system must be carried out before a rational exploitation of these nanocomposite magnetic systems may be accomplished. In this work, bilayer thin films of CoPt/Co were prepared as a simplified one-dimensional exchange-coupled model system to investigate the fundamental relationships between microstructure and exchange-coupling.

EXPERIMENT

15-25 nm-thick CoPt films with near-equiatomic compositions (52-53^{at}% Co) were sputter-deposited on oxidized Si(100) substrates at room temperature. An arc-melted alloy of CoPt was used as the sputtering target cathode. Ar-4%H_2 was used as the sputtering process gas in order to ensure a reducing atmosphere during deposition, as suggested by K. Coffey [3]. The base pressure before deposition was below 5×10^{-8} torr and the sputtering gas pressure was 3×10^{-3} torr.

25 nm-thick CoPt films were deposited as underlayers for CoPt/Co bilayer films. 1.2-1.5 μm-thick freestanding films were prepared by a lift-off technique using Cu underlayers. These latter films were used for compositional analysis using wavelength dispersive spectrometry (WDS) and for determination of the order/disorder transition temperature using differential thermal analysis (DTA). The 25 nm-thick CoPt films were annealed in a tube furnace in Ar-4% H_2 at a flow rate of 100 ml/min at 700°C in the manner of Ristau [4] to produce magnetically hard, ordered $L1_0$ CoPt. For the preparation of thin film CoPt/Co bilayers, the 25nm-thick CoPt films were annealed for 120 minutes. These single-layer films were then reinserted into the UHV sputtering chamber for deposition of an additional layer of magnetically-soft Co. In this manner, bilayer CoPt/Co films with Co thicknesses in the range of 10 and 90% (Co thickness 2.8-225 nm) of the total bilayer thickness were prepared.

Plan-view transmission electron microscopy (TEM) samples of 25 nm-thick CoPt films subjected to various annealing times were prepared by chemical back-etching of the Si substrate. The microstructure was studied using a Philips EM400T operating at 120 kV. Magnetic sample coupons of 5 x 5 $(mm)^2$ films on Si substrate were prepared for SQUID magnetometry. Two oxidized Si wafer pieces of dimensions 5 mm x 60 mm were used to minimize the diamagnetic signals from the substrates during the magnetic measurements [5]. Magnetic characterization was performed at room temperature in the range $-5\ T \leq H \leq +5T$ with the field applied parallel to the film plane. In order to assess the exchange coupling qualitatively from our single-layer and bilayer samples, magnetic recoil curves were obtained for a number of recoil fields at 1000 Oe intervals along the demagnetization curve of a previously saturated sample. The mean slope of each recoil curve was then measured to analyze the change of reversible magnetic susceptibilities of the samples [6].

RESULTS AND DISCUSSION

The cross-sectional geometry of CoPt single-layer and CoPt/Co bilayer samples used in this study are shown in Fig. 1. The 25 nm-thick single-layer films were magnetically hardened through annealing as described above. The microstructural development of CoPt film as a function of annealing time is shown in Fig. 2. The films were transformed from a disordered fcc to an ordered $L1_0$ phase, evidenced by the appearance of the {110} and {011} superlattice reflections. Grain growth and <111> fiber texturing also occurred during annealing. The mean grain size of the film annealed for 120 minutes was 45 nm. Our results on ordering, grain growth and fiber texture development are in qualitative agreement with those of Ristau [4], who recently investigated the kinetics of these processes in 10 nm-thick, nominally equiatomic CoPt films.

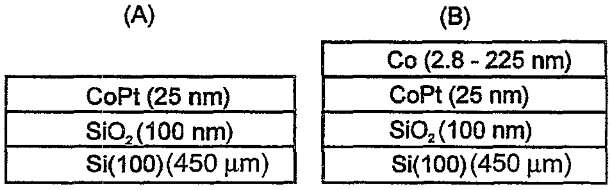

Fig. 1 - Schematic diagrams of the thin film samples used in this study.
(A) Single-layer CoPt
(B) Bilayer CoPt/Co

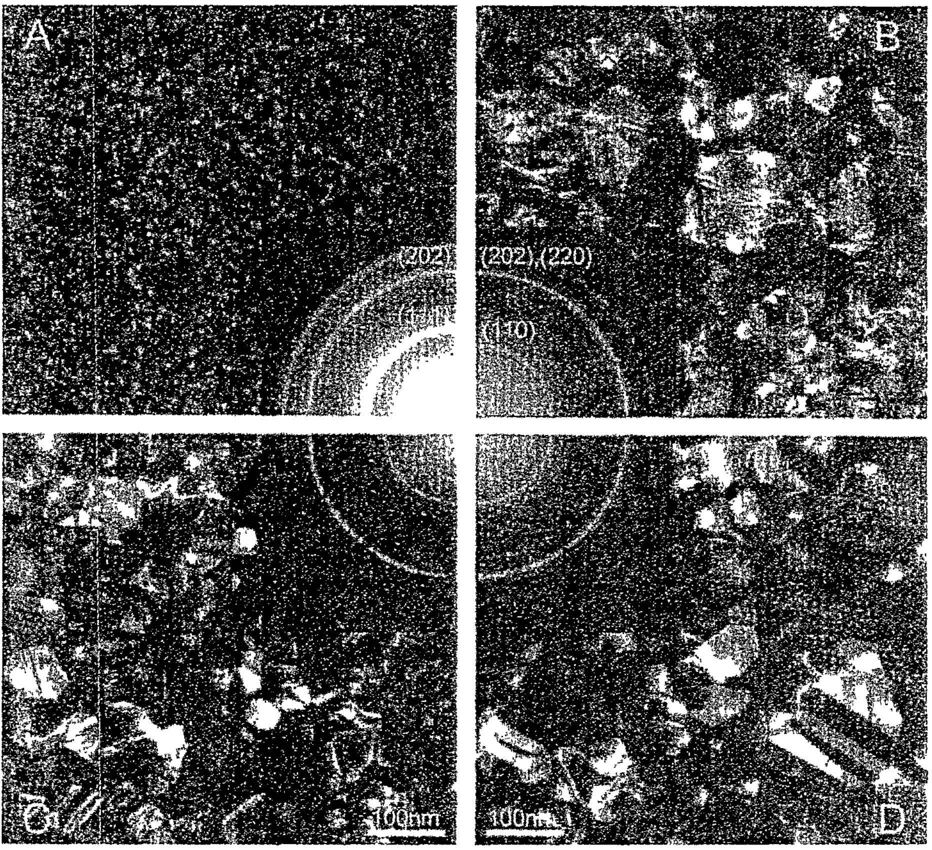

Fig. 2 - Transmission electron micrographs of 25 nm-thick CoPt films annealed for (A) 0 min (as-deposited), (B) 30 min, (C) 120 min, and (D) 900 min at 700°C.

The change of coercivity with annealing time for CoPt single-layer films of various thicknesses is shown in Fig. 3. The coercivity of 25 nm-thick CoPt films increased with annealing time and reached a plateau of 9.5 kOe after 120 min. This coercivity was maintained up to 900 min of annealing without agglomeration of the film. The lower coercivities in the present films compared with those of Ristau's are believed to be related to the Co-rich composition of our films. However, it is worth noting that, unlike Ristau's films [4], the samples used here can be annealed for considerably longer times without the films breaking up. Based on our coercivity vs. annealing time results, 120 min annealed CoPt films were used as hard phase underlayer for CoPt/Co bilayer composites. The average grain size of the 120 min-annealed CoPt films (45 nm) is about 6 times smaller than the calculated single magnetic domain size of 288 nm ($D_c \approx \gamma / M_s^2$, where γ (domain wall energy) $\approx$ 18.4 erg/cm^2, $M_s \approx$ 800 emu/cm^3[7,9]). It is postulated that the grains are exchange-coupled into multi-grain interaction domains which may or may not support distinct domain walls. The change of magnetization under influence of an external field may proceed by a rotational-type process, in which no clear domain walls exist but magnetization changes by an avalanche-type reversal sequence, or it may proceed by the more conventional route of domain wall motion. A combination of data obtained from diverse techniques is necessary to determine the presence and nature of domain walls in the system, and will be the subject of future work.

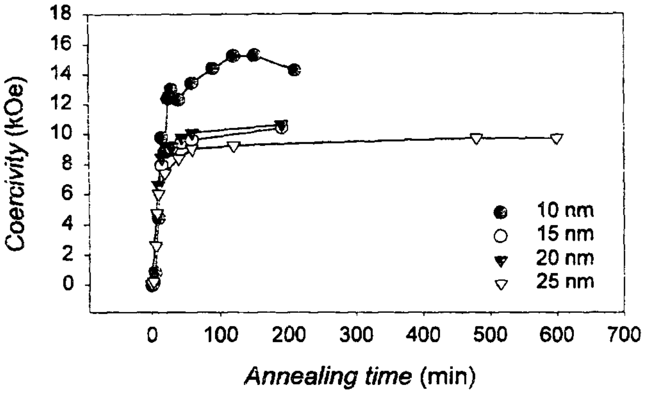

Fig. 3 - Coercivity as a function of annealing time at 700°C for CoPt films of differing thicknesses. Lines are drawn to guide the eye. The 10 nm-thick CoPt film data of Ristau [4] are included for comparison.

The fraction of $L1_0$ ordered, hard CoPt phase in the single-layer film increased with annealing time as the ordering proceeded [4]. Thus, this hard phase coexisted with the disordered fcc soft phase at all times during the microstructural development, resulting in a two-phase exchange-coupled nanocomposite. No discontinuities in the second-quadrant demagnetization curves were visible for any of the single-layer CoPt films. The mean slope of recoil curves to zero internal field, which, as the curves are nearly linear, is equivalent to the reversible susceptibility and is presented as a function of the recoil field in Fig. 4.

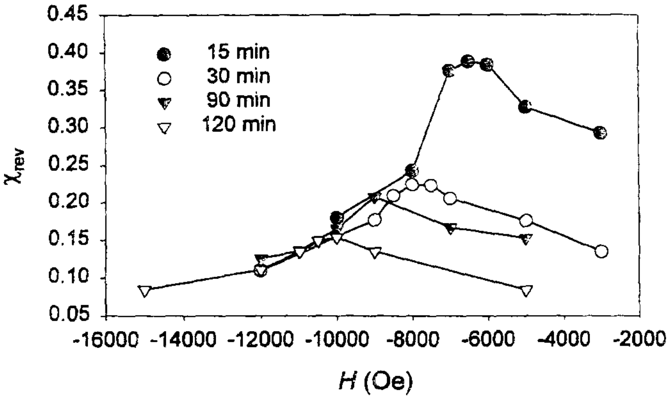

Fig. 4 - Mean slope of the linear recoil curves, as a function of recoil field, of 25 nm-thick CoPt single layer films subjected to different annealing times (χ_{rev} denotes reversible susceptibility).

All films show a maximum in the mean slope at the coercive field of the film. As the recoil field defines a particular remanent magnetization state for the film, and hence for a particular irreversible magnetization state, the plot in Fig. 4 shows equivalent information to a plot of reversible magnetization *vs.* irreversible magnetization. It has been shown by Crew, Woodward and Street [8] that such a plot with an extremum in the reversible magnetization indicates the presence of domain walls in the material which are able to bow reversibly under the action of a field. This conclusion is true even for the well-annealed film samples and indicates that although the grains are small enough to be single domain in isolation, exchange interactions group them into interaction domains which can support a domain wall. The observed increase in the coercivity with annealing time, and the decrease of the mean slope of the recoil curves with annealing time, is consistent with the increasing amount of hard phase. The increased ordered fraction was quantitatively measured for annealed 10 nm-thick CoPt films by Ristau [4].

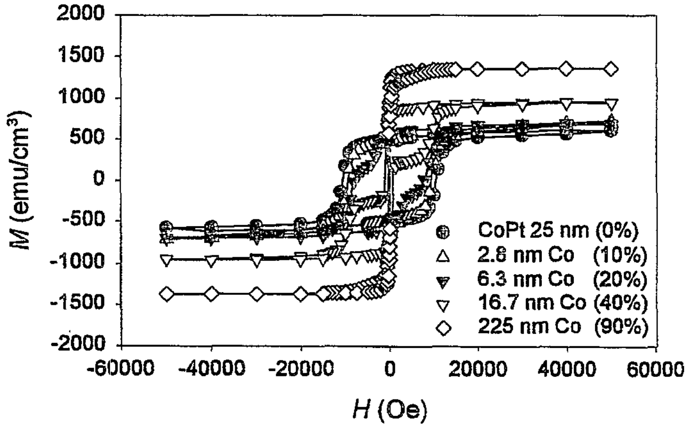

Fig. 5 - *M-H* loops for bilayer CoPt/Co films, with Co thicknesses ranging between 10% and 90% of the total film thickness.

The change of the shape of the magnetic hysteresis loop with Co (soft phase) thickness in the bilayer thin film systems is shown in Fig. 5. As the Co layer thickness increased, the coercivity decreased and the saturation magnetization increased. While the 2.8 nm Co layer (10% of the total thickness of the bilayer) was exchange-coupled to the CoPt layer for its entire thickness, the CoPt/Co bilayer with a Co layer thickness of 6.3 nm (20% of total thickness of the bilayer) began to decouple, as evidenced by the constricted hysteresis loop. Further increases in the Co layer thickness beyond 16.7 nm (40% of the total thickness) caused the coercivity to almost disappear. At these thicknesses, the magnetic behavior of the composite is dominated by that of the decoupled soft Co layer.

The theoretical domain wall width in ordered CoPt is calculated to be 4.6 nm (using the formula, $\delta \approx \sqrt{A/K}$, where A (exchange constant) ≈ 8.5 x 10^{-6} erg/cm^2 and K (magnetocrystalline anisotropy constant) ≈ 4 x 10^7 erg/cm^3 [7,9]). Thus, it is anticipated that complete exchange coupling will break down for Co overlayer thicknesses greater than approximately 5 nm, or 17% of the total bilayer thickness. The mean slope of the linear recoil curves as a function of recoil field is illustrated in Fig. 6 for the samples that are fully exchange-coupled (CoPt, 120 min; 10% Co/CoPt) and partially exchange coupled (20% Co/CoPt).

The mean slope of the linear recoil curves increased with thickness of the soft phase layer, indicating that a greater amount of material was contributing to the reversible remagnetization processes. As was demonstrated earlier with the single-phase CoPt films, there was a maximum value of the mean slope of the recoil curve which occurred at the coercive field of each sample. Thus it may be postulated that domain walls are also present in these bilayer system, and are free to move under the influence of a reverse field.

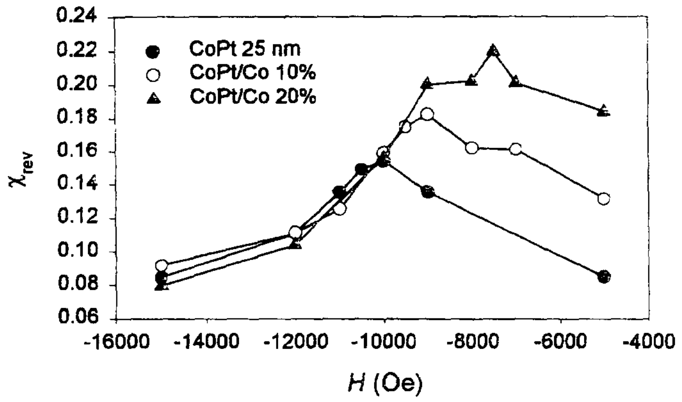

Fig. 6 - Mean slope of the linear recoil curves as a function of recoil field. (χ_{rev} denotes reversible susceptibility).

CONCLUSIONS

Interlayer exchange coupling was studied using CoPt/Co model bilayers to relate the exchange-coupling behavior of hard/soft nanocomposite magnets to fundamental microstructural factors. Exchange coupling is suggested to be present between the ordered $L1_0$ and disordered fcc phases in the single layer of CoPt. The interlayer exchange coupling between CoPt and Co increased with Co soft phase content below the critical dimension predicted from the domain wall width of the ordered CoPt phase, as evidenced by the second-quadrant demagnetization curves. The behavior of the mean slope of the linear recoil curves indicated that there were domain walls present in both the single-layer and the bilayer systems, and that these domain walls are free to move under the influence of a reverse field.

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