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A deep look into exchange-coupled films: advances through nuclear resonant scattering of synchrotron radiation

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Abstract

We introduce a new method to image the internal spin structure in exchange-coupled thin films. The technique relies on nuclear resonant scattering of synchrotron radiation from ultrathin layers of Mössbauer isotopes embedded in different depths of the sample. The technique is applied to image the spiral spin structure that forms in exchange-spring bilayers in external magnetic fields.

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1. Introduction

The magnetic structure of nanoscale devices plays an increasingly important role in many branches of applied physics and materials science. This is particularly true for the rising field of magnetoelectronics where electronic transport properties are controlled by the spin structure within thin magnetic films. In recent years, one could observe an impressive progress in the development of efficient methods to characterize magnetic properties on the atomic scale. While many of these methods offer a very high lateral resolution they cannot access the magnetic struc-

ture of buried layers and interfaces. This is the domain of scattering methods with polarizationsensitive probes like photons [\[1\]](#page-4-0) and neutrons [\[2\]](#page-4-0) that penetrate into the sample. The availability of high-brilliance synchrotron radiation has recently opened new avenues for the application of X-ray scattering methods in this field [\[1\]](#page-4-0). If the X-ray energy is tuned to a resonance of one of the constituting atomic species, these methods become element specific and the magnetic properties of individual layers can be probed, see, e.g. [\[3\]](#page-4-0). The specificity can be further enhanced if the scattering process exhibits isotopic sensitivity. Magnetic properties are studied with very high spatial resolution using ultrathin layers of a Mössbauer isotope that are embedded in selected depths of the sample. This was applied in a number of studies

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involving conversion electron Mössbauer spectroscopy [\[4–7\].](#page-4-0) The technique has gained new momentum in recent years due to the availability of high-brilliance synchrotron radiation sources [\[8–11\]](#page-4-0).

2. The experimental method

The pulsed time structure of synchrotron radiation suggests to perform hyperfine spectroscopy in the time domain rather than in the energy domain. Hyperfine-split nuclear energy levels (e.g. the hyperfine sextett of $57Fe$ in ferromagnetic materials) are simultaneously excited by the radiation pulses, comparable to hitting a set of slightly detuned tuning forks. Correspondingly, one observes a temporal beating in the time evolution of the nuclear decay that proceeds on a time scale given by the natural lifetime τ_0 of the resonant isotope. In the experiments described here, we have employed the 14.4-keV transition of $57Fe$ with $\tau_0 = 141$ ns. From the analysis of the temporal beat pattern, $I(t)$, the magnitude and the orientation of magnetic fields in the sample can be determined with high accuracy [\[11\].](#page-4-0) The laserlike collimation of synchrotron radiation renders this technique very suitable for the investigation of nanostructures on surfaces, thin films and multilayers.

The incident radiation typically hits the sample under grazing angles of a few mrad, and the reflected signal is recorded by the detector, as sketched in the upper left of Fig. 1. Due to the strong polarization dependence of the scattering process, the time dependence of the reflectivity sensitively depends on the orientation of the magnetic fields in the sample. This is illustrated for some selected cases of a unidirectional magnetization \hat{m} in Fig. 1, for which the characteristic time spectra $I(t)$ are shown. For that reason, ultrathin layers enriched in $57Fe$ can be used to probe the depth dependence of magnetic properties in thin films. This is sketched in Fig. 1. Due to the slope of the probe layer, a certain depth D in the sample is probed by adjusting the transverse displacement Δx of the sample relative to the narrow incident beam.

Fig. 1. Top: Scattering geometry applied to investigate the magnetic spin structure in Fe/FePt exchange-spring bilayers. In the soft-magnetic Fe layer, an isotopic probe layer of 57Fe is embedded. Bottom: For selected orientations of the magnetization \hat{m} relative to the wavevector \hat{k}_0 of the incident photon, the time spectra $I(t)$ of nuclear resonant scattering are shown. $\hat{\sigma}$ and $\hat{\pi}$ are the vectors of the polarization basis.

3. The magnetic structure of exchange-spring bilayers

The exchange coupling between different magnetic materials plays an important role in many micromagnetic systems. For example, nanophase materials consisting of hard-and soft-magnetic phases are promising candidates for new magnetic materials with very high energy products beyond

 1 MJ/m^3 [\[12\]](#page-4-0). Bilayers consisting of hard- and soft-magnetic thin films are ideal model systems to investigate the magnetic coupling between such phases. The exchange interaction leads to a pinning of the soft-magnetic moments at the interface of the hard-magnetic film. With increasing distance from the interface, the exchange coupling becomes weaker and the magnetization may rotate under the action of an external field. If, for example, the external field is applied orthogonally to the magnetization direction of the hard layer, the magnetic moments in the soft layer arrange in a spiral structure along the normal, as shown schematically on the left of [Fig. 1](#page-1-0). Due to the reversible nature of this rotation, this is called the exchange-spring effect [\[13\].](#page-4-0) A number of micromagnetic models have been developed to describe this behaviour [\[14–16\].](#page-4-0) In this article, we apply nuclear resonant scattering from ultrathin probe layers of 57 Fe to directly measure the in-depth magnetic structure of exchange-spring bilayers.

The experiments were performed at the Nuclear Resonance beamline (ID18) of the European Synchrotron Radiation Facility (ESRF) [\[17\]](#page-4-0). The sample was a bilayer consisting of 11 nm softmagnetic Fe on 30 nm hard-magnetic FePt. To minimize oxidation of the Fe layer, it was coated with a 3 nm thick Ag layer. All the layers were prepared by RF-magnetron sputtering in a highvacuum system with a base pressure of $2 \times$ 10^{-7} mbar [\[10\]](#page-4-0). The FePt layer was produced by co-sputtering of Fe and Pt to obtain a composition close to $Fe₅₅Pt₄₅$. Subsequent annealing of this layer resulted in the formation of the hardmagnetic $L1_0$ phase with a coercivity of 0.95 T at room temperature. An ultrathin tilted probe layer of 57Fe was embedded within the Fe layer, as shown in [Fig. 1](#page-1-0). The sample was mounted in a cryomagnet system and exposed to an external field of about 3:5 T to saturate the FePt and introduce a remanent uniaxial magnetization along the direction of the incident wave vector \vec{k}_0 . During the measurements, the sample was cooled to 4:2 K and subjected to variable external fields \vec{H} perpendicular to the beam direction. To measure the depth dependence of the spin rotation, the sample was displaced transversely to the beam. The time spectra taken at various positions Δx are shown in [Fig. 2](#page-3-0) for an external field of $H = 160$ mT. The characteristic changes in the beat pattern reflect the rotation of the magnetization from an almost parallel alignment at $\Delta x = 0$ mm towards increasing perpendicular orientation with increasing Δx . This corresponds to a gradual transition from geometry A to geometry B in [Fig. 1.](#page-1-0) From each of these spectra the in-plane rotation angle ϕ of the magnetization was deduced by a fit of the data (solid lines). The resulting depth dependence of the rotation angle is shown in [Fig. 2](#page-3-0) for external fields of 160, 240 and 500 mT: Dashed lines are simulations according to a one-dimensional micromagnetic model as outlined in Ref. [\[15\],](#page-4-0) assuming the magnetic properties to be homogeneous throughout the Fe layer with an exchange constant of $A = 1.0 \times 10^{-6}$ erg/cm [\[10\]](#page-4-0). However, close to the Fe/Ag interface the simulation clearly deviates from the measured data. This can be explained by a reduction of the exchange constant to $A = 3.0 \times$ 10^{-7} erg/cm in the top 3 nm of the Fe layer. The solid lines are corresponding simulations, yielding a much better description of the experimental data.

The modification of the magnetic properties in the interface region is strongly correlated with the distribution of hyperfine fields in the sample as derived from evaluation of the time spectra. The simulation with a single hyperfine field of 33:8 T leaves significant discrepancies for time spectra that were recorded close to the Ag/Fe interface, as shown in [Fig. 3a.](#page-3-0) However, the assumption of a broad-hyperfine field distribution around 27:0 T leads to a much better simulation of the data, see [Fig. 3b.](#page-3-0) This points to a magnetic compound in the interfacial region with a hyperfine field that is significantly reduced compared to that of bulk Fe. The relative fraction of this compound decreases with increasing depth in the Fe layer, as shown in [Fig. 3c.](#page-3-0) The origin of this compound may be due to intermixing of Fe and Ag during the sputter process or by diffusion of oxygen through the Ag capping layer.

4. Conclusion and outlook

Due to the very high brilliance of modern synchrotron radiation sources, ultrathin isotopic probe layers can be used to directly image the internal spin structure of thin films. This technique was used here to reveal the depth profiles of the

Fig. 3. Effect of the hyperfine field distribution on the measured time spectra. (a) Simulation of the time spectra with a single hyperfine field of 33:8 T; (b) Simulation of the same data with an additional field distribution around 27 T as shown above; (c) Depth dependence of the relative fraction of the corresponding magnetization component.

spin structure within the soft magnetic layer of a Fe/FePt exchange-spring bilayer capped with ag. Due to the very high sensitivity of the method, a depth resolution of the magnetic structure below

Fig. 2. Top: Measured time spectra of grazing incidence reflection in a perpendicular magnetic field of $H = 160$ mT at various lateral positions Δx . The change in the beat pattern reflects the rotation of the magnetization direction as a function of depth D: Solid lines are theoretical simulations from which the rotation angle ϕ , as defined in [Fig. 1,](#page-1-0) was derived. Bottom: Depth dependence of the spin rotation in the Fe layer for external magnetic fields of 160 mT (\bullet) , 240 mT (\bullet) , and 500 mT (\blacksquare) . Lines are simulations according to the model described in [Ref. \[15\]](#page-4-0) with the magnetic parameters given in the text.

1 nm could be achieved. The measurements reveal a transition region at the Fe/Ag interface where the magnetic properties are significantly altered. Moreover, the simulations indicate a rotation of the magnetization in the hard magnetic layer already below the switching field. Furthermore, the technique has been applied to investigate the coupling of ferromagnetic films through nonmagnetic spacer layers [18]. Future applications include the investigation of the spin structure in the hard-magnetic layer and the extension to other elements like Eu, Sm and Dy which are important constituents of new magnetic materials.

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