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Critical focused issues

Role of the antiferromagnetic bulk spins in exchange bias

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ABSTRACT

This "Critical Focused Issue" presents a brief review of experiments and models which describe the origin of exchange bias in epitaxial or textured ferromagnetic/antiferromagnetic bilayers. Evidence is presented which clearly indicates that inner, uncompensated, pinned moments in the bulk of the antiferromagnet (AFM) play a very important role in setting the magnitude of the exchange bias. A critical evaluation of the extensive literature in the field indicates that it is useful to think of this bulk, pinned uncompensated moments as a new type of a ferromagnet which has a low total moment, an ordering temperature given by the AFM Néel temperature, with parallel aligned moments randomly distributed on the regular AFM lattice.

1. Introduction

Exchange bias (EB) is characterized by the shift of the magnetic hysteresis loop along the field axis, generally observed in Antiferromagnetic (AFM)/Ferromagnetic (FM) bilayered hybrids [1]. This interesting, basic research effect is also the basis for many applications in the spintronics area such as magnetic data storage and sensor devices. The essential characteristics which determine the properties of an exchange biased system are: the magnitude of the shift, its sign, asymmetry of the hysteresis loop, blocking temperature (above which the EB disappears), training effect and time dependence. Although much work has been dedicated to understand the phenomenology of EB [2], each one of these important characteristics presents interesting puzzles, which give complementary clues regarding the essential physics of the effect. EB is generally considered to be a consequence of the interfacial interaction between the FM and AFM constituents [3]. This is attributed to the pinned, uncompensated magnetic moments [1,4–6] at the interface originating from the AFM.

Originally it was postulated that only the AFM interface controls the EB, i.e. EB is a purely interfacial phenomenon in which the role of the AFM bulk is restricted to pinning the interfacial magnetic moments. However, the interface is always coupled to the AFM bulk. Therefore the AFM bulk may affect the precise magnetic state of the interface with the consequent effect on the exchange bias. There is by now much compelling evidence that the bulk magnetic state of the AFM may affect the exchange bias, which implies that EB is not a purely interfacial phenomenon. Although its ultimate origin is the exchange interaction at the AFM/FM interface, the pinned, uncompensated spin distribution at the interface might be determined by the AFM bulk. In this "Critical Focused Issue" we highlight the role and microscopic origin of the pinned, uncompensated moments (PUM) present in the bulk of the AFM. More specifically, we emphasize the important experiments, which provide clues regarding the microscopic mechanism that governs exchange bias. We conclude by describing potential new directions in which this field can move and connected open questions.

EB is initiated by cooling the FM/AFM bilayer in an externally applied magnetic field below the AFM Néel temperature. The exchange coupling between the FM and the AFM, which shifts the hysteresis loop along the field axis, is determined by an effective "exchange field" or by a "unidirectional" anisotropy energy. The AFM crystallinity, its morphology (e.g., grains) and intrinsic anisotropy are crucial parameters which determine the magnitude of the EB. In general, two types of exchange-biased systems, which show distinctly different behavior, can be distinguished. Type 1 are highly textured or epitaxial systems such as FeF_2 or CoO. On the

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other hand, type 2 are usually small-grained, polycrystalline systems, such as the classic archetypes IrMn or FeMn. It should be noted that the anisotropy energy, the central quantity determining the EB magnitude, depends on both the effective anisotropy constant as well as the crystal volume. Consequently, some polycrystalline AFMs may behave as either type 1 or type 2 depending on the crystallite size, the inter-crystallite magnetic coupling, which may lead to a larger effective particle volume, and the microstructure (e.g., growth mode), which may yield an effective increase of the anisotropy constant [7,8]. It is also important that the properties of the exchange bias bilayers are not only determined by the AFM's physical structure, but also its magnetic structure. Even if the crystallographic orientation of the AFM/FM interface is well defined, the spin orientation may become very complicated since in some cases equivalent crystallographic directions may not be magnetically equivalent. For instance, NiO is a classic example in which the (111) crystallographic plane has 4 structurally equivalent, but magnetically inequivalent directions.

Type 1 systems have in general a large exchange bias, the blocking temperature coincides with the AFM Néel temperature, and training and time dependences are practically absent. In type 2 systems the blocking temperature can be considerably reduced compared to the Néel temperature, and they may exhibit large training as well as time dependent effects. The change from negative (NEB) to positive (PEB) exchange bias shift can be present in both types of systems if the exchange coupling at the interface is antiferromagnetic and the surface layer of the AFM couples to the increasing external cooling field [9,10]. Like with many other situations in physics, there is no clear demarcation between type 1 and type 2 systems; these are just two extreme cases. For instance, there may be situations in which the blocking temperature coincides with the AFM Néel temperature but the systems exhibit large training effects [11–13]. This may occur even within the same combination of materials, since sometimes there are large structural differences within the same system. A classic example is Co/CoO where the CoO may be polycrystalline, textured, or epitaxial depending on the specific preparation method.

There are a number of additional extrinsic experimental complications, which may cause confusion. Sometimes the exchange bias is much smaller than the coercivity, the hysteresis loops are sheared and/or there are large contributions from other (presumably irrelevant) parts of the sample such as substrates. In either case, small shifts along the field axis may be caused by artifacts such as vertical loop shifts and may complicate the identification of the EB. Other important issues which are not discussed here include intrinsic and extrinsic effects such as interfacial roughness, interdiffusion, variation in thickness and reduced magnetization and/or formation of interfacial compounds at interfaces and surfaces. Of course, in order to avoid erroneous conclusions the physical and chemical properties of these systems must be thoroughly characterized quantitatively using a comprehensive battery of tests.

2. Issues

In spite of all the above-mentioned complications, it seems that a single physical mechanism determines the exchange bias. There is overwhelming evidence that the origin of EB resides in the pinned, uncompensated moments (PUM) present in the AFM. The only possible exceptions are interfaces with sizeable Dzyaloshinskii–Moriya interaction [14], which breaks mirror symmetry and may lead to EB at perfectly compensated interfaces. Moreover, it is crucial that in addition to the PUM there is evidence for the presence of unpinned, uncompensated moments (UUM), which do not influence the EB, but may affect the coercivity [15]. It should also be mentioned that the presence of intentionally introduced non-magnetic sites (impurities and/or defects) may affect the domain state (and consequently the PUM) of the AFM as theoretically implied by the “Imry–Ma argument” [16]. It refers to the statistical imbalance of the number of impurities on the two AFM sublattices within any *finite* region, leading to a net AFM magnetization.

The following issues arise naturally regarding the PUM: (a) Do they reside on the surface and/or in the bulk of the AFM? (b) What is their microscopic origin? And (c) do bulk spins/moments play any role? Generally it is assumed that EB is a purely interfacial effect in which bulk moments provide the pinning matrix for the interfacial PUM. However, recent experiments show clear evidence that bulk AFM spins/moments play an active role in determining the EB features.

In this “Critical Focused Issue” we will discuss this particular important characteristic and highlight unanswered questions that are still open for further research. We will not discuss the role of PUM at the AFM/FM interface as this has been extensively done in previous articles [17–20]. We will focus on pure type 1 and type 1-like AFM systems, in which thermal fluctuations play a minor role due to the high anisotropy, epitaxial nature, large grain size, relevant inter-gain coupling and/or low enough measurement temperature. This also excludes systems in which training and other history dependent phenomena may be associated with metastable magnetic structures, e.g. spin glasses, present in the bulk of the AFM [21]. On the other hand, we exclude pure type 2 materials consisting of small, uncoupled AF grains as, e.g., IrMn. Their behavior has been addressed by a phenomenological model based on thermal activation of AFM grains with distributed grain sizes to explain loop shifts, training and changes in coercivity with temperature [22]. In this particular case the microscopic mechanism for the EB and the key role that PUM play is still elusive.

3. Experimental evidence

In this section, we will summarize the different classes of experiments which imply that bulk PUM play a major role in exchange bias.

3.1. Dilution in the bulk

Uncompensated moments were generated intentionally only in the bulk AFM using nonmagnetic defects [23] and keeping the interface the same for all dilutions. Nonmagnetic defects create a statistical imbalance in the ideally equal number of spins in the two sublattices of the AFM. This imbalance results in a net number of uncompensated spins which couple to the external magnetic cooling field. This was accomplished in the strong-anisotropy AFM CoO by diluting the bulk magnetic Co sites with nonmagnetic Mg [23]. The samples were prepared from a ferromagnetic Co layer, grown on (0001)-oriented single crystalline sapphire Al_2O_3 (Fig. 1(a)). To assure that *all samples had an identical interface*, a 0.4-nm thick antiferromagnetic CoO layer containing a nominally minimum defect concentration was then deposited on top of the Co layer. The subsequently deposited epitaxial antiferromagnetic CoO layers were diluted by inserting nonmagnetic Mg substitutions in $Co_{1-x}Mg_xO$ or Co defects in $Co_{1-y}O$. In this fashion, a variable concentration of defects was generated away from the FM/AFM interface, within the volume part of the AFM layer.

Fig. 1(b) shows the dependence of the EB as a function of Mg dilution x in the CoO bulk at different temperatures. The changes in the bulk of the AFM cause major changes of the EB field. For example at 20 K the EB field is enhanced (over the background of about 20 mT) by a factor of three due to 10% nonmagnetic Mg

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