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Technical Progress Report

ARTIFICIALLY STRUCTURED MAGNETIC MATERIALS
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I. INTRODUCTION

This document reports the progress made during the past year of our three-year DOE grant on "Artificially Structured Magnetic Materials."

Following this Introduction, the remaining sections of this report describe progress with DOE funding during the past 12 months; description of the research to be conducted during the remaining few months of the current grant year (through March 31); a description of the status of the graduate students working on this research; lists of the invited talks, seminars and colloquia, of other recognition of our research, and of the publications crediting DOE sponsorship; and a summary of current and pending federal support.

Our research efforts are directed toward studies of magnetism at surfaces and interfaces in well-characterized materials prepared by Molecular Beam Epitaxy (MBE) and sputtering. We have a very well equipped laboratory for these studies, with:

Thin film preparation equipment: two heavily instrumented MBE machines, two computer-controlled, multi-target sputtering machines, and an electron beam gun evaporator;

Characterization equipment: various x-ray diffraction instruments including Bragg-Brentano θ-2θ, low-angle, wide-film Debye-Scherrer (Read), Laue, and Seemann-Bohlin; Scanning Tunneling and Atomic Force Microscopy (STM and AFM); Auger, X-Ray Photoelectron Spectroscopy (XPS), Ion Scattering Spectroscopy (ISS), and Secondary Ion Mass Spectroscopy (SIMS); Reflected High and Low Energy Electron Diffraction (RHEED and LEED); and Scanning and Transmission Electron Microscopy (SEM and TEM);

Equipment to study magnetic properties of surfaces of ultra-thin magnetic films, and interfaces in multilayers and superlattices: *in situ* (to 2.2 kOe at 10^{-10} torr) and *ex situ* (to 15 kOe) Surface Magneto-Optic Kerr Effect (SMOKE) and variable-temperature Vibrating Sample Magnetometer (VSM).

Excellent progress and recognition of our DOE-funded research continued this past year, as evidenced by the 12 publications, 7 invited talks at international conferences, 8 contributed abstracts, and 8 seminars during 93-94 which credit DOE support.

II. RESEARCH ACCOMPLISHMENTS: OCTOBER 1, 1993–SEPTEMBER 30, 1994

A. Introduction

As a result of our discovery of an anomalous magnetic anisotropy in ultra-thin Co films, our work this past year has concentrated on the determination of the underlying mechanisms responsible for this phenomenon. A detailed discussion of our recent findings are given below.

B. *In Situ* Kerr Effect Measurements of Ultra-thin Co Films

Our recent results on ultra-thin Co films suggests that the interface anisotropy is more strongly dependent on the electronic structure of the constituents than on the microscopic structure of the interface. To investigate this further, we have been studying the evolution of the magnetic anisotropy as an interface with a non-magnetic overlayer is progressively formed.

Prior to this past year, our technique to measure the evolution of the interface anisotropy with overlayer coverage involved performing *in situ* Kerr effect measurements after each of many separate sub-monolayer depositions. Although effective, this measurement process was labor intensive and time consuming. We therefore modified our MBE system to allow the formation of multiple coverage samples. A computer-controlled stepper motor was attached to the main sample transfer mechanism. In this way, a large diameter substrate (2-3") could be moved in discrete steps from behind a shutter during overlayer deposition, producing a stepped-wedge thickness profile of the deposited film. Hence, several different coverages can be investigated after one deposition. The sample is then precisely moved by the motor to the Kerr effect chamber where magneto-optic data are collected for each of the different coverages.

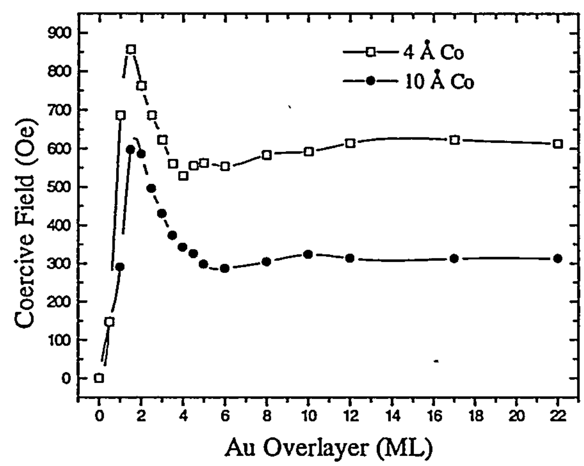

Figure 1 Coercive field H_C versus Au overlayer thickness on 4 Å and 10 Å Co films grown on Cu(111).

One of the interesting questions raised by our work on overlayer-induced anisotropy is the role of the first underlying interface. We have shown that Cu overlayers on Co grown on a Au(111) buffer layer display an unusual peak in anisotropy near one atomic layer. In order to study the effect of reversing the layer order, we have grown several samples on a Cu(111) buffer layer on which we deposited Co films of various thicknesses. Overlayers of

Au were then deposited and the Kerr effect measured. Figure 1 shows the coercive field H_C versus Au overlayer thickness on 4 Å and 10 Å Co films grown on Cu(111). We see a pronounced peak in the coercivity at 1.5–2 atomic layers Au coverage. Comparing these results with those obtained on Pd(111) and Au(111) buffer layers, we see that the order of deposition does not significantly affect the overall anisotropy behavior.

C. Influence of crystal structure on the anisotropy

The role of crystal structure in our observed coverage-dependent anisotropy is an important question that is difficult to address. Because magnetic anisotropy is very sensitive to the local environment, subtle changes in atomic spacings could cause significant effects. For this reason, we have been using RHEED and LEED to investigate coverage-dependent changes in both the in-plane and out-of-plane surface lattice spacings. Details of our recent results are discussed below.

C-1. Out-of-plane spacings determined from LEED

We have extended and refined our technique to determine the out-of-plane lattice spacings of the top few monolayers using Low Energy Electron Diffraction (LEED). We are now able to obtain information about the out-of-plane spacings by performing intensity *vs.* voltage (I-V) measurements. Although not as rigorous as a full dynamical analysis, one can determine the average out-of-plane lattice spacing of the top few monolayers from measured I-V curve of the specular or (0,0) beam. This simple analysis is only valid for materials which show weak or no multiple scattering. The average lattice spacing is deduced by comparison of the resulting intensity peak locations to calculated Bragg peaks. In the case of Cu deposited on Co(111), the overlayer is a different material from the substrate and therefore this analysis provides a combined weighted average lattice spacing of both materials. As an example of the sensitivity of this technique, Figure 2 shows an I-V curve of a clean Co(111) surface and that from a bulk-like thick Cu film deposited on top of the Co surface. The two representative curves show well-defined Bragg peaks which are slightly shifted from each other. To measure the effect of the overlayer as it is grown, I-V curves were taken after each of many Cu depositions of ~ 1 Å each (the equivalent of 0.5 atomic layers of Cu). As the bare Co is increasingly covered with Cu we observe a continuous shift from the Co peak positions to the Cu peak positions. Further deposition of Cu beyond 8 Å coverage does not change the location of the peaks. These peaks can be associated with Bragg diffraction by taking into account an inner potential correction, indicating that both the materials Co and Cu behave nearly kinematically. The peak location of the specularly reflected beam can be calculated from the following relation:

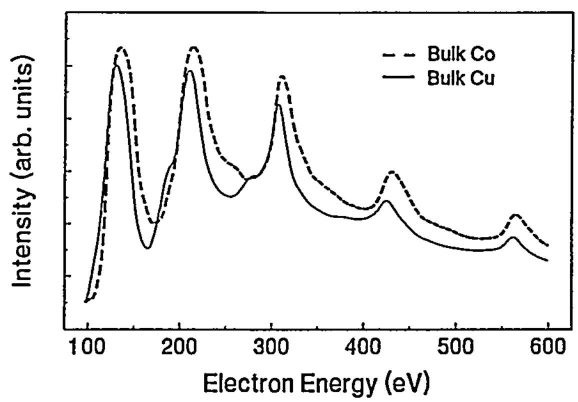

Figure 2 LEED I-V curves of (0,0) specular beam for Co(111) surface and a thick Cu film (200 Å) deposited on top of it. See text for explanation.

$$2(E^B + V_0)\cos^2\theta = (n\pi/d)^2 \qquad (1)$$

where E^B are the Bragg energies, V_0 is the inner potential, θ is the angle of incidence, n the order of the peak, and d the out-of-plane lattice constant. The energies and distances are given in Hartrees, 1h = 27.18 eV, and Bohrs, 1b = 0.529 Å, respectively. The angle of incidence of the electron beam was ~7 degrees in the <11-2> azimuth. We have measured the inner potential shift for bulk Co and bulk Cu to be 7.7 eV and 8.5 eV, respectively. It should be noted that we do not know how the inner potential V_0 varies as Cu is deposited. However, because the two bulk values are nearly the same, we used their average (8.1 eV) in all of our calculations. This causes only a small uncertainty in our determined lattice spacings and is included in the error estimates.

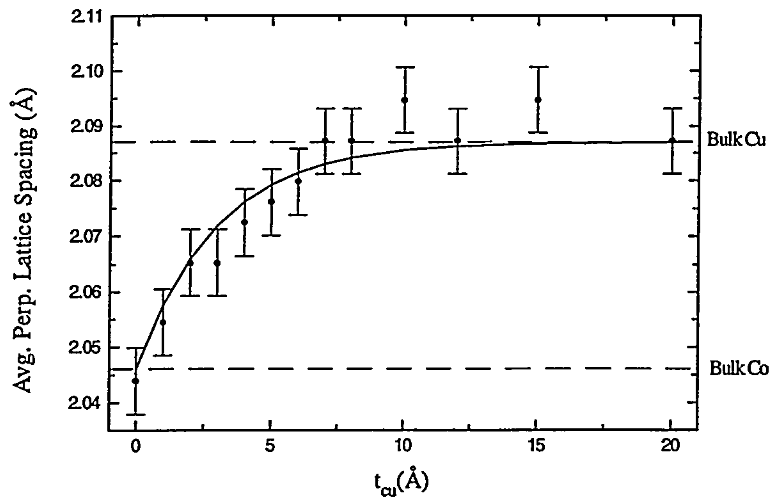

Figure 3 Change in average perpendicular lattice spacings determined from shifts in lowest energy peak locations in Fig. 2. The curve is a calculation discussed in text.

From the shifts in the Bragg peaks upon coverage we can calculate changes in the average out-of-plane lattice spacing according to equation (1), and which are shown as the circles in Figure 3. Here we have used the lowest energy peak at 137 eV which is the most surface sensitive with a mean sampling depth of only ~ 3 Å. The mean free path of LEED electrons at 137 eV is roughly 6 Å. However, in a reflection-diffraction experiment in which a monoenergetic beam must enter and exit the crystal, the mean sampling depth is half the value of the mean free path. We have also calculated the coverage dependence of the average lattice constant that would be expected if both the Co and Cu remain at their bulk spacings. This average spacing is calculated using depth dependent weighting factors derived assuming an exponential decay of the LEED electrons with a probing depth of 3 Å. The calculated solid curve in Figure 2 is in good agreement with the measured data indicating that, to within our uncertainty, Cu grows at its bulk perpendicular lattice constant on the Co surface. We see no evidence of any abrupt structural changes of greater than ~ 0.6 % at ~ 1 atomic layer coverage that would correlate with our observed peak in the anisotropy at this coverage.

We have also taken LEED I-V spectra of the specular beam for Au and Pd overlayers on Co. Both materials show strong multiple scattering where the peaks do not correspond to Bragg peaks and therefore the above simple analysis cannot be applied.

D. Effect of an Insulating Overlayer

In order to further clarify the role of electronic interactions in the interface anisotropy, we have measured the influence of an insulating overlayer, MgO on the perpendicular magnetic properties. For the noble metals Ag, Au and Cu, for which we observe the anomalous anisotropy behavior near 1 atomic layer coverage, there is also a hybridization between the ferromagnetic metal and the overlayer electronic states. In the case of an insulating MgO overlayer this electronic interaction between the ferromagnetic material and overlayer is extremely weak. Therefore, one would not expect significant changes in anisotropy upon insulator coverage.

The deposition of the MgO was carried out by e-beam evaporation. RBS analysis of the MgO film showed a 1:1 stoichiometric ratio of Mg to O, in agreement with previously reported results from e-beam evaporated MgO. We have also carried out Auger electron spectroscopy on the samples immediately after deposition. The Auger spectra of the deposited MgO film agree with those in the literature. Thus we do not expect any oxidation of the metal surface due to dissociation of MgO.

Figure 4 shows polar Kerr ellipticity loops of 18 Å Co deposited on a thick Au (111) buffer layer, 2 Å Cu deposited on the bare Co, and ~120 Å MgO deposited on top of the Cu. The initial magnetic moment of the bare Co is in-plane. As we previously reported, upon coverage with just 2 Å of Cu we see a large increase in the perpendicular anisotropy. Interestingly, after depositing a MgO cap of ~ 120 Å thickness we observe a decrease in the perpendicular anisotropy. The MgO shows polycrystalline growth as evidenced by the disappearance of the Cu RHEED streak pattern. In a different experiment we started with a perpendicularly magnetized 4 Å Co film with a coercive field of $H_c \approx 470$ Oe; subsequent deposition of ~ 30 Å polycrystalline MgO reduces the coercive field to $H_c \approx 70$ Oe with no change in the magnitude of the measured ellipticity.

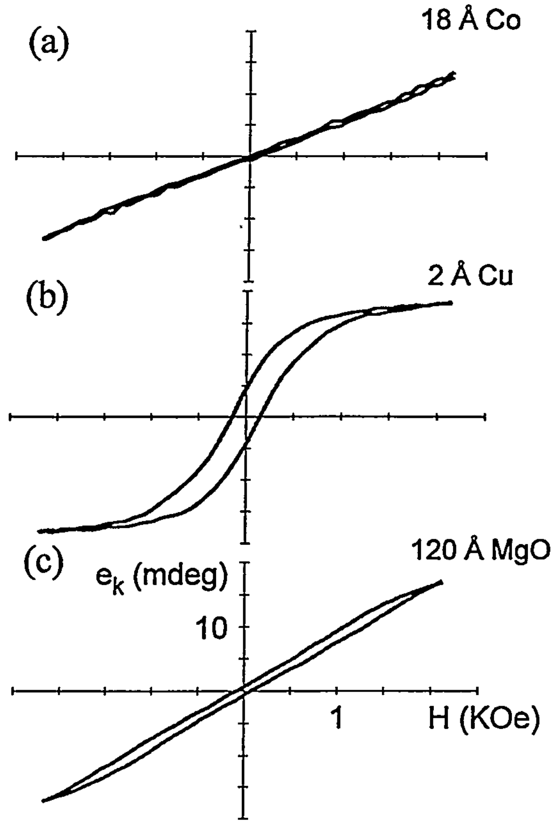

Figure 4 *In situ* polar Kerr ellipticity loop from: (a) uncovered 18 Å Co on Au(111); (b) 2 Å Cu deposited on 18 Å Co; (c) 120 Å MgO deposited on 2 Å Cu.

It is surprising to find such a large change in anisotropy since the electronic interaction between the metals and MgO should be very weak. It is possible that the MgO overlayer induces strain in the Co. Unfortunately, because MgO grows polycrystalline in this case we are unable to use *in situ* structural characterization techniques to investigate changes due to MgO coverage. Work is in progress to investigate other insulating materials.

E. Summary

To summarize this Section on research accomplishments during the most recent year of DOE funding, considerable progress was made in all three major areas of our program: deposition, characterization, and measurement of physical properties of multilayered materials. Although our results were only briefly described above, this research is described in detail in the 12 publications crediting DOE support which are listed below in Section V.

III. Research to be Conducted Through March 31, 1995

In the remaining months of the current year our work will continue to emphasize the production and characterization of high-quality, single-crystal MBE-grown materials for studies of the magnetic properties. In particular, we will concentrate the rest of the year on anisotropy studies of ultra-thin Co-based films grown along different crystal orientations. Our collaborations with other groups to study some of the magnetic properties by light scattering and neutron diffraction will continue. A number of *in situ* Surface Magneto-Optic Kerr Effect (SMOKE) measurements on MBE-grown ultra-thin films will be made during this next period. We also will study the magnetic properties of our multilayer materials using our Vibrating Sample Magnetometer (VSM).

IV. Status of Graduate Students Working on this Research

One student was funded by this DOE grant during the past year. As a result of his research for this project, Michael Wiedmann was awarded a Ph.D this past August. Dr. Wiedmann has now moved to a postdoctoral research position with Professor Albert Fert at the University of Paris-Sud in Orsay, France. We have recently hired another student, Mr. Dana Biddulph. Mr. Biddulph is a second year Ph.D. graduate student in the Physics Department, who has passed all of the department's examinations.

V. PUBLICATIONS, INVITED TALKS, ABSTRACTS, AND SEMINARS CREDITING DOE SUPPORT 93-94

A. Publications

1. **Anomalous Magnetic Anisotropy in Ultra-Thin Transition Metals.** Brad N: Engel, Michael H. Wiedmann, Robert A. Van Leeuwen and Charles M. Falco, Phys. Rev. B—Rapid Communications B 48, 9894 (1993).

2. **Properties of MBE-Grown Superlattices and Ultra-Thin Films.** Charles M. Falco and Brad N. Engel, in Proc. International Conference on the Physics of Transition Metals, (World Scientific, 1993) p. 446.

3. **Effect of Transition-Metal Overlayers on the Perpendicular Magnetism of MBE-Grown Ultra-Thin Co Films.** Brad N. Engel, Michael H. Wiedmann, Robert A. Van Leeuwen and Charles M. Falco, J. Appl. Phys. 73, 6196 (1993).

4. **Electronic Influence of Transition Metals on the Perpendicular Magnetism of MBE-Grown Co/ Pd.** Robert A. Van Leeuwen, Michael H. Wiedmann, Brad N. Engel, and Charles M. Falco, J. Magnetics Soc. of Japan 17, Suppl. S1, 136 (1993).

5. **Anomalous Perpendicular Anisotropy in Ultra-Thin Co Films.** Michael H. Wiedmann, Brad N. Engel, Robert A. Van Leeuwen, Ko Mibu and Charles M. Falco, Proc. of the Materials Research Society, 313, 531 (1993).

6. **Influence of Transition Metal Overlayers on the Perpendicular Magnetism of MBE-Grown Co Films.** Brad N. Engel, Michael H. Wiedmann, Robert A. Van Leeuwen, and Charles M. Falco, J. of Magnetism and Magnetic Materials 126, 532 (1993).

7. **Evidence for Collective Exchange Modes in Co/ Pd Multilayers Observed by Brillouin Light Scattering.** B. Hillebrands, J. V. Harzer, R. L. Stamps, G. Güntherodt, C. D.

England and Charles M. Falco, Proc. of the Materials Research Society – IN PRESS.

8. **Magnetic Anisotropies in MBE-Grown and Sputtered Co/ Pt(001), Co/ Pt(111) and Co/ Pd(111) Multilayers Investigated by Brillouin Light Scattering.** J. V. Harzer, B. Hillebrands, G. Güntherodt, R. Farrow, C. H. Lee, E. E. Marinero, H. Notarys, D. Weller, B. Engel, C. D. England and C. M. Falco, Proc. of the Materials Research Society – IN PRESS.

9. **Overlayer-Induced Perpendicular Anisotropy in Ultra-thin Co Films**
Brad N. Engel, Michael H. Wiedmann and Charles M. Falco
Journal of Applied Physics 75, 6401 (1994)

10. **Spatial Modulation of the Magnetic Moment in Co/ Pd Superlattices Observed by Polarized Neutron Reflectivity**
J.A. Borchers, J.F. Ankner, C.F. Majkrzak, B.N. Engel, M.H. Wiedmann, R.A. Van Leeuwen, and C.M. Falco
Journal of Applied Physics 75, 6498 (1994)

11. **Magnetic Anisotropy of Metal/ Co/ Metal and Metal/ Co/ Insulator Sandwiches**
Michael H. Wiedmann, Brad N. Engel, and Charles M. Falco
Journal of Applied Physics – IN PRESS

12. **Perpendicular Magnetic Behavior of Ultra-Thin Co Sandwiches**
Michael H. Wiedmann, Christian Marliere, Brad N. Engel and Charles M. Falco
Journal of Magnetism and Magnetic Materials – IN PRESS

B. Invited Talks at International Conferences Crediting DOE Support 93-94

1. **Magnetic Anisotropy in Magneto-Optical Multilayers.** Brad N. Engel and Charles M. Falco. Topical Meeting on Application of Magnetic Multilayers. Sendai, Japan. March 8, 1993.

2. **Structural Influence on Anisotropy of Co/ Pd Superlattices and Ultra-Thin Co Films.** Brad N. Engel and Charles M. Falco. Topical Meeting on Magneto-Optics of the Magnetics Society of Japan. Tokyo, Japan. March 9–10, 1993.

3. **Elastic Properties of Metallic Superlattices.** Charles M. Falco, John Dutcher, Jeha Kim, Sukmock Lee and George Stegeman. NATO Advanced Study Institute on Nanophase Materials. Corfu, Greece. June 20–July 2, 1993.

4. **Magnetic Properties of Metallic Multilayers.** Charles M. Falco and Brad N. Engel. European Magnetic Materials and Applications Conference. Košice, Czechoslovakia. August 24–27, 1993.

5. **Overlayer-Induced Perpendicular Anisotropy in Ultra-Thin Co Films.** Brad N. Engel, Michael H. Wiedmann, and Charles M. Falco. 38th Conference on Magnetism and Magnetic Materials. Minneapolis. November 15–18, 1993.

6. **Magnetic Properties of Surfaces and Ultra-Thin Films.** Charles M. Falco, Brad N. Engel and Michael H. Wiedmann. 3rd Magnetic Thin Films Symposium. Tsingdao, China. May 20–24, 1994.

7. **Preparation and Characterization of Ultra-Thin Magnetic Films.** Charles M. Falco. Tutorial on Multilayer Materials at the 6th Joint Intermag/Magnetism and Magnetic Materials meeting. Albuquerque. June 19, 1994.

C. **Abstracts of Contributed Talks Crediting DOE Support 93-94**

1. **Effect of Transition-Metals (TM) on the Perpendicular Magnetism of MBE-Grown Pd/ Co/ TM/ Sandwich and Multilayer Films.** B. N. Engel, M. H. Wiedmann, R. A. Van Leeuwen and C. M. Falco, International Symposium on Metallic Multilayers. Kyoto, Japan. March 1–5, 1993.

2. **Anomalous Magnetic Anisotropy in Ultra-Thin Transition Metals.** Brad N. Engel, Michael H. Wiedmann, Robert A. Van Leeuwen and Charles M. Falco, Bull. Am. Phys. Soc. 38, 371 (1993).

3. **Temperature-Dependent Pd Polarization Observed in a Co/ Pd(111) Superlattice Using Neutron Reflectometry.** J. F. Ankner, J. A. Borchers, C. F. Majkrzak, B. N. Engel, M. H. Wiedmann, R. A. Van Leeuwen and Charles M. Falco, Bull. Am. Phys. Soc. 38, 618 (1993).

4. **Anomalous Perpendicular Anisotropy in Ultra-Thin Co Films**
Michael H. Wiedmann, B. N. Engel, R. A. Van Leeuwen, K. Mibu, T. Shinjo, and C. M. Falco, Materials Research Society Spring Meeting. San Francisco, CA. April 12-16, 1993

5. **Structure and Magnetic Anisotropy of Ultra-thin Co Sandwiches**
Michael H. Wiedmann, Brad N. Engel, and Charles M. Falco
American Physical Society Meeting. Pittsburgh, PA. March 21-25, 1994.

6. **Magnetic Anisotropy of Metal/ Co/ Metal and Metal/ Co/ Insulator Sandwiches**
Michael H. Wiedmann, Brad N. Engel, and Charles M. Falco
The 6th Joint MMM-Intermag Conference. Albuquerque, NM. June 20-23, 1994.

7. **Perpendicular Magnetic Behavior of Ultra-Thin Co Sandwiches**
Charles M. Falco, Michael H. Wiedmann, Christian Marliere, and Brad N. Engel
14th International Conference on Magnetic Films and Surfaces (ICMFS). Düsseldorf, Germany. August 29 - September 2, 1994.

D. **Seminars and Colloquia Resulting from our DOE-Funded Research 93-94**

1. **Influence of Transition Metal Overlayers on the Perpendicular Magnetism of MBE-Grown Co Films.** Brad N. Engel, Institute for Chemical Research, Kyoto University, Kyoto, Japan. February 16, 1993

2. **Effect of Transition Metal Overlayers on the Perpendicular Magnetism of MBE-Grown Co Films.** Brad N. Engel, Department of Physics, Nagoya University, Nagoya, Japan. February 26, 1993.

3. **Anomalous Magnetic Anisotropy in Ultra-thin Transition Metals.** Brad N. Engel, Toshiba Research Center, Kawasaki, Japan. March 10, 1993.

4. **Anomalous Magnetic Anisotropy in Ultra-thin Transition Metals.** Brad N. Engel, Electro-Technical Laboratory, Tsukuba Science City, Japan. March 11, 1993.

5. **Artificial Metallic Superlattices.** Charles M. Falco, Department of Chemistry, University of Arizona. Tucson. November 1, 1993.

6. **Magnetism at Surfaces and Interfaces.** Charles M. Falco, Physics Department, University of California, San Diego. December 1, 1993.

7. **Metallic Superlattices and Ultra-Thin Films: Magnetic and X-Ray Optical Properties.** Institut d'Optique, Université de Paris–Sud. Orsay, France. March 4, 1994.

8. **Magnetism at Surfaces and Interfaces.** Materials Science Division, Argonne National Laboratory. Argonne, Illinois. August 8, 1994.

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