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Probing Interfacial and Bulk Magnetism: A Magneto-Optical Comparative Analysis

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ABSTRACT

The magnetic properties of materials are fundamentally determined by their atomic structure and electronic configuration. In thin films and nanostructures, surface and interfacial effects can profoundly alter the magnetic behavior compared to the bulk material, leading to unique phenomena relevant for spintronics and magnetic recording. This article conceptually investigates the disparities between surface and bulk magnetization, with a particular emphasis on how magneto-optical techniques can effectively differentiate and characterize these distinct magnetic responses. We explore the principles of magneto-optical effects, such as the Kerr effect, which are inherently sensitive to the surface and near-surface magnetic moments. The proposed methodology involves utilizing these techniques to probe the magnetic hysteresis loops and reversal mechanisms at the surface, in comparison with bulk magnetic measurements (e.g., SQUID magnetometry). By examining the conceptual outcomes related to phenomena like exchange bias and training effects, we aim to elucidate how interfacial coupling can create asymmetry and shifts in the surface hysteresis, distinct from the underlying bulk. This comparative analysis provides critical insights into the microscopic origins of magnetism at interfaces, offering a powerful framework for designing advanced magnetic materials with tailored surface functionalities.

KEYWORDS

Interfacial magnetism, bulk magnetism, magneto-optical analysis, Kerr effect, magnetic thin films, spin dynamics, magnetic domains, comparative study, optical characterization, ferromagnetic materials.

INTRODUCTION

Magnetism, a fundamental force of nature, manifests across various length scales, from atomic moments to macroscopic domains. The magnetic properties of a material are dictated by its electronic structure and the collective behavior of its constituent spins. While the "bulk" properties of a material represent the average behavior within a large volume, the magnetic characteristics at the surface and interfaces can differ significantly, especially in thin films, multilayers, and nanostructures [1, 2]. These deviations arise due to factors such as reduced coordination of atoms, broken symmetry, strain, intermixing, and unique electronic states at the boundary, leading to phenomena distinct from the interior [16, 17]. Understanding these surface and interfacial magnetic properties is not merely an academic pursuit but is crucial for advancing technologies in areas like magnetic data storage, spintronics, and magnetic sensors [1, 2].

One of the most prominent interfacial magnetic phenomena is exchange bias (EB) [1, 2]. Exchange bias occurs in systems comprising a ferromagnetic (FM) layer coupled to an antiferromagnetic (AFM) layer [1, 2, 8]. When such a

system is cooled below the Néel temperature of the AFM in the presence of an applied magnetic field, a unidirectional anisotropy is induced. This manifests as a shift in the magnetic hysteresis loop of the FM layer along the magnetic field axis and, often, an increase in its coercivity [1, 2, 16]. The magnitude of this shift is known as the exchange bias field (HEB), while the increase in coercivity is reflected in the coercive field (HC) [20, 21]. Early work by Nogués and Schuller (1999, 2005) laid the foundation for understanding exchange bias, highlighting its dependence on various parameters, including the interface quality, AFM bulk properties, and external conditions like temperature and annealing fields [1, 2, 12, 18]. Recent studies continue to explore robust exchange bias phenomena in systems like Co/CoO films for flexible spintronics [7] and isotropic exchange bias in twinned epitaxial Co/Co3O4 bilayers [5].

While bulk characterization techniques like Vibrating Sample Magnetometry (VSM) or Superconducting QUantum Interference Device (SQUID) magnetometry provide macroscopic insights into the integrated magnetic response of a material, they often lack the surface sensitivity required to disentangle the contributions from the surface/interface versus the bulk. This is where magneto-optical (MO) techniques become indispensable. Magneto-optical effects, such as the Magneto-Optical Kerr Effect (MOKE), involve the change in polarization or intensity of light reflected from a magnetized surface [3]. These effects are inherently sensitive to the magnetic state of the material within the optical penetration depth of the light, which is typically on the order of tens of nanometers, thereby providing a powerful tool for probing surface and interfacial magnetism without interference from the deeper bulk [3].

The objective of this article is to conceptually investigate the distinct magnetic behaviors of surfaces/interfaces compared to the bulk, and to highlight the unique capabilities of magneto-optical techniques in discerning these differences. We will explore how MO measurements can reveal asymmetries, shifts, and other specific features in surface hysteresis loops that are indicative of interfacial magnetic phenomena like exchange bias, contrasting them with bulk measurements. By synthesizing theoretical understanding and anticipated experimental observations, this study aims to provide a comprehensive framework for understanding and leveraging the subtle yet profound influences of surfaces and interfaces on the overall magnetic properties of advanced materials.

METHODS

This study is a conceptual investigation drawing upon established principles of magnetism, materials science, and magneto-optical phenomena. The "materials" refer to the types of samples typically investigated in such studies, and the "methods" delineate the experimental and analytical techniques employed to probe and compare surface and bulk magnetic properties.

Sample Systems

For a comprehensive comparative analysis of surface versus bulk magnetization, ideal sample systems would be those where interfacial effects are significant and controllable. Thin films and multilayer structures are particularly suitable.

- Ferromagnetic (FM) Thin Films: Single-layer ferromagnetic films (e.g., Co, FeNi, Fe) deposited on non-magnetic substrates would be used to establish a baseline for thickness-dependent magnetic properties and to understand the intrinsic surface anisotropy. The surface properties of these films, due to reduced symmetry and altered electronic structure, can deviate significantly from their bulk counterparts [23].
- Exchange-Biased Bilayers: The most illustrative systems are ferromagnetic/antiferromagnetic (FM/AFM) bilayers (e.g., CoO/Co, FeNi/FeMn, CoFe/IrMn) [4, 6, 7, 9, 10, 16]. These systems are explicitly designed to exhibit strong interfacial magnetic coupling, leading to the exchange bias phenomenon [1, 2, 9, 10, 16]. The choice of AFM

(e.g., CoO, IrMn, FeMn) and its specific properties (e.g., ordering temperature, uncompensated spins) are critical for the strength and nature of the exchange bias [9, 10, 12, 16, 19].

o Controllable Parameters: The deposition methods (e.g., sputtering, molecular beam epitaxy) would allow control over film thickness, interface roughness, and crystallographic orientation [4, 5]. Post-deposition treatments such as annealing (e.g., annealing in a magnetic field below the Néel temperature) [18] or ion irradiation [13, 14, 15] can be used to tailor the interface and induce or modify the exchange bias.

Magneto-Optical Kerr Effect (MOKE) Measurements

MOKE is the primary technique for probing surface and near-surface magnetism. It exploits the interaction of linearly polarized light with a magnetized material, resulting in changes in the light's polarization state (rotation of the plane of polarization and change in ellipticity) upon reflection [3, 23]. The magnitude of the Kerr rotation and ellipticity is directly proportional to the magnetization component perpendicular to or parallel to the plane of incidence, depending on the MOKE geometry.

- Experimental Setup: A typical MOKE setup involves:
- 1. Light Source: A monochromatic light source (e.g., a laser) to ensure well-defined optical properties.
- 2. Polarizer: To produce linearly polarized incident light.
- 3. Electromagnet/Helmholtz Coils: To apply a magnetic field to the sample, allowing for the measurement of hysteresis loops.
- 4. Sample Stage: Mounted in a cryostat for temperature control (e.g., for cooling through the Néel temperature to establish exchange bias) [3].
- 5. Analyzer: To analyze the polarization state of the reflected light.
- 6. Photodetector: To measure the intensity of light after passing through the analyzer.
- MOKE Geometries: Different MOKE geometries (longitudinal, transverse, polar) are sensitive to different components of magnetization and provide varying probing depths. Longitudinal MOKE (magnetic field in the plane of the film, parallel to the plane of incidence) and polar MOKE (magnetic field perpendicular to the film plane) are commonly used to measure in-plane and out-of-plane magnetization, respectively.
- Surface Sensitivity: The shallow optical penetration depth (typically 10-20 nm for visible light in metals) ensures that MOKE primarily probes the magnetic properties of the surface or the interface of thin films. This intrinsic surface sensitivity makes MOKE ideal for distinguishing surface from bulk magnetism [3, 23].
- Hysteresis Loop Measurement: By sweeping the applied magnetic field and simultaneously measuring the Kerr signal, the magnetic hysteresis loop of the probed region (surface/interface) can be obtained. This allows for the determination of surface coercive fields (HC), exchange bias fields (HEB), and saturation behavior [3, 23, 24].

Bulk Magnetization Measurements

To provide a comparative baseline for the entire sample volume, standard bulk magnetometry techniques would be employed.

- Vibrating Sample Magnetometer (VSM): A VSM measures the magnetic moment of the entire sample by vibrating it in a uniform magnetic field and detecting the induced voltage in pickup coils. VSM provides the macroscopic magnetic hysteresis loop, yielding bulk HC, HEB, and saturation magnetization (MS).
- Superconducting QUantum Interference Device (SQUID) Magnetometer: A SQUID offers highly sensitive

measurements of magnetic moment over a wide range of temperatures and magnetic fields. It is particularly useful for low-moment samples or for characterizing temperature-dependent magnetic properties like blocking temperatures in exchange bias systems [18, 24].

Data Analysis and Comparison

The acquired hysteresis loops from both MOKE and bulk magnetometry would be analyzed and compared.

- Extraction of Parameters:
- o Coercivity (HC): The field at which magnetization becomes zero.
- o Exchange Bias Field (HEB): The shift of the hysteresis loop center from zero field [1, 20, 21].
- o Saturation Magnetization (MS): The magnetization at high fields where the sample is fully magnetized.
- o Remanent Magnetization: The magnetization remaining after the field is reduced to zero.
- Training Effect: The reduction of HEB and HC upon successive cycling of the magnetic field would be investigated for exchange-biased samples [24]. The training effect is a signature of the reorientation of AFM spins at the interface.
- Comparison of Loops: Direct overlay and quantitative comparison of MOKE and VSM/SQUID hysteresis loops would reveal differences in HC, HEB, and loop shapes between the surface/interface and the bulk.
- Temperature Dependence: Measurements across a range of temperatures would reveal the blocking temperature of the AFM layer (above which exchange bias vanishes) and how the surface and bulk properties evolve with temperature [18].

This dual-technique approach ensures a comprehensive understanding by providing both surface-sensitive and volume-averaged magnetic information, allowing for a precise differentiation of interfacial and bulk magnetic phenomena.

RESULTS

Based on the proposed magneto-optical and bulk characterization methodology, and drawing from the rich literature on thin film magnetism and exchange bias, the following conceptual results are anticipated:

Baseline: Ferromagnetic (FM) Single Thin Films

- MOKE vs. VSM/SQUID: For FM single thin films, MOKE and VSM/SQUID hysteresis loops would generally show similar coercive fields and squareness, assuming the film thickness is within the optical penetration depth of MOKE or if the bulk properties largely reflect the surface [23].
- Thickness Dependence: As the film thickness increases beyond the MOKE penetration depth, MOKE would continue to probe primarily the surface/near-surface region, while VSM/SQUID would integrate the magnetic response over the entire film volume. Any inherent surface anisotropy or dead layers could cause subtle differences in saturation behavior or effective coercivity between the two measurements.

Exchange-Biased Bilayers (FM/AFM)

This is where the most significant and illustrative differences between surface and bulk magnetization are expected.

• Observation of Exchange Bias (EB) and Coercivity Enhancement: Both MOKE and VSM/SQUID measurements on field-cooled FM/AFM bilayers (e.g., Co/CoO [3, 4, 6, 7, 16], FeNi/FeMn [13, 14, 15]) are expected to show clear signs of exchange bias:

- o Horizontal Shift (HEB): The center of the hysteresis loop will be shifted away from zero magnetic field, indicating a unidirectional anisotropy induced by the FM-AFM interface.
- o Coercivity Enhancement (HC): The width of the hysteresis loop (coercivity) will be significantly larger compared to an uncoupled FM film of the same thickness.
- Asymmetry of Magnetization Reversal: MOKE measurements are particularly sensitive to the asymmetry of magnetization reversal caused by exchange bias [3, 23]. This means the pathways of magnetization reversal (e.g., nucleation and propagation of domains) might differ significantly for the ascending versus descending branches of the hysteresis loop. While also present in bulk, MOKE's surface sensitivity can resolve subtle interfacial effects on this asymmetry.
- Differences in HEB and HC between Surface (MOKE) and Bulk (VSM/SQUID):
- o Surface-Dominated HEB: The exchange bias field (HEB) is fundamentally an interfacial phenomenon [1, 2]. Therefore, MOKE measurements, which probe the interface directly, are expected to provide a highly accurate and sometimes larger HEB value compared to VSM/SQUID, especially if the AFM layer has a complex domain structure or if the FM layer exhibits non-uniform magnetization across its thickness.
- o Coercivity Discrepancies: The coercive field (HC) measured by MOKE might differ from that measured by VSM/SQUID. These differences can arise if the magnetic reversal mechanisms at the surface/interface are distinct from those in the bulk. For instance, if pinning at the interface is stronger than in the bulk, the MOKE HC might be higher. Conversely, if the bulk is more strongly pinned by defects or grain boundaries, the VSM/SQUID HC could be larger.
- o Loop Shape Differences: Subtle variations in the shape of the hysteresis loops (e.g., squareness, slope of the branches) would be observed between MOKE and VSM/SQUID, reflecting the distinct contributions of surface/interface vs. bulk spins to the overall magnetization reversal process.
- Training Effect: Both MOKE and VSM/SQUID measurements on exchange-biased systems are expected to exhibit the training effect [24]. Upon successive cycling of the magnetic field, both HEB and HC will typically decrease before saturating at lower values. This phenomenon is attributed to the irreversible reorientation of frustrated spins within the AFM layer near the interface [1, 2, 24]. MOKE's ability to monitor this training effect specifically at the interface offers critical insights into the microscopic dynamics of the AFM spin structure that drives exchange bias.
- Temperature Dependence: The blocking temperature (TB) of the AFM layer, below which exchange bias is observed, can be precisely determined by tracking HEB with increasing temperature for both MOKE and VSM/SQUID. The values should largely coincide, but subtle differences might reflect interfacial versus bulk AFM ordering [18, 22].

These conceptual results demonstrate the power of magneto-optical techniques in providing unique, surface-sensitive insights into complex magnetic phenomena like exchange bias, complementing and enriching the volume-averaged information obtained from bulk magnetometry.

DISCUSSION

The conceptual results presented underscore the critical importance of distinguishing between surface/interfacial and bulk magnetic properties, particularly in advanced magnetic thin films and nanostructures. Magneto-optical techniques, especially MOKE, emerge as indispensable tools for this differentiation, providing insights that are inaccessible to conventional bulk magnetometry. The observed disparities in hysteresis loop characteristics between MOKE and VSM/SQUID measurements, especially in exchange-biased systems, are profound and carry

significant physical implications.

The distinct behavior observed at the surface/interface compared to the bulk fundamentally stems from the broken translational symmetry and reduced coordination of atoms at the material's boundary. At an interface, atoms experience different local environments, leading to altered exchange interactions, magnetic anisotropy, and even spin configurations. For instance, the uncompensated spins in an antiferromagnetic (AFM) layer, crucial for establishing exchange bias, primarily reside at the interface or within a few atomic layers [1, 2, 9]. MOKE's shallow optical penetration depth allows it to directly probe these interfacial spins, providing a more accurate representation of the active magnetic elements driving phenomena like exchange bias [3, 23].

The observation of a potentially larger exchange bias field (HEB) via MOKE compared to VSM/SQUID is consistent with the interfacial nature of EB. Bulk measurements average the response over the entire film, which may include regions not contributing to the bias or even regions with opposing effective biases, thereby diluting the measured effect [12]. MOKE, by focusing on the active interface, can capture the maximal local HEB induced by the AFM layer. The asymmetries in magnetization reversal observed by MOKE [3] further highlight that the energy barriers and mechanisms for reversing spins are different for the two field directions due to the unidirectional anisotropy at the interface. This provides crucial information for understanding the spin configurations and defect structures within the AFM that pin the FM magnetization [17].

The training effect, a signature phenomenon in exchange bias systems, is also particularly informative when studied with surface sensitivity. The gradual reduction of HEB and HC upon repeated field cycling reflects the dynamic reorientation of uncompensated AFM spins at the interface, which initially contribute to the bias but become less rigidly pinned with repeated reversals [1, 2, 18, 24]. MOKE allows for a direct observation of these interfacial spin dynamics, providing experimental evidence for models like the domain state model proposed by Miltényi et al. (2000) [9] or more recent descriptions of relaxation phenomena [18]. Understanding the microscopic origins of the training effect is crucial for device applications where magnetic stability under cycling is required.

Practical implications for magnetic materials design are substantial. By quantitatively differentiating surface from bulk properties, researchers can:

- Optimize Interface Engineering: Tailor deposition conditions, annealing protocols [18], or ion irradiation parameters [13, 14, 15] to engineer specific interfacial spin configurations and enhance desired magnetic properties like stronger exchange bias or reduced training.
- Develop Advanced Spintronic Devices: Devices like spin valves and magnetic tunnel junctions rely heavily on interfacial magnetic phenomena. MOKE provides a direct means to characterize and optimize the performance of these nanoscale interfaces.
- Understand Thermal Stability: The interplay between blocking temperature (TB) and the exchange bias magnitude, as revealed by temperature-dependent MO measurements, is vital for designing thermally stable magnetic components [18, 22].

Limitations and Future Directions:

While powerful, MOKE has its limitations. Its optical penetration depth, though shallow, is not atomically precise; it averages over several atomic layers. Techniques like X-ray magnetic circular dichroism (XMCD), which can be element-specific and surface-sensitive to an even finer degree, could complement MOKE for a more atomic-scale understanding of interfacial magnetism. The interpretation of MOKE signals in complex multilayers can also be challenging due to interference effects.

Future research could explore:

- Time-Resolved MOKE: Investigating the ultrafast dynamics of magnetization reversal at surfaces and interfaces [3].
- Micro-MOKE: Probing magnetic domains and their interactions at high spatial resolution.
- Combined Techniques: Integrating MOKE with other surface-sensitive techniques (e.g., photoemission electron microscopy with XMCD contrast) for a multi-modal view of interfacial magnetism.
- Theoretical Modeling: Developing more sophisticated theoretical models that accurately predict the MOKE response based on detailed interfacial spin structures and compare them with experimental data.

In conclusion, the ability to directly compare and contrast surface and bulk magnetization using magneto-optical and bulk techniques provides a holistic understanding of magnetic materials. This differentiation is not just an academic nuance but a fundamental requirement for designing next-generation magnetic devices that leverage precise control over interfacial magnetic phenomena.

CONCLUSION

This article has conceptually underscored the critical distinction between surface/interfacial and bulk magnetization in magnetic materials, emphasizing the invaluable role of magneto-optical techniques in discerning these differences. Through a proposed comparative methodology utilizing both surface-sensitive Magneto-Optical Kerr Effect (MOKE) measurements and volume-averaged bulk magnetometry, we outlined how key magnetic parameters like coercive field, exchange bias field, and the training effect manifest uniquely at interfaces.

The anticipated results highlight that MOKE measurements can reveal more pronounced exchange bias fields and distinct magnetization reversal asymmetries at the surface, directly reflecting the crucial role of interfacial spin configurations. This precise differentiation provides fundamental insights into how broken symmetry and unique atomic environments at boundaries dictate magnetic behavior. Understanding these disparities is not only pivotal for advancing the theoretical understanding of magnetic phenomena but also holds immense practical significance for the design and optimization of advanced magnetic thin films and nanostructures for spintronic devices, data storage, and other cutting-edge technologies. The continued integration of highly sensitive surface characterization techniques is essential for harnessing the full potential of interfacial magnetism.

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