Optical Properties of Magnetic Surfaces, Interfaces, Thin Films, Overlayers and Superlattices

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A brief general review of tlie optical properties of magnetic surfaces, interfaces, thin films, overlayers and superlattices is preseiited. It includes general background material, aiid some cf the latest developments in tlie field.

I. Introduction

Magnetis n is an electronically driven phenomenon, and therefore susceptible to study by optical means. It is however a weak phenomenon, as compared with electrostatic m d electric-dipole effects, and much more difficult to detect. It is also very subtle in its manifestations. It origins are purely quantum mechanical, with its foundations in the Pauli Exclusion Principle and tlie existence of tlie electron spin. It leads nonetheless to sliort- and loiig-range forces, aiid to both classical and quantum-ineclianical effects. As a consequence tliere is a rich variety of textures aiid properties found in magnetic systems

It is also possible to alter these systems drastically by means of small changes in temperature, by changes in the specific chemistry and morphology of tlie samples, and by relatively modest forces. such as tliose arising from magnetic fields, and uniform and non-uniform stresses. Tlierefore magnetic systems lead to a large number of useful engineering and technical applications.[1–5]

II. Magneto-optics

Tlie first rragneto-optic effect was reported by Faraday in the middle of the XIX century. He discovered that (in general) elliptically polarized light passing tlirough a magnetized, transparent substance (i) changes the ellipticity and (ii) rotates tlie major axes of its polarizat ion.

About 30 years later, in 1876, Kerr reported an analogous plienomenon in reflection. The Kerr effect is observable in the majority of ferromagnets, wliicli are seldom transparent and therefore not suitaùle for tlie Faraday effect. In fact Kerr only reported a rotation in tlie plane of polarization, but did not notice tliat tliere was also a change in tlie ellipticity of tlie reflected liglit. Kerr's explanation of tlie effect was that tlie magnetization of tlie ferromagnet gives rise to a component of tlie electric-field vector of the liglit wliicli oscillates in a direction perpendicular to tlie plane of vibration of tlie incident beain (and in phase with it). If tlie incoming light is polarized in tlie plane of incidence (p-polarization), tlie reflected liglit lias its strong component in tlie same plane because of the laws of ordinary reflection. However there is another component, in tlie reflected beain, polarized in tlie plane of the surface (s-polarizatioii), perpendicular to tlie first one. If, as Kerr assumed, tlie two coinponents are in phase, simple vector addition would give a polarization wliicli is also linear, but with a polarization direction rotated by an angle Θ (the Kerr angle) from the p-direction. In fact tlie two coinponents are not in pliase, and the reflected light is thus elliptically polarized. In a typical experiment the main axis of the ellipse is rotated by a few minutes of arc from tlie original orientation (of tlie order of 10 ininutes for the longitudinal Kerr effect, and about 40 minutes for tlie polar Kerr effect), and tlie ellipticity (ratio of the minor to the major axis of the ellipse for light originally linearly p-polarized) is of tlie order of 0.001. These are all very small but easily detectable effects.

Given that there are many vectors involved in the problein, the possible geometrical arrangements of the experiinent are inany. In particular tlie following vectors are relevant:

Incident light: Propagation vector k; Instantaneous polarization vector ϵ perpendicular to k.

Reflected light: Propagation vector **k';** Instantaneous polarization vector ϵ' perpendicular to **k'.**

Reflecting magnetic surface: Unit vector normal to tlie surface ai; Direction of the magnetization M.

According to tlie elementary lnws of reflection, **k,** k' and **n** are all three in one plane, the plane of iiicideiice:

because of the *laws of reflection* **n** is always parallel to **(k'** - k),

because of the laws of *reflection* **n** is always perpendicular to **(k'** + **k).**

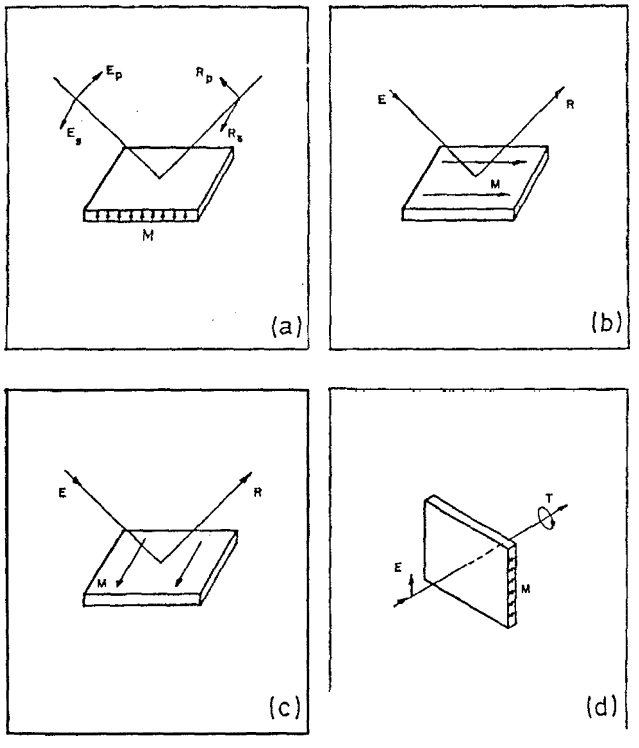

Figure 1: Tlie arrangement of tlie various vectors in tlie magneto-optic experiments. Tlie various letters indicate: E: tlie incident beam; R: tlie reflected beam; T: the transmitted beam; M: tlie sample magnetization. (a) Tlie Polar Kerr effect;(b) Tlie Longitudinal Kerr effect; (c) Tlie Transverse Kerr effect; (d) Tlie Faraday effect.

Tlie Polar *Kerr Effect* corresponds to tlie case in which M is parallel to n, i.e., tlie magnetization is normal to tlie surface and also in tlie plane of incidence:

POLAR ICERR EFFECT: M lies parallel to n.

In tlie Longitudinal *Kerr Effect* tlie magnetization M is perpendicular to **n** (i.e., it lies in the surface plane) and is in the plane of incidence:

LONGITUDINAL ICERR EFFECT: M lies perpendicular to n,

LONGITUDINAL KERR EFFECT: M lies parallel to **(k'** + k).

In tlie *Transverse Kerr Effect* the magnetization is once again perpendicular to **n** (i.e.. it also lies ill the surface plane) and is also perpendicular to tlie plane of incidence:

TRANSVERSE KERR EFBECT: M lies perpendicular to **n,**

TRANSVERSE KERR EFFECT: M lies perpendicular to **(k'** + k).

All these arrangements are shown in Figure 1.

Since tlie interaction between electric and magnetic fields is largest when tlie two vectors are perpendicular to each otlier, tlie Kerr rotation is maximum whenever E and M are perpendicular to one another (e.g., the polar and longitudinal effects for s—polarization), sizeable when tliey form a not-very-small angle, aiid negligible when ϵ; ϵ aiid M are parallel or almost parallel.

Even though both magneto-optic effects (Faraday aiid Kerr) were first discovered and studied in tlie second lialf of the XIX ceiitury, tliey are both now enjoying a renaissance as tools in basic aiid applied research. On tlie basic-research side it lias been recently deinonstrated tliat tlie Kerr effect can be iiscd to detect monolayer and even submonolayer magnetism.[6] In tlie applications area the effects can be useful in connection witli the commercial potential of materials for high-density magneto-optical data storage.[7] In addition, recent developments in Merr microscopy to image magnetic domains aiid to observe magnetic-switching phenomena have also lielped revitalize tlie classic field of micromagnetics.[8,9]

Tlie Surface Magneto-Optic Kerr Effect (SMOKE) provides a valuable in situ characterization probe of tlie magnetic and magneto-optic properties of magnetic films during tlie growth process.[10] Since tlie magneto-optic coupling is caused by the spin-orbit interaction it is indeed a very small effect. Tlie technique requires the application of an external magiietic field to reverse tlie magnetization direction of tlie sample in tlie growtli chambers. All other parts, including tlie optical components, are outside tlie vacuum system. Typically tlie system consists of[11] a laser source, a polarizing analyzer, and a photodiode detector (see Figure 2). Magnetic hysteresis curves are obtained by monitoring the light intensity at tlie detector as tlie field is swept. To address key issues associated witli the surface magnetic anisotropy, tlie field can be in tlie film plane (longitudinal and transverse Kerr effects) or perpendicular to it (polar Kerr effect). Tlie temperature dependence of tlie hysteresis loops can be used to monitor tlie magne-

tization and coercivity. Tlie Kerr effect was also used to obtain the magnetization exponent β in the critical regime for the system Fe/Pd (100), and a good agreement was found witli tliat expected tlieoretically ($\beta = 0.125$) for a two-dimensional Ising system[12]. Tlie Kerr effect can be used as well to monitor tlie Curie temperature as a function of tliickness, which provides a fundamental cliaracterizatioii paraineter of tlie films of interest.

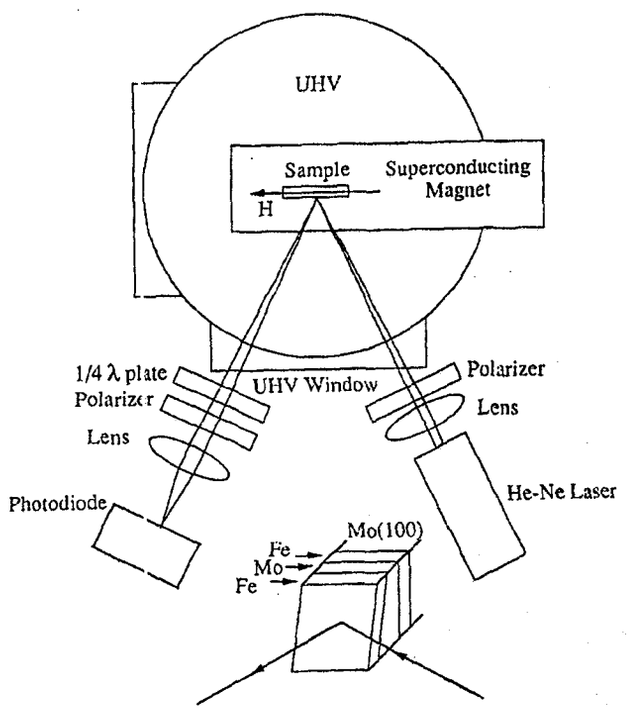

Figure 2: Schematic experimental arrangement for the Surface Magneto-Optic Kerr Effect (SMOKE) on a trilayer sample of Fe/Mo/Fe. (Figure courtesy of S. D. Bader. See reference [11])

It sliould be possible in the very near future to use tunable photon sources (synclirotron sources) in tlie optical-frequency region to monitor tlie Kerr rotation of magnetic monolayers and ultra-tliin, metastable phases. Tliis iòrm of the Kerr spectroscopy will provide electronic striictural information in tlie form of a joint density of states weiglited by tlie magneto-optic niatrix elements.[13] The spectral information sliould complement that obtained from k-dependent probes of the band structure, such as angle-resolved, spin-polarized photoemissior .

It should be emphasized that the Kerr effect is not an inherently surface-sensitive probe. The optical penetraticn deptli in metals is between 100 and 200 Å, and tlierefore tlie probe goes many atomic layers into tlie bulk. Tlie surface sensitivity, necessary to study surface and iiiterface magnetism, is derived from the sainple fabrication tecliniques that create extremely thin epitaxial inagnetic films. It is of interest to use compleinentary techniques witli different probing deptlis to understand coupled magnetic layers, for iiistance, It sliould be possible to develop tlie Kerr effect into such a probe by using non-linear optical processes; surface sensitivity will be obtained by monitoring tlie Kerr rotation in the Second-Harmonic Generation (SIIG) mode.[14,15] Tlie SHG teclinique has receiitly gained prominence as an advanced surface-analysis technique.[16]

III. Light-scattering techniques

Brillouin liglit scattering (inelastic liglit scattering by low-frequency excitation modes witli well defined energy and wavevector) lias also proven valuable to obtain information about tlie magnetization, and the excliange and anisotropy constants. Tlie scatteriiig modes (bosonic excitations created and destroyed) in tliis case are tlie spin waves (magnons). Tliese studies can be performed *in situ* on overlayers,[17] or as a post-growth cliaracterization tool on superlattice and sandwicli structures[18,19] in air or in controlled liigli- or low-teinperature environments. The information obtained is quantitative and cross-correlates well with other techniques, sucli as ferromagnetic resonance data.[20]

IV. Angle-resolved photoemission spectroscopy

Even witliout resolution of the spin polarization of the emitted electron, angle-resolved photoelectron spectroscopy (ARPES) is a very powerful tool to study the magnetic properties of valence electrons in solids. Peaks in ARPES are produced by direct (vertical) transition, in wliich tlie k-vectors in tlie initial (before plioton absorption) and final (after photon absorption) electron states are conserved (within a vector of tlie reciproca1 lattice in the crystal). From tlie measured energy and angle of the emitted electron, and given that the frequency of tlie radiation is known, one can determine uniquely the energy of tlie initial state, and the component of its k-vector in the plane of the emitter's surface. Since spin-up and spin-down electrons have different band structures in polarized materials, the detection of specific features in the photoemission spectrum

– witli well determined eiiergy aiid surface components of tlie k-vector – permit comparison witli tlieoretical calculations hased oii various magiietic structures, tlie elimination of many non-compatible models, and tlie determinatioii (in some cases) of tlie correct structure.

A successful example of tlie application of this technique is tlie study of tlie (100) surface of antiferromagnetic Cr. It was known from theoretical studies[21–23] that tlie (100) free surface of aatiferromagnetic Cr should order itself il1 tlie forin of alteriiating ferromagnetic planes. Tlie surface outer plane lias a inagnetic moinent considerably eiilianced from its bulk value [between (+2.4) and ($3.0) Bolir magnetons, compared with a bulk value of 5.59 Bolir magnetons]. Tlie second layer, whicli is ferroinagnetic but aligned antiparallel to the outer one, has a niagnetic moment of about -1.5 Bolir magnetoiis; tlie tliird layer a moment of +1.0 Bohr magnetons. Such a stroiig magiietic structure (wliicli is, on tlie whole, antiferromagnetic in nature but – because of only partial compensation between successive layers – has a net surface magnetization) yields well defined features in tlie band structure, in particular surface electronic states that are concentrated either on tlie surface or on tlie secoiid atomic layer.

Analysis of tlie ARPES data[24] confirms tlie existente of two sharp features. Feature 1, at an energy 0.08 eV below tlie Fermi level, lias a k-vector wliose surface-parallel component lies at tlie center of tlie two-dimensional (surface) Brillouiii zone; polarizatioii and photon-energy analysis indicates that it originates froin states witli tlie so-called Δ_1 syminetry in tlie bulk. Feature 2, at an energy 0.63 eV below tlie Fermi level, lies also at tlie center of tlie two dimensional surface Brillouiii zone, but originates froin bulk states with Δ_5 syminetry. These features can be put in a one-to-one correspondence to tliose predicted by theory.[21–23] It is highly unlikely that any other magnetic structure could give rise to features whicli would also agree tliat well witli tlie experimental data.

Only recently, however, lias tlie ferroinagnetic character of tlie (100) surface of antiferromagnetic cliroinium been observed directly. Tlie difficulty witli tlie direct observation is tlie existence, in all real surfaces, of surface steps. Witli each step, one monolayer in heiglit, a different terrace is exliibited. Since alternating terraces have opposite magnetizations (a consequence of tlie antiferromagnetic arrangeineiit of bulk Cr), there is in fact no net magnetization at tlie stepped surface. Tlie effect was nonetheless observed by ineans of a double Scanning Tunneling Microscopy (STM) experiment.[25] When a (100) chromium surface was observed witli an STM witli a tungsten tip, many identical steps (all of 1.4 Å lieiglit) were recorded. Tlie lieiglit corresponds exactly to one lialf of tlie 2.8 Å of the cubic parameter in body-centered cliroinium. A secoiid cxperiinent, in wliicli tlie tip was made of chromium dioxide (a magnetic semimetal in whicli only one spin orientation can tunnel either in or out), produccd alternating steps of 1.2 Å aiid 1.6 Å lieiglit, proving tliat for electroiis of a given spin (tliose that can tunnel in or out of tlie chromium dioxide tip) consecutive terraces present different tunneling probabilities, i.e., terraces with alternating ferromagnetic arrangements.

V. Spin-polarized photoemission spectroscopy

Direct information on tlie ferroinagnetic electronic structure at surfaces can be obtained by means of spin-polarized photoemission studies. Early studies[26] measure tlie polarization of tlie photo-yield as a function of photon energy, without energy analysis. Such measurements have tlie advantage that they can be performed as a function of applied magnetic field perpendicular to tlie surface up to tlie magiietic saturation of tlie sainple.

Synchrotron radiation, witli its liigli intensity, permits energy analysis of tlie electrons photoemitted from a material magnetized in tlie plane of tlie surface.[27] A inovable spin and eiiergy analyzer allows investigation along different directions of k-space. One can tlius obtain a complete mapping of tlie spin-dependent electronic band structure over tlie entire Brillouin zone.[28]

With higli-iiitensity vacuum ultra-violet and soft x-ray sources otlier investigations become possible. Studies of surface shifts in shallow core levels, such as the $4f$ levels in rare earths, allow one to distinguish varying magnetization as tlie surface is approached.[29] With x-ray photoemission spectroscopy tlie polarization of electrons emitted from inultiplet split core levels, sucli as tlie 3s or $3p$ level iii Fe, gives element-speciiic magiietic information,[30] similar to tliat obtained from polarized Auger spectroscopy but easier to interpret. Froin these studies it is also possible to extract quantitative values of atoinic magnetic moments at surfaces.[31]

VI. Polarized Auger spectroscopy

Auger electron spectroscopy is a powerful tool for surface analysis because of its surface sensitivity and

its chemical-element specificity. In tlie case ferromagnets tlie Auger electrons may also be spin polarized. Such polarization arises from tlie different occupation of tlie spin split valence-conduction band. When tlie electrons at tlie top of tlie Fermi distribution ill a ferromagnet are directly involved in tlie Auger emission process, tlit emitted electrons are naturally polarized. For core electrons, on tlie otlier liaiid, tliere also may be polarized emission because of tlie excliange interaction between tlie valence-electron spin density and tlie filled core levels.

Through spin-polarized Auger spectroscopy one obtains an element- specific probe of tlie local magnetization at a given site. It can provide information not only on tlie magnetic properties of a surface but, in films of a few atomic layers, on tlie inagnetic properties of substrates and interfaces.

A classic study of this kind[32] has determined univocally tlie magnetization of a monolayer of Gd evaporated on an Fe(100) crystal surface. Tlie spin polarization of tlie Auger electrons corresponding to tlie **R** $[M(23)M(45)M(45)]$ line lias opposite polarization to that corresponding to the Gd $[N(45)O(23)N(67)]$ and Gd $[N(45)N(67)N(67)]$ lines, indicating tliat tlie magnetic momeiits in tlie Gd overlayer are coupled so as to lie in a direction aiitiparallel to those in tlie Fe substrate. In tl e same study it was possible to measure independently tlie teinperature dependence of tlie magnetization of the Gd layer and Fe interface layers taking advantage of tlie elemental specificity of tlie Auger process.

VII. Magnetic superlattices

Metallic magnetic superlattices[1] are made by depositing, in an orderly fashion and witli clean interfaces, alternating metallic thin films of two or more chemical compositions, at least one of which is magnetic. These systems exliibit a wide range of very interesting new physical phenomena, in particular magneto-optics, magnetoresistance, magnetostriction, magnetostatics, inagnetic excliange coupling, unusual microwave properties, and anisotropic magnetic beliavior.

A variety cdprocedures have been used to grow tliese superlattices. Tlie preferred metliods have been sputtering, evaporstion, molecular beam epitaxy, and chemical vapor deposition. A typical superlattice consists of a sometimes complex substrate e.g., gallium arsenide, magnesium oxide, strontium titanate, copper, molybdenum, followed by a repetition (a given number of times, between one and several hundred) of a tliickiiess x of a ferromagnetic metal (e.g., iron, cobalt, permalloy) and a tliickness y of a non-magnetic metal (e.g., copper, chroiniuin, molybdenum). Tlie non-magnetic metal is usually called tlie spacer. Two closely related effects appear in tliese systems:

1. Successive ferromagnetic layers arrange themselves witli their magnetization either parallel or antiparallel to eacli otlier (see Figure **3).** Tlie (parallel or antiparallel) magnetic arrangement is a function of tlie tliickiiess and nature of tlie iiitervening non-ferromagnetic metal and tlie quality and structure of tlie interfaces. For a given system, prepared iii a systeinatic way, tlie coupling is an oscillatory function of tlie tliickness of tlie spacer[33,34] (see Figures below[11]).

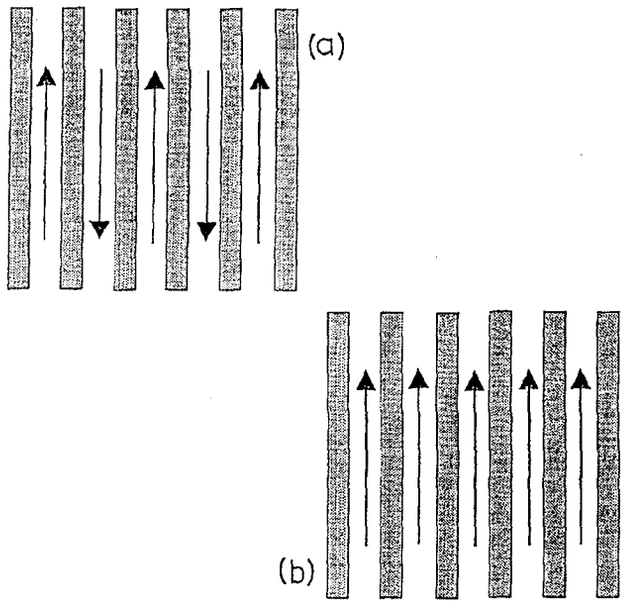

Figure 3: Schematic representation of a magnetic superlattice. In (a) successive magnetic layers are arranged witli their magnetizations antiparallel to eacli otlier; tlie electrical resistance is high. In (b) tliere is a parallel magnetization arrangement; tlie resistance is low. If tlie application of a magnetic field changes (a) into (b) one obtains a *negative magnetoresistance.*

2. Tlie second effect takes place in samples in wliicli tlie inagnetic alignment is antiparallel. The application of a strong enougli magnetic field changes

tlie arrangement of the magnetization. The antiferromagnetic coupling is overcoine, and tlie magnetic moments of all ferromagnetic layers are forced to lie in the same direction. A macroscopic magnetic moment develops. Simultaneously tlie electrical resistaiice of tlie superlattice, in all directions, decreases.

The change in the magnetic structure as a function of the stiucture of tlie sample and tlie applied magnetic field lias been moiiitored by optical (SMOKE, Brillouin scattering) and non-optical (neutron and electron scattering techniques, ferromagnetic resonance) means.

SMOKE experiments provide in situ characterization of the samples.[11] By monitoring tlie Kerr angle as a function of the applied magnetic field, one determines the Iiysteresis loops of tlie various samples (tlirough a derivative property). Samples with parallel arrangement exliibit a simple, single loop, similar to those found in ordinary, bulk ferroinagnets. Samples with antiparallel arrangements in the absence of an applied field exliibit a double loop, corresponcling to tlie three possible configuration: $\{...,\uparrow,\uparrow,\uparrow,\uparrow,...\}$ (for strong magnetic fields up); $\{...,\uparrow,\downarrow,\uparrow,\downarrow,...\}$ (for weak fields); and $\{...,\downarrow,\downarrow,\downarrow,\downarrow,...\}$ (for stroiig magnetic fields down).

(See Figures 4, 5, and 6.)

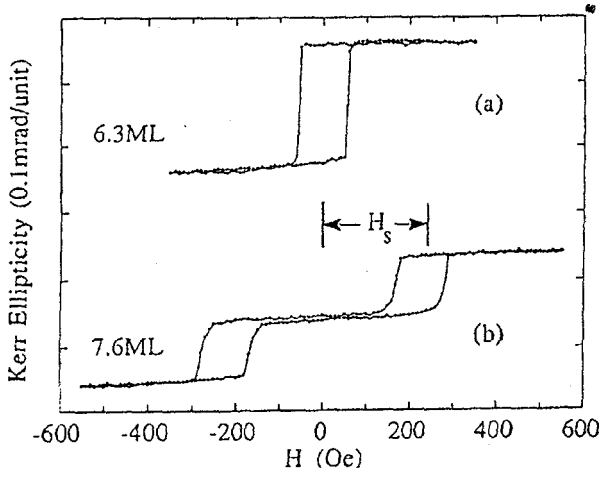

Figure 4: Ferromagnetic and antiferromagnetic liysteresis loops in trilayer samples of Fe/Mo/Fe, with varying thickness of tlie Mo spacer. The data were taken with tlie SMOKE technique. See Figure 2 for the experimental arrangement. (Figure courtesy of S. D. Bader. See reference [11]).

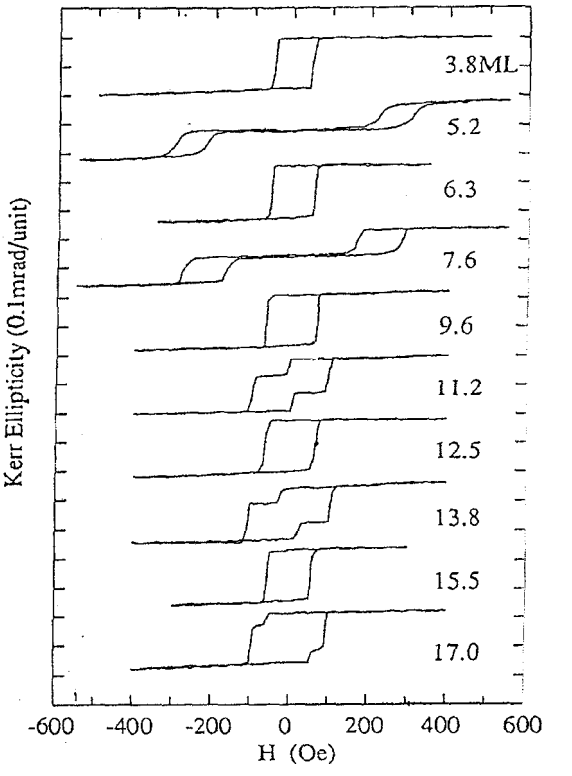

Figure 5: Hysteresis loops measured by SMOKE teclinique in trilayer samples of Fe/Mo/Fe, as a function of tlie tliickness of tlie Mo spacer. The data were taken witli the SMOKE technique. The parallel (single loop) and antiparallel (double loop) arrangements are an oscillatory function of the tliickness. (Figure courtesy of S. D. Bader. See reference [11]).

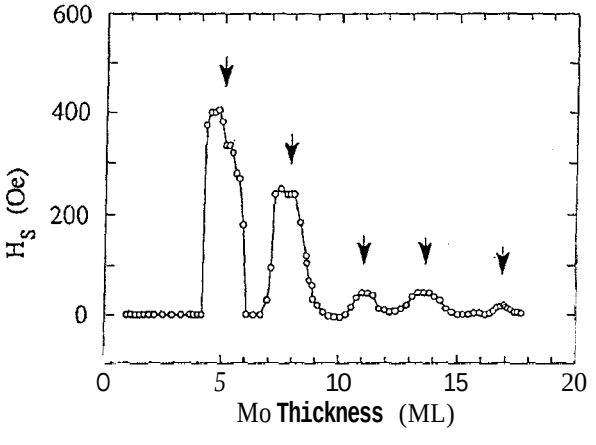

Figure 6: Oscillations in tlie switching field, as defined in Figure 4, as a function of the Mo spacer tliickness in Fe/Mo/Fe trilayers. Finite switching fields indicate antiparallel arrangement of tlie two Fe layers. Zero switcliing fields are an indication of parallel arrangement of the Fe layers. (Figure courtesy of S. D. Bader. See reference [11]).

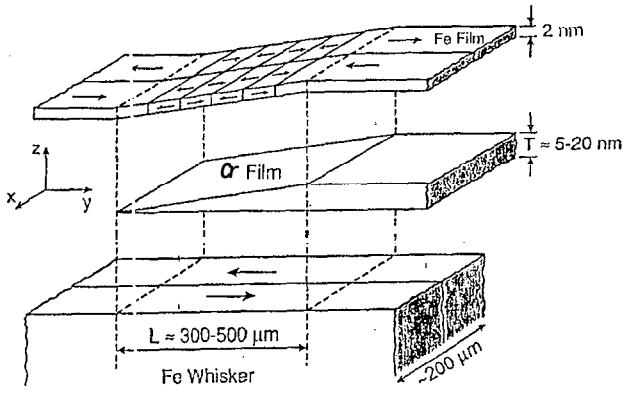

Figure 7: Exploded scliematic view of a wedge sample of an Fe/CrlFe trilayer. Tlie arrows in tlie Fe indicate the direction of magnetization in eacli domain. Tliere are two domains in the Fe whisker substrate, and many domains in tlie overlaying (top) Fe film. The coupling betweeii tlie Fe wliisker and tlie Fe film – parallel or antiparallel – is a function of tlie thickness of tlie Cr spacer. It is also a function of the rougliness of tlie Fe/Cr interfaces. Note that tlie vertical and horizontal scales are very different. Tlie actual angle of tlie wliisker was in fact 0.001 degrees. (Figure courtesy of R. J. Celotta and D. T. Pierce. See reference [35]).

A prominent technical success of tlie last three years is the growtl; of a spacer wedge whose thickness varies continuously across tlie sample.[35] Typically, its thickness varies between 0 and 20 Å over a distance of 0.5 mm. Such a sliallow wedge permits tlie conversion of rather poor horizontal resolution into vertical resolutions in the atomic scale or even better. Wedge samples have been prepared with a variety of elements under a large variety of growth conditions. It should be noted that on the scale of the wedge, it is in fact a succession of "broad" terraces, eacli one atomic layer higher than tlie next as shown in Fig. 7.

Acknowledgments

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