Manuscript**MAGNETO-OPTICAL STUDIES OF ULTRATHIN FERROMAGNETIC FILMS*****S.D. Bader**

*Materials Science Division
Argonne National Laboratory, Argonne, IL 60439*

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

INVITED PAPER: "Microstructure of Magnetic Materials", 1992 Arizona State University High Resolution Electron Microscopy Workshop, Wickenburg, AZ, January 8-11, 1992 (special issue- Journal of Ultramicroscopy)

jmc

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*Work supported by the U.S. Department of Energy under contract #W-31-109-ENG-38.

MASTER

Magneto-Optical Studies of Ultrathin Ferromagnetic Films*

S. D. Bader

Material Science Division, Argonne National Laboratory, Argonne, IL 60439

Recent developments are reviewed in the general area of surface magnetic anisotropy of ultrathin ferromagnetic films of 3d-transition metals. The surface magneto-optic Kerr effect (SMOKE) is introduced as a probe of magnetic anisotropy effects in ultrathin ferromagnetic films. SMOKE results from the author's laboratory are highlighted, including the study of the stabilization of vertical easy axes of magnetization in a variety of epitaxial Fe film systems.

Invited Paper:

Workshop on Microstructure of Magnetic Materials; Wickenburg, AZ; Jan. 8-11, 1992.

To be published in a special issue of the journal Ultramicroscopy.

- * Work supported by U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under Contract #W-31-109-ENG-38.

I. Introduction:

The focus of this paper is on surface magnetic anisotropy of 3d ferromagnetic transition metals. While the exchange interaction is responsible for the net magnetization associated with ferromagnetism, it is the magnetic anisotropy energetics that determine the direction of that magnetization with respect to the crystallographic axes and to the shape of the sample. Van Vleck^[1] originally proposed that the magnetic anisotropy of the ferromagnetic transition metals was due to the spin-orbit interaction. Néel^[2] was then the first to introduce the concept that the surface can have its own strong magnetic anisotropies due to the lowered symmetry relative to the bulk crystal. Gradmann and coworkers^[3] were first experimentally to observe the consequences of strong surface anisotropies in terms of vertical easy axes of magnetization for ultrathin films, whereas the easy axes are in-plane for thicker films for which surface effects can no longer dominate.

The study of surface magnetic anisotropy has grown in popularity with the advent of vertical recording and magneto-optic data storage media. It is now clear that surface magnetic anisotropy can endow a material with device potential and marketability. When this is compounded with the realization that ultrathin films and artificially layered materials can be atomically engineered to have the desired anisotropies, the renewed interest is quite understandable. These developments have led Kaneyoshi^[4] recently to point out that the surface magnetic anisotropy is "of paramount importance in determining the magnetic properties of surfaces and thin films."

Both theoretical and experimental activities are underway currently. The theoretical studies are being pursued along a variety of avenues. Phenomenological theorists utilize power-series expansions of the energetics to define anisotropy constants^[5] and to relate these constants to experimental observables.^[6] First-principles theorists^{[7][8][9]} utilize band-structure techniques to calculate total energies in order to evaluate the energetics due to the spin-orbit interaction. The band-structure results are still at the stage where discussions of numerical accuracy can lead to lively controversies.^[10] Another aspect of

theoretical investigation explores the consequences of the surface magnetic anisotropy in influencing other fundamental properties, such as the magnitude of the Curie temperature T_C in ultrathin films, or the critical thickness d_c at which the easy axes reorient from vertical ($\perp$) to in-plane ($\parallel$). At d_c the effective anisotropy goes to zero because the vertical surface anisotropy and in-plane shape anisotropy balance out each other. It is believed that in such a situation it should be impossible to sustain long-range ferromagnetic order. This is because the Mermin-Wagner theorem^[11] has shown that for an isotropic Heisenberg model there is no finite-temperature ordering in two dimensions (2D). Bander and Mills^[12] and Bruno^[13] have investigated the T_C behavior recently. The spin-wave behavior^[14] and instabilities that occur in the spin-wave spectrum near the reorientation transition have also been explored.^[15] Efforts to incorporate surface roughness into the modeling are also in progress.^[16]

Experimental efforts have focused on both quantifying the magnetic anisotropy constants and on searching for systems and conditions that support vertical easy axes. An overview of experimental activities is provided in the reference listings that accompany the torsion magnetometry studies of Elmers *et. al.*^[17] Other techniques that have been used to analyze anisotropy constants include Mössbauer spectroscopy,^{[18][19]} ferromagnetic resonance,^{[20][21]} and Brillouin light scattering,^[22] as well as micromagnetic modeling^[23] to interpret images taken by means of scanning electron microscopy with polarization analysis (SEMPA). Tabulations of approximate anisotropy constants appear in many of the works referred to above, as well as in a paper devoted to the subject by Bruno and Renard.^[24]

The sphere of interest expands as one acknowledges the relationship between surface and interface anisotropy. Then studies in the superlattice field help in the cross-pollination of ideas with the monolayer magnetism field.^[25] Some recent examples include magnetic anisotropy studies of epitaxial superlattices,^[26] of ultrathin amorphous

multilayers,^[27] and of vertical recording media based on rare-earth-transition-metal amorphous alloy films^[28] or multilayers.^[29]

In addition to efforts to quantify anisotropy constants, the direct observation of vertical easy axes also has become an exciting endeavor. For example, SEMPA has been used to observe a continuous reorientation transition that takes place as a function of Co thickness for epitaxial Co/Au(111) films.^[23] Spin-polarized secondary-electron spectroscopy has been used to discover a reversible reorientation transition ($\perp \leftrightarrow \parallel$) that occurs as a function of temperature for ultrathin fcc Fe grown on Cu(100).^[30]

Most of the remainder of the present paper will be devoted to highlighting results for ultrathin iron films that exhibit interesting behavior related to their surface magnetic anisotropy. The examples are based on surface magneto-optical Kerr effect (SMOKE) studies carried out in the author's laboratory. The interest to the microscopy community is that when strong surface anisotropies are found to dominate the physical properties, the goal then becomes to correlate the magnetic properties with the atomic structure, microstructure and morphology. This is where there are great future opportunities and challenges for diverse types of microscopists.

II. Background Information:

Before proceeding with the examples some background information is in order. The films to be considered are grown epitaxially onto single crystal substrates in ultrahigh vacuum (UHV) of 10^{-10} - 10^{-11} Torr. The growth method, known as molecular beam epitaxy (MBE), utilizes techniques standard to the surface science community. Auxilliary characterization probes used throughout the studies are Auger electron spectroscopy to monitor surface cleanliness and growth mode characteristics, and low-energy electron diffraction (LEED) to monitor surface structural order. The above conditions and tools constitute the minimal requirements for meaningful studies of surface phenomena. For a

broad overview of the experimental techniques used in surface and thin-film magnetism research the reader is referred to the articles by Falicov *et al.*^[31] and by Bader.^[32]

SMOKE is the adaptation of the conventional magneto-optical Kerr technique to study ultrathin, or monolayer-range, signals *in situ*. A beam of linearly polarized He-Ne laser light enters the growth chamber through a UHV window and reflects off of the magnetized sample. The magneto-optic interaction, which also is due to the spin-orbit interaction, rotates the polarization of the light and introduces an ellipticity. The reflected light reemerges through the UHV window, passes through a polarizing analyzer crystal whose fast axis is almost crossed with that of the incident light, and the resultant signal is detected by a photodiode. All of the optical components are outside of the UHV chamber; the only component located inside the chamber is a conventional electromagnet used to magnetize the sample. The orientation of the applied magnetic field H is either in-plane or perpendicular to the plane of the film. The resultant configuration is referred to as the longitudinal or polar Kerr effect, respectively. The polar effect is the considerably stronger one, which is part of the reason for its importance in vertical recording applications. A hysteresis loop is generated as H is swept and the output of the detected light changes. The Kerr intensity measures the height of the loop, is proportional to the magnetization, and can be studied directly or used to obtain rotation or ellipticity values. Typically, the maximum possible polar Kerr rotation value is ~ 0.1 - 0.2 mrad for an Fe monolayer. Easy axes can be identified by the shape of the hysteresis loop. Loops for which the remanent Kerr signal is comparable to the saturation value are characteristic of field alignments along easy axes. For a review of the SMOKE technique the reader is referred to the article by Bader.^[33]

III. Vertical Easy Axes in Ultrathin Magnetic Films

In this section four systems will be highlighted and discussed in the light of the opportunities they offer for further study. The studies identify critical thickness values for vertical easy axes. Figure 1 shows the thickness dependence of the Kerr intensity and

coercivities H_c for epitaxial Fe films grown on the indicated substrates. Strong polar Kerr signals initially increase with thickness until d_c is reached, after which a relatively weak longitudinal Kerr signal is detected. The d_c values are in the 3-6 monolayer (ML) range. The coercivities tend to peak in the monolayer range, which is another manifestation of the strong surface anisotropy. A most interesting aspect of the results summarized in Fig. 1 is that, although in each case Fe films of similar thickness are under investigation, the systems are quite diverse. Bcc-Fe(100) grows on Au(100) because of the lattice match between bcc-Fe and the primitive square unit cell of unreconstructed Au(100). This unit cell is rotated 45° from the fcc unit cell of Au. In contrast to this behavior, we find that relatively strain-free fcc-Fe grows on Cu(100). Fcc-Fe exists in the Fe phase diagram at high temperatures T (910-1390°C). Both ferromagnetic and antiferromagnetic (AF) forms of fcc-Fe have been found to exist experimentally and theoretically.^[34] Our epitaxially-stabilized fcc-Fe is clearly of the ferromagnetic variety; it was grown at low T (~ 100 K) to suppress interdiffusion,^[35] as was the Fe/Au(100).^[36] It is now becoming accepted that the AF phase of fcc-Fe can be grown on Cu(100) at *high* T ($\sim 200^\circ\text{C}$) stabilized by an intermixed Fe-Cu buffer layer at the interface.^[37] The most recent structural studies indicate that the ferromagnetic phase is slightly tetragonally distorted, while the AF phase is undistorted.^[38] Evidence has been found in LEED studies for a low- T surface reconstruction^[39] for films grown near room temperature, but correlations have not been pursued yet with magnetic properties.

The interesting question for microscopists is to what extent the d_c values correlate with the thickness at which misfit dislocations set in to relieve strain. The role of strain in stabilizing vertical easy axes in thin epitaxial films is also an important question to pursue. To provide a sense of the strength of the surface magnetic anisotropy in these systems, note that the shape anisotropy corresponds to a field of $\sim 20\text{kOe}$ per layer of Fe; at d_c the field associated with the surface anisotropy balances out this field. If $d_c = 5$ ML, the surface anisotropy field is $\sim 50\text{kOe}$, since there are two surface or interface layers.

Cu and Au are good choices as substrates because their d-bands are filled, so they should not electronically perturb the Fe d-electrons greatly. However, for this same reason they have low surface free energies relative to Fe. Thus, the growth process is unstable with respect to intermixing and segregation.^[40] For this reason transition metal substrates were also explored in Fig. 1. Fe grows pseudomorphically on Pd(100), but because of the lattice mismatch there is a strong tetragonal distortion and the resultant structure is bct.^[41] Thus, a new structure is created via epitaxy that does not exist in the bulk phase diagram. Additional interest in Fe/Pd(100) is that the interfacial Pd is ferromagnetically polarized,^[42] which leads to higher than expected T_C values in the ultrathin regime,^[43] and to the observation of ferromagnetic hysteresis well into the submonolayer dosage regime.^[44] The submonolayer morphology appears to consist of a random-site occupancy, rather than of a 2D island structure.^[45]

The Fe/Ru(0001) results in Fig. 1 differ in a number of ways from those of the other systems. Firstly, the substrate in this case is the basal plane of a hexagonal material. The pseudomorphy would suggest the growth of Fe(111) planes that are expanded in-plane and, therefore, expected to be trigonally compressed, rather than fcc or hcp. However, Tian *et al.*^[46] have recently proposed, based on their LEED studies, that only the first monolayer is pseudomorphic, and that thicker films consist of 3D bcc-Fe(110) domains in two equivalent Kurdjumov-Sachs orientations. Secondly, vertical easy axes are found for room-temperature growth,^[47] while low-T growth was used in the other cases in Fig. 1. This is presumably due to the reduced tendency towards interfacial mixing relative to that for the other systems under consideration. Finally, the first two monolayers of Fe neither yield a ferromagnetic signal nor extrapolate to be ferromagnetically active. It has been reported that Fe/Ru superlattices also have two Fe dead layers at each interface. These results have led to a variety of theoretical speculations,^{[48][49]} including that at least the first Fe monolayer is antiferromagnetically ordered.^[50]

It would be interesting to follow the growth of any of the above systems by means of a scanning tunneling microscope (STM). This would seem to be feasible, as borne out by recent successes in studying the growth of Cu and Au on Ru(0001),^[51] and of Fe on Au(111).^[52]

IV. Summary:

Interest in surface and interface magnetism has increased in recent years. There are many basic challenges as well as possibilities for applications in the magnetic recording field. In the present work a review was presented highlighting recent activities in the field. Examples of vertical easy axes taken from SMOKE studies in the author's laboratory were presented and discussed within the context of future research directions. It is anticipated that the field will continue to attract attention and that studies based on the use of diverse microscopy techniques will help the field to mature.

Acknowledgment

This work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under contract #W-31-109-ENG-38. I thank my colleagues C. Liu and Z. Qiu for helpful discussions.

References:

- [1] J. H. Van Vleck, Phys. Rev. 52 (1937) 1178.
- [2] L. Néel, J. de Phys. Rad. 15 (1954) 225.
- [3] U. Gradmann and J. Mueller, Phys. Status Solidi (b) 27 (1968) 313; U. Gradmann, Appl. Phys. 3 (1974) 161; W. Kümmerle and U. Gradmann, Solid State Commun. 24 (1977) 33; Phys. Stat. Sol. (a) 45 (1978) 171; U. Gradmann, J. Magn. Magn. Mater. 6 (1977) 173; and U. Gradmann and H. O. Isbert, J. Magn. Magn. Mater., 15-18 (1980) 1109; U. Gradmann, J. Magn. Magn. Mater., 54-57 (1986) 733.
- [4] T. Kaneyoshi, in Physics of Magnetic Materials, edited by W. Gorzkowski, M. Gutowski, H. K. Lachowicz and H. Szymczak (World Scientific, Singapore, 1991) p.29.
- [5] F. Gautier, Mater. Sci. Forum 59 & 60 (1990) 361.
- [6] G.T. Rado, in Physics of Magnetic Materials, edited by W. Gorzkowski, M. Gutowski, H. K. Lachowicz and H. Szymczak (World Scientific, Singapore, 1991) p.3.
- [7] J. G. Gay and R. Richter, Phys. Rev. Lett. 56 (1986).
- [8] C. Li, A. J. Freeman, H. J. F. Jansen, and C. L. Fu, Phys. Rev. B 42, (1990) 5433.
- [9] P. Bruno, Phys. Rev. B 39, (1989) 865.
- [10] W. Karas, J. Noffke, and L. Fritsche, J. Chem. Phys. 85 (1989) 861.
- [11] N.D. Mermin and H. Wagner, Phys. Rev. Lett. 17 (1966) 1133.
- [12] M. Bander and D. L. Mills, Phys. Rev. B 38 (1988) 12015.
- [13] P. Bruno, in Magnetic Thin Films, Multilayers and Surfaces, edited by S. S. P. Parkin, Mater. Res. Soc. Symp. Vol. 231 (MRS, Pittsburgh, 1991) in press.
- [14] P. Bruno, Phys. Rev. B 43 (1991) 6015.

- [15] R. P. Erickson and D. L. Mills, "Magnetic instabilities in ultrathin ferromagnets", unpublished preprint.
- [16] P. Bruno, J. Phys. F 18 (1988) 1291; J. Appl. Phys. 64 (1988) 3153.
- [17] H. J. Elmers, T. Furubayashi, M. Albrecht and U. Gradmann, J. Appl. Phys. 70 (1991) 5764.
- [18] U. Gradmann, J. Korecki, and G. Waller, Appl. Phys. A 39 (1986) 101.
- [19] N. C. Koon, B. T. Jonker, F. A. Volkening, J. J. Krebs, and G. A. Prinz, Phys. Rev. Lett. 59 (1987) 2464.
- [20] G. A. Prinz, G. T. Rado, and J. J. Krebs, J. Appl. Phys. 53 (1982) 2087; J. J. Krebs, P. Lubitz, A. Chaiken, and G. A. Prinz, J. Appl. Phys. 69 (1991) 4795.
- [21] B. Heinrich, J. F. Cochran, M. Kowalewski, J. Kirschner, Z. Celini, A. S. Arrott, and K. Myrtle, Phys. Rev. B 44 (1991) 9348.
- [22] B. Hillebrands, P. Baumgart, and G. Güntherodt, Phys. Rev. B 36 (1987) 2450.
- [23] R. Allenspach, M. Stampanoni, and A. Bischof, Phys. Rev. Lett. 65 (1990) 3344.
- [24] P. Bruno and J.-P. Renard, Appl. Phys. A 49 (1989) 499.
- [25] T. Shinjo, Surf. Sci. Reports 12 (1991) 49.
- [26] B. N. Engel, C. D. England, R. A. Van Leeuwen, M. H. Wiedmann, and C. M. Falco, Phys. Rev. Lett. 67 (1991) 1910.
- [27] R. J. Hicken, G. T. Rado, G. Xiao, and C. L. Chien, , Phys. Rev. Lett. 64 (1990) 1820.
- [28] H. Fu, M. Mansuripur, and P. Meystre, Phys. Rev. Lett. 66 (1991) 1086.
- [29] Z. S. Shan and D. J. Sellmyer, Phys. Rev. B 42 (1990) 10433; Z. S. Shan and D. J. Sellmyer, S. S. Jaswal, Y. J. Wang, and J. X. Shen, Phys. Rev. B 42 (1990) 10446.
- [30] D. P. Pappas, K.-P. Kämper, and H. Hopster, Phys. Rev. Lett. 64 (1990) 3179.

- [31] L. M. Falicov, D. T. Pierce, S. D. Bader, R. Gronsky, K. B. Hathaway, H. J. Hopster, D. N. Lambeth, S. S. P. Parkin, G. Prinz, M. Salamon, I. K. Schuller, and R. H. Victora, *J. Mater. Res.* 5 (1990) 1299.
- [32] S. D. Bader, *Proc. IEEE*, 78 (1990) 909.
- [33] S. D. Bader, *J. Magn. Magn. Mater.* 100 (1991) in press.
- [34] For references to the literature see S. D. Bader and E. R. Moog, *J. Appl. Phys.* 61 (1987) 3729; and F. J. Himpsel, *Phys. Rev. Lett.* 67 (1991) 2363.
- [35] C. Liu, E. R. Moog, and S. D. Bader, *Phys. Rev. Lett.* 60 (1988) 2422.
- [36] C. Liu and S. D. Bader, *J. Vac. Sci. Technol. A* 8 (1990) 2727.
- [37] W. A. A. Macedo and W. Keune, *Phys. Rev. Lett.* 61 (1988) 475.
- [38] H. Magnan, D. Chandesris, B. Villette, O. Heckmann, and J. Lecante, *Phys. Rev. Lett.* 67 (1991) 859; *Surf. Sci.* 251/252 (1991) 597.
- [39] W. Daum, C. Stuhlmann, and H. Ibach, *Phys. Rev. Lett.* 60 (1988) 2741; H. Landskron, G. Schmidt, K. Heinz, K. Müller, Stuhlmann, U. Beckers, M. Wuttig, and H. Ibach, *Surf. Sci.* 256 (1991) 115.
- [40] K. Sano and T. Miyagawa, *Jpn. J. Appl. Phys.* 30 (1991) 1434; 30 (1991) 1434; and "Initial stages of deposition-concurrent surface segregation in Fe/Au epitaxial multilayers on GaAs(001)", *Proc. MRS Fall Mtg.* (1991) Boston, in press. Also, see Bader and Moog in Ref. 34.
- [41] J. Quinn, Y. S. Li, H. Li, D. Tian, F. Jona, and P. M. Marcus, *Phys. Rev. B* 43 (1991) 3959.
- [42] S. Blügel, M. Weinert, and P. H. Dederichs, *Phys. Rev. Lett.* 60 (1988) 1077; S. Blügel, B. Drittler, R. Zeller, and P. H. Dederichs, *Appl. Phys. A* 49 (1989) 547.
- [43] K. J. Strandburg, D. W. Hall, C. Liu, and S. D. Bader, "Monte-Carlo simulations of the Curie temperature of ultrathin ferromagnetic films", unpublished preprint.
- [44] C. Liu and S. D. Bader, *J. Appl. Phys.* 67 (1990) 5758.

- [45] C. Liu and S. D. Bader, Phys. Rev. B 44 (1991) 2205.
- [46] D. Tian, H. Li, F. Jona, and P. M. Marcus, "Study of the growth of Fe on Ru(0001) by low-energy electron diffraction', unpublished preprint.
- [47] C. Liu and S. D. Bader, Phys. Rev. B 41 (1990) 553.
- [48] J. Kübler, Solid State Commun. 72 (1989) 631.
- [49] D. Knab and C. Koenig, J. Magn. Magn. Mater. 98 (1991) 10.
- [50] R. Wu and A. J. Freeman, Phys. Rev. B 44 (1991) 4449.
- [51] G. Pötschke, J. Schröder, C. Günther, R. Q. Hwang, and R. J. Behm, Surf. Sci. 251-252 (1991) 592.
- [52] B. Voigtländer, G. Meyer, and N. M. Amer, Surf. Sci. Lett. 255 (1991) L529.

Figure Caption:

Figure 1 Critical thicknesses d_c to sustain vertical easy axes of magnetization (left panels) and the corresponding coercivities (right panels) are shown for epitaxial Fe films grown on the indicated substrates. The information is derived from the height and width, respectively, of the magnetic hysteresis loops obtained using the surface magneto-optic Kerr effect. The strong polar Kerr signals (square symbols) indicate vertical easy axes, and the weak longitudinal Kerr signals (triangles) indicate easy axes that are in the film plane.

END

**DATE
FILMED
5/13/92**