

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188	
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1 AGENCY USE ONLY (Leave blank)	2 REPORT DATE 05/13/99	3 REPORT TYPE AND DATES COVERED Final Progress Report, 08/15/98-02/14/99		
4 TITLE AND SUBTITLE Processing of Sm-Co Films in High Magnetic Field for Inducing Texture and Anisotropy		5 FUNDING NUMBERS DAAG55-98-1-0450		
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9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211		10. SPONSORING / MONITORING AGENCY REPORT NUMBER DAAG55-98-1-0450 ARO 29058.1 - MS		
11 SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.				
12a DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.		12b DISTRIBUTION CODE 19990706 041		
13 ABSTRACT (Maximum 200 words) The objectives of this study were to induce texture and anisotropy in the film plane by crystallizing amorphous Sm-Co films below the magnetic ordering temperature in the presence of high magnetic field along the film plane. Magnetically isotropic amorphous films were grown by magnetron sputtering and were crystallized at 450-600 C (below the magnetic ordering of Sm-Co alloys) in magnetic fields up to 29 T. However, no texture or anisotropy along the magnetic field direction was observed. The experiments were also conducted on anisotropic Sm-Co films that were grown on Cr/Ag/Si templates and had a significant in-plane anisotropy. When these films were subjected to high magnetic field up to 29 T, with their easy axis aligned along the magnetic field, and heated to 450-600 C no enhancement in magnetic anisotropy due to magnetic field effect was observed, rather already existing magnetic anisotropy was lost due to thermal treatment. These experiments lead to the conclusion that magnetic energy term is insignificant relative to the other energies involved in the process in this temperature range. However, at higher temperatures, the magnetic energy term may become significant. Further experiments at elevated temperatures are planned in this context.				
14 SUBJECT TERMS		15. NUMBER OF PAGES		
		16. PRICE CODE		
17 SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED	18 SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

Processing of Sm-Co thin films in a high magnetic field for inducing texture and anisotropy

Foreword

The performance of permanent magnets is determined by intrinsic and extrinsic properties. Saturation magnetization M_s , anisotropic field H_A and Curie temperature T_c are intrinsic to the hard magnetic phase the magnet is made from. The extrinsic properties, remanence M_r , coercive field H_c , and the energy density $(BH)_{max}$ additionally depend strongly on the microstructure, such as grain size and crystallographic texture. Thin films of permanent magnet material are of particular interest for miniature devices, where strong local permanent fields are desired. An optimum permanent magnetic film consists of nanocrystalline grains of a hard magnetic phase with high uniaxial magnetocrystalline anisotropy for maximum coercivity. Additionally all the grains should have their easy magnetization direction (EMD) aligned in one direction, which leads to a nearly square shaped hysteresis with a remanence value close to the saturation magnetisation. Nanocrystalline SmCo films, as can be prepared by annealing amorphous deposits, allow very high coercivity values due to the small grain size of 30-50 nm. The crystallization process, however, leads to an isotropic orientation of the c-axes and the remanence value is limited to $1/2 M_s$. Consequently the energy density for these high coercive materials reaches only $1/4$ of the maximum possible value for a completely aligned sample. Therefore, texturing and orienting of grains in a particular direction is highly desired for permanent magnet applications.

Statement of the problem studied

The texturing of a permanent magnet sample improves remanence, loop squareness and energy density. It can possibly be achieved when a magnetic material is subjected to high magnetic fields at temperatures close to the Curie temperature, provided the Curie temperature is sufficiently high so that thermal diffusion and atomic mobility can be realized in the processed material. We conducted such magnetic annealing experiments at the National High Magnetic Field Laboratory (NHMFL) to test the possibility of texturing nanocrystalline SmCo thin films. This material is chosen as it has excellent intrinsic hard magnetic properties. The crystalline SmCo phases ($SmCo_5$, $SmCo_7$), which form in the range from 12 to 21 at% Sm, have a very high uniaxial anisotropy and a high Curie temperature of 700 to 900°C as required for the mentioned experiment. Furthermore, nanocrystalline SmCo films can be prepared by crystallizing amorphous films at temperatures above 450°C. Applying a magnetic field during the crystallization of the hard magnetic phase may lead to an anisotropic nucleation, which is in favor of a texturing during grain growth. Based on these considerations three sets of experiments have been performed.

(1) The crystallization of amorphous SmCo films has been studied by annealing the samples without a magnetic field to evaluate the critical process parameters, annealing temperature and annealing time. The SmCo films with composition around $Sm_{20}Co_{68}Fe_7Cu_4Zr_1$ were sputter deposited on polycrystalline Al_2O_3 substrates using

commercial SmCo sinter targets. The substrate temperature was below 200°C, which leads to the formation of soft magnetic, amorphous SmCo. The films were subsequently heated *in-situ* without removing them from the sputter chamber. The vacuum during annealing was better than 1×10^{-6} Torr. The films were heated for 1h at temperatures between 400 and 600°C

(2) Amorphous SmCo film have been crystallized in the presence of a high magnetic field of 24-29T. Small samples of amorphous SmCo films, prepared as in (1), were sealed under vacuum in a quartz tube together with additional Ti and Nd file cuttings. They were mounted with a magnetic field oriented in the film plane and annealed for 1 h at temperatures between 450 and 600°C.

(3) Magnetic annealing experiments have been conducted on SmCo films with already existing uniaxial texture. The samples used for this study were prepared by sputtering SmCo on a textured Cr//Ag//Si template and were obtained from Carnegie Melon University [1]. They have a small but distinct uniaxial in-the-plane texture and coercive fields around 500 Oe. Diffraction studies both with XRD and TEM reveal no crystallographic structure, but the existence of magnetic texture indicates an anisotropic short-range order inside the SmCo film. Magnetic annealing experiments at 450 and 500°C for 1 h were performed with a field of 29 T present along the easy axis of the magnetic film, to see whether anisotropy and texture can be enhanced.

Summary of the most important results

(1) The amorphous films after sputtering on a 200°C substrate exhibit isotropic, soft magnetic behavior and the saturation magnetization of $M_s \approx 1T$ is determined by the amount of ferromagnetic transition metal elements (Co, Fe) in these films. Annealing at 400°C does not change the crystallographic and magnetic properties. Annealing above 450°C leads to the crystallization of the desired SmCo₅ phase with its high uniaxial magnetocrystalline anisotropy. The nanocrystalline grain size leads to high coercivities around $\mu_0 H_c = 2T$, the random distribution of nanograins, however, results in an isotropic magnetic behavior. In the temperature range from 450°C to 600°C the samples possess very similar properties. As examples for still amorphous and crystallized SmCo, the hysteresis loops for films annealed at 400°C and 520°C are seen in Fig. 1. The kink observed in Fig. 1b at small fields is attributed to a small amount of oxidized material. Although the annealing took place *in-situ* with a high vacuum, outgasing of the chamber during the annealing may introduce some reactive gases, which affect the sample. A first magnetic annealing experiment at the NHMFL with the sample mounted in streaming Argon gas showed an even more pronounced shoulder due to oxidation. These observation show, that a careful sealing of the samples is required for the annealing experiments to be successful.

(2) The magnetic properties of an amorphous SmCo thin film annealed 1 h at 500°C in a field of 24T are seen in Fig. 2a. The sample shows a smooth hysteresis with a high coercivity of $\mu_0 H_c = 2.5 T$, but the properties are identical in all 3 principal sample

directions. Similar properties were found in films after annealing for 1h at 450, 550 and 600°C in a field of 24T and for a film processed for 6h at 450°C in a field of 29T. This reveals that neither the nucleation of the SmCo₅ grains nor its subsequent growth in the amorphous matrix at temperatures up to 600°C can be influenced by a magnetic field.

(3) Fig. 3a shows the anisotropic magnetic behavior of the sample sputtered on a textured Cr//Ag//Si template in the initial state (before annealing, solid lines) and after annealing for 1h at 450°C in a magnetic field of 29T aligned along the easy axis (dashed lines). A similar result is obtained for a heat treatment at 500°C. The heat treatment reduces both coercivity and in-the-plane texture, indicating that the anisotropic short-range order cannot be enhanced or preserved via annealing, even in the presence of a magnetic field. Furthermore the low coercivity values indicate that no crystallization of the SmCo₅ phase takes place, although it can be expected for the given composition and annealing temperature. No significant structural changes upon the annealing can be observed in the XRD experiment (Fig. 3b).

The above experiments lead to the conclusion that magnetic energy term is insignificant relative to the other energies involved in the crystallization process in the temperature range investigated. However, at higher temperatures the magnetic energy term may become significant. The short duration and non extendable nature of the project did not permit to do further experiments. However, experiments at higher temperatures may provide further information in the above context.

Acknowledgement: We are thankful to G. Zangari for providing the Sm-Co films deposited on Cr//Ag//Si templates.

References:

- [1] G. Zangari, B.Lu, D.E. Laughlin, J. Appl. Phys. 85, 5759 (1999).

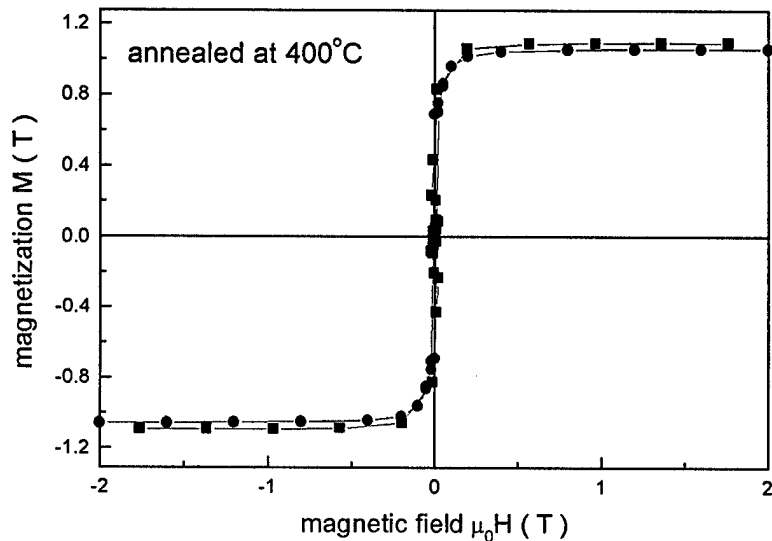


Fig 1 (a): Hysteresis loop, measured in two directions, of an amorphous SmCo film after annealing at 400°C. No crystallization occurred at this temperature and the sample shows isotropic, soft magnetic behavior.

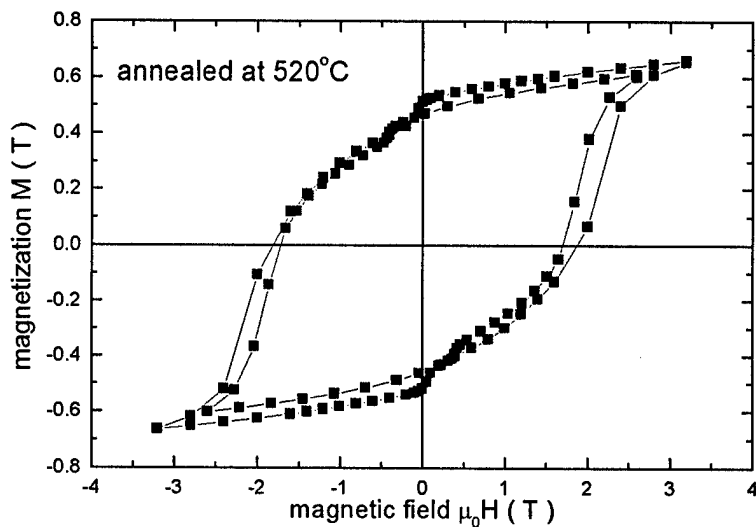


Fig 1 (b): Hysteresis loop, measured in two directions, of an amorphous SmCo film after annealing at 520°C. The hard magnetic SmCo_5 phase has formed and leads to high coercivity. The crystallization process, however, is isotropic.

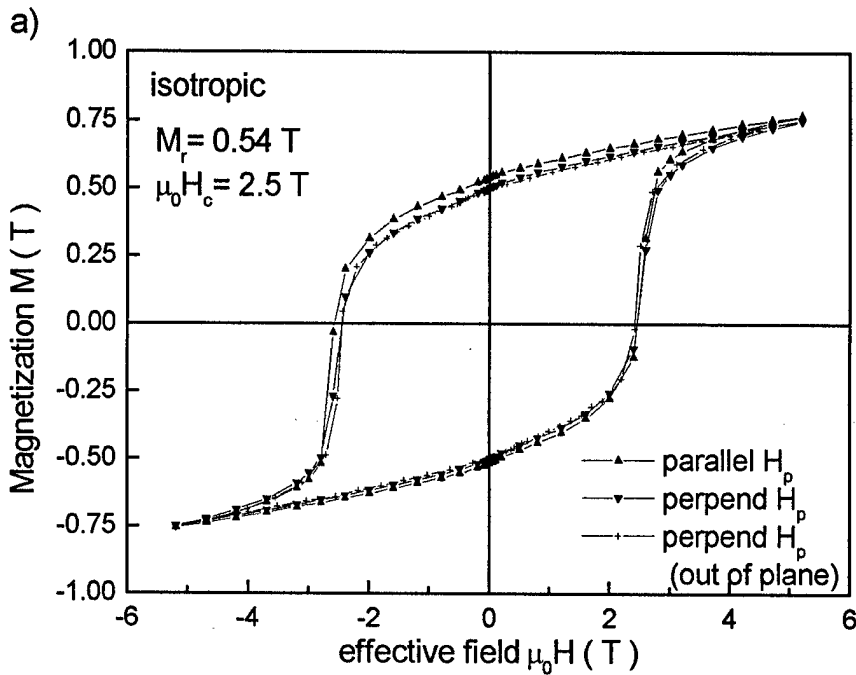


Fig. 2 (a): Hysteresis loop of SmCo sputtered on Al_2O_3 ($T_{\text{sub}} = 200^\circ\text{C}$) and annealed in a field $\mu_0 H_p = 24$ T at 500°C . The two measurements in the film plane (parallel and perpendicular to the process field) and the measurement perpendicular to the film plane give identical magnetization curves, indicating the isotropic behavior of the film.

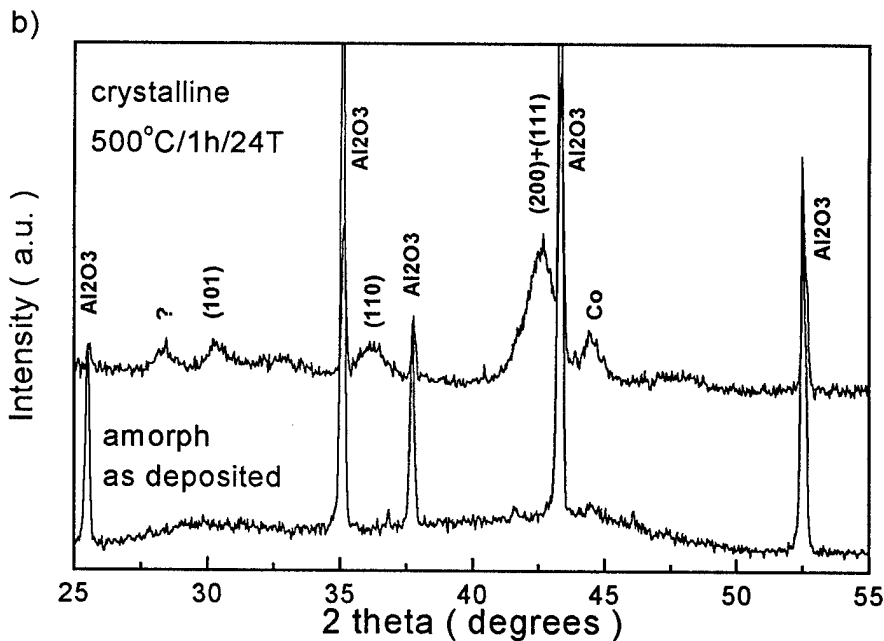


Fig. 2 (b): XRD trace of the sample shown in (a) before and after annealing. Except for the reflection at $2\theta \approx 28.4^\circ$ the processed film can be indexed according to the SmCo_5 phase and some elemental Co.

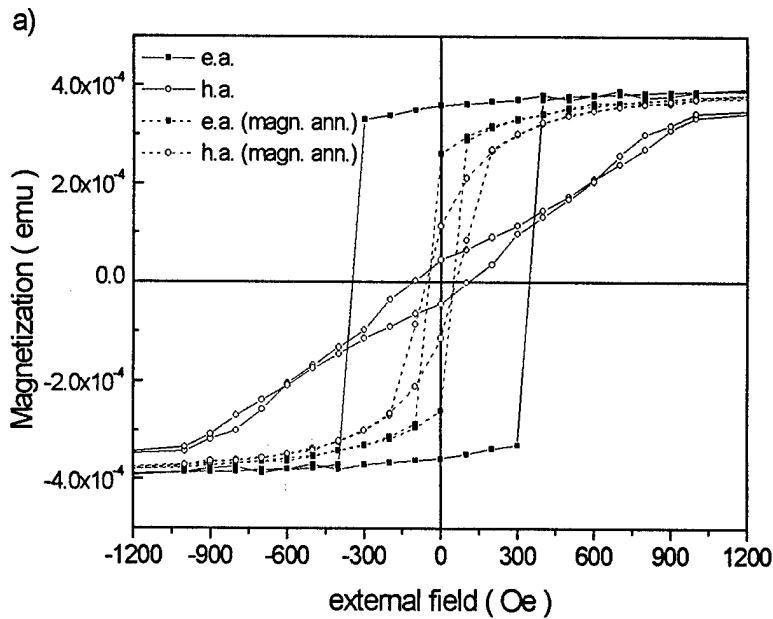


Fig.3 (a): Hysteresis loops of SmCo sputtered on a textured Cr//Ag//Si template with pronounced in-the-plane texture (solid lines), which vanishes after annealing for 1h at 450°C in a field of 29T (dashed lines); (b): XRD traces of the samples shown in (a). No significant structural changes can be observed upon magnetic annealing.

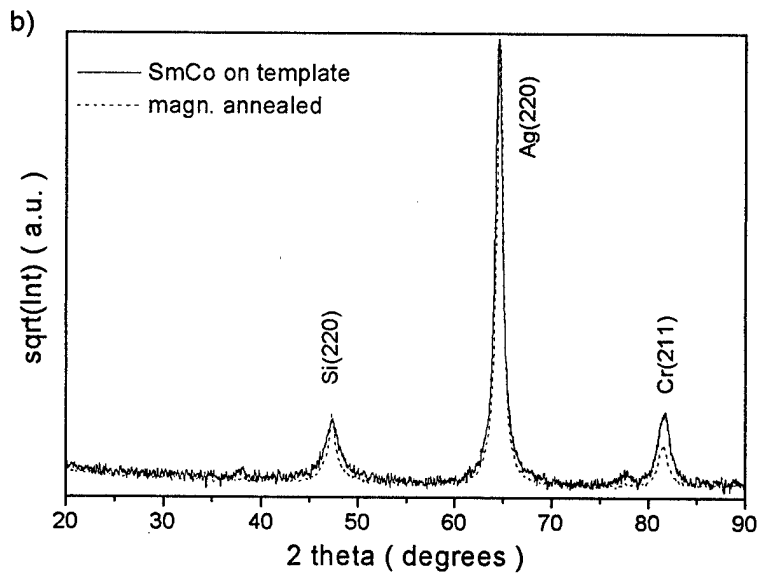


Fig. 3 (b): XRD traces of the samples shown in (a). No significant structural changes can be observed upon magnetic annealing.