Electronic Supplementary Information (ESI): From negative to positive Exchange-Bias in dipole coupled trilayers: experiment and theory

Felipe Torres^{1,2}, Rafael Morales^{3,4}, Ivan K. Schuller⁵, Miguel Kiwi^{1,2}

¹Depto. de Física, Facultad de Ciencias, Universidad de Chile, Casilla 653, Santiago, Chile 7800024

²Centro para el Desarrollo de la Nanociencia y la Nanotecnología,

CEDENNA, Avda. Ecuador 3493, Santiago, Chile 9170124

³Department of Chemical-Physics & BCMaterials,

University of the Basque Country UPV/EHU, 48940 Leioa, Spain

⁴IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain. and

⁵Department of Physics and Center for Advanced Nanoscience,

University of California San Diego, La Jolla, California 92093, USA.

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ADDITIONAL EXPERIMENTAL DETAILS

An FeF₂(70nm)/Au(t_{Au})/Ni(30nm)/Al(2nm) wedge-shaped trilayer was fabricated by electron beam evaporation, at a base pressure of 5×10^{-7} Torr. FeF₂ was deposited onto a MgF₂ (110) single crystal at 300°C. The temperature was reduced to 150°C for deposition of Au, Ni and the Al protecting layer. A shadow blade covered progressively the sample during Au growth, in order to obtain the wedge-shaped Au layer, which varies in thickness from $t_{Au} = 0$ to 13 nm. As a consequence a Au wedge with a slope $\Delta t_{Au} = 0.5$ nm/mm is obtained. A schematic representation of the wedge-shaped trilayer is shown in Fig. (1). FeF₂ grows epitaxially on MgF₂ following the same (110) The external magnetic field is always applied along the FM and AFM easy axis. Magneto-optical hysteresis loops are scanned through the Au-wedge by a laser spot of 100 um. Therefore, proving an area with a thickness variation around 0.05 nm orientation. This crystallographic plane exhibits a magnetically compensated spin structure in a bulk single crystal.



FIG. 1: (color-online). FeF₂(70nm)/Au(t_{Au})/Ni(30nm)/Al(2nm) wedge-shaped trilayer for $0 < t_{Au} < 13$ nm. The Au-wedge is orthogonal to both the magnetic easy axes of the system, and the direction of the external field that is applied during cooling and measurements.

THE MODEL

We assume that the largest number of domains imprinted on the AFM, which are responsible for EB, are created during the field cooling process, and that they remain frozen over a large range of external magnetic fields. The spin structure at the FM surface is determined by the the competion of the Zeeman and dipole energies. Let us consider the case for intermediate field cooling, i.e. when the antiferromagnetic dipole coupling through the PM spacer is compensated by the Zeeman energy. The FM/AFM domain interaction energy density E_{int} can then be written as

$$E_{int}(\theta_1, \theta_2) = - K_{\rm FM}[\cos^2(\beta - \theta_1) + \cos^2(\beta - \theta_2)] - M_{\rm FM}\mu_0[\cos(\theta_1) + \cos(\theta_2)]H + E_{dip},$$
(1)

where μ_0 is the vacuum permeability, β is the angle between the applied field (*H*) direction and the anisotropy axis. θ_1 (θ_2) is the angle between FM domain-1 (domain-2). $K_{\rm FM}$ is the uniaxial anisotropy energy density and M_{FM} is the saturation magnetization of the FM. The dipole energy density is given by

$$E_{dip} = -\frac{\mu_0}{4\pi} \left[\frac{\mathbf{M}_{\rm FM}^{(1)} \cdot \mathbf{M}_{\rm AFM}^{(1)}}{t_{\rm PM}^3} + \frac{\mathbf{M}_{\rm FM}^{(2)} \cdot \mathbf{M}_{\rm AFM}^{(2)}}{t_{\rm PM}^3} \right],\tag{2}$$

where, $\mathbf{M}_{\mathrm{FM}}^{(1,2)}$ is the magnetization of FM domain-1 (domain-2), and $\mathbf{M}_{\mathrm{AFM}}^{(1,2)}$ is the magnetization of its closest AFM domain-1 (domain-2). It is further assumed that FM domain-1 and FM domain-2 are initially oriented in the same direction. Thus $\mathbf{M}_{\mathrm{FM}}^{(1)} \cdot \mathbf{M}_{\mathrm{AFM}}^{(1)} = M_{\mathrm{FM}} r_{\mathrm{FM}}^{(1)} M_{\mathrm{AFM}}^{(1)} r_{\mathrm{AFM}}^{(1)} \cos(\theta_1)$, and $\mathbf{M}_{\mathrm{FM}}^{(2)} \cdot \mathbf{M}_{\mathrm{AFM}}^{(2)} = -M_{\mathrm{FM}} r_{\mathrm{FM}}^{(2)} M_{\mathrm{AFM}}^{(2)} r_{\mathrm{AFM}}^{(2)} \cos(\theta_2)$, where $r_{\mathrm{FM}}^{(1,2)}$ is the FM domain size, and the $r_{\mathrm{AFM}}^{(1,2)}$ AFM domain size. Using, $M_{\mathrm{AFM}}^{(1)} = m_{\mathrm{AFM}}/(r_{\mathrm{AFM}}^{(1)})^2$, and $M_{\mathrm{AFM}}^{(2)} = m_{\mathrm{AFM}}/(r_{\mathrm{AFM}}^{(1)})^2$, we obtain

$$E_{dip} = -\frac{\mu_0 M_{\rm FM} m_{\rm AFM}}{4\pi t_{\rm PM}^3} \left[\frac{r_{\rm FM}^{(1)}}{r_{\rm AFM}^{(1)}} \cos(\theta_1) - \frac{r_{\rm FM}^{(2)}}{r_{\rm AFM}^{(2)}} \cos(\theta_2) \right].$$
(3)

Here, the average magnetic moment $m_{\text{AFM}} = \mu_B \sum_{\mathbf{r}} \langle S_{\alpha}^z(\mathbf{r}) - S_{\beta}^z(\mathbf{r}) \rangle \neq 0$, where \mathbf{r} denotes a lattice site, and μ_B is the Bohr magneton. When the cooling field is applied along the easy axis of the AFM, quantum fluctuations of the frustrated spins break the balance between the two magnetic sublattices, and therefore $|\langle S_{\alpha}^z(\mathbf{r}) \rangle| \neq |\langle S_{\beta}^z(\mathbf{r}) \rangle|$, where $|\langle S_{\alpha}^z(\mathbf{r}) \rangle|$, and $|\langle S_{\beta}^z(\mathbf{r}) \rangle|$ are the average magnetic moments of the AFM sublattices. Defining two EB fields, namely,

$$H_{\rm NEB} = H_{\rm EB}^{(1)} = \frac{m_{\rm AFM}}{4\pi t_{\rm PM}^3} \left(\frac{r_{\rm FM}^{(1)}}{r_{AFM}^{(1)}}\right),\tag{4}$$

$$H_{\rm PEB} = H_{\rm EB}^{(2)} = \frac{m_{\rm AFM}}{4\pi t_{\rm PM}^3} \left(\frac{r_{\rm FM}^{(2)}}{r_{AFM}^{(2)}}\right).$$
(5)

and, replacing Eqs. (3-5) into Eq. (2), one obtains

$$E_{int}(\theta_1, \theta_2) = - K_{\rm FM}[\cos^2(\beta - \theta_1) + \cos^2(\beta - \theta_2)] - M_{\rm FM}\mu_0(H + H_{\rm EB}^{(1)})\cos(\theta_1) - M_{\rm FM}\mu_0(H - H_{\rm EB}^{(2)})\cos(\theta_2) .$$
(6)

This way, the energy cost associated with the reversal of these additional magnetic fields generates double hysteresis loops. To obtain the magnetization M we seek the solution of $\partial E_{int}(\theta_1, \theta_2)/\partial \theta_1 = 0 = \partial E_{int}(\theta_1, \theta_2)/\partial \theta_2$. Hence

$$M = M_{sat} \left[\frac{r_{\rm FM}^{(1)}}{r_{\rm AFM}^{(1)}} \cos(\theta_1) + \frac{r_{\rm FM}^{(2)}}{r_{\rm AFM}^{(2)}} \cos(\theta_2) \right].$$
(7)

Magnetic domain size

The size of the FM domains can be estimated by using the model by Gaunt, which relates the magnetic viscosity to the thermal activation energy of the reversal mode. For a single domain under an applied field H opposite to the magnetization M_{FM} , the activation energy is given by

$$E_{\rm Act} = K_{\rm FM} t_{\rm FM} r_{\rm FM}^2 (1 - \mu_0 {\rm HM}_{\rm FM} / 2K_{\rm FM})^2 , \qquad (8)$$

Consequently, the thermal energy k_BT activates a FM domain of size

$$r_{\rm FM}(H) = \sqrt{\frac{k_B T}{K_{\rm FM} t_{\rm FM}}} \frac{1}{(1 - \mu_0 H M_{\rm FM} / 2K_{\rm FM})} \,. \tag{9}$$

If we assume that, that due to the AFM dipole coupling, $r_{AFM} \sim r_{FM}$, then from Eq. (9) $r_{AFM}^{(1,2)} \sim r_{FM}(H=0) \sim 300$ Å. Since $M_{FM}^{(1)}$ and $M_{FM}^{(2)}$ initially are oriented in opposite directions

$$\frac{r_{\rm FM}^{(1)}}{r_{\rm AFM}^{(1)}} = \frac{\rho_1}{(1 - \mu_0 (H_{\rm FC} - H_{\rm dip}^{\rm AFM}) M_{\rm FM} / 2K_{\rm FM})} , \qquad (10)$$

$$\frac{r_{\rm FM}^{(2)}}{r_{\rm AFM}^{(2)}} = \frac{\rho_2}{(1 + \mu_0 (H_{\rm FC} + H_{\rm dip}^{\rm AFM}) M_{\rm FM} / 2K_{\rm FM})} , \qquad (11)$$

where $\rho_k = r_{\rm FM}(H=0)/r_{\rm AFM}^{(k)}$ is the ratio between FM domain-k and the respective AFM domain-k size. These definitions of ρ_k are valid in the absence of field cooling and the dipolar interaction.

AFM average magnetic moment

The FeF₂(110) AFM is modeled as a set of magnetically compensated planes parallel to the AFM/PM interface, labeled by $\ell \geq 0$ (with $\ell = 0$ specifying the interface layer) the AFM Hamiltonian is given by

$$\mathcal{H}_{AFM} = J_{AFM} \sum_{\ell,\ell'} \sum_{\mathbf{R},\mathbf{R}'} \mathbf{S}_{\alpha}(\ell,\mathbf{R}) \cdot \mathbf{S}_{\beta}(\ell',\mathbf{R}') -g\mu_{B}H_{FC} \sum_{\ell,\mathbf{R}} \left((H_{FC} + H_{dip}^{FM})S_{\alpha}^{z}(\ell,\mathbf{R}) + (H_{FC} - H_{dip}^{FM})S_{\beta}^{z}(\ell,\mathbf{R}) \right) -K_{AFM} \sum_{\ell,\mathbf{R}} \left((S_{\alpha}^{z}(\ell,\mathbf{R}))^{2} + (S_{\beta}^{z}(\ell,\mathbf{R}))^{2} \right), \qquad (12)$$

where J_{AFM} is the AFM exchange interaction, **R** and **R'** are the in-plane lattice vectors, and K_{AFM} is the magnitude of the uniaxial anisotropy along the in plane $\hat{\mathbf{z}}$ direction. The field cooling H_{FC} is applied along $\hat{\mathbf{z}}$, and the magnetic dipole field $H_{\text{dip}} = \mu_0 m_{\text{FM}} / (4\pi t_{\text{PM}}^3)$ (m_{FM} is the FM average magnetic moment) created by the FM is applied along $-\hat{\mathbf{z}}$, and the double summation in the first term corresponds to the AFM intra and inter plane exchange interactions. The Holstein-Primakoff transformations

$$S^{z}_{\alpha}(\ell, \mathbf{R}) = 1/2 - a^{\dagger}(\ell, \mathbf{R})a(\ell, \mathbf{R}),$$

$$S^{z}_{\beta}(\ell, \mathbf{R}) = -1/2 + b^{\dagger}(\ell, \mathbf{R})b(\ell, \mathbf{R}),$$

$$S^{+}_{\alpha}(\ell, \mathbf{R}) = \sqrt{2s}a(\ell, \mathbf{R}),$$

$$S^{+}_{\beta}(\ell, \mathbf{R}) = \sqrt{2s}b^{\dagger}(\ell, \mathbf{R}),$$

(13)

allow to recast the Hamiltonian of Eq. (12) in terms of elementary bosonic excitations created (destroyed) by the operators a^{\dagger}, b^{\dagger} (a, b). We ignore magnon-magnon interactions and consequently quadratic and higher order terms are neglected. Since there is in-plane translational symmetry the boson operators can be expanded in parallel to the interface spin waves, which allows to decouple the Hamiltonian into a set of independent semi-infinite linear chains,

$$\mathcal{H}_{AFM} = J_{AFM} \sum_{\ell,\ell'} \sum_{\mathbf{k}} \left[\gamma_{\mathbf{k}} \left(a(\ell, \mathbf{k}) b(\ell', -\mathbf{k}) + a^{\dagger}(\ell, \mathbf{k}) b^{\dagger}(\ell', -\mathbf{k}) + b^{\dagger}(\ell, \mathbf{k}) a^{\dagger}(\ell', -\mathbf{k}) + b(\ell, \mathbf{k}) a(\ell', -\mathbf{k}) \right) \right. \\ \left. + a^{\dagger}(\ell, \mathbf{k}) a(\ell, \mathbf{k}) + b^{\dagger}(\ell', \mathbf{k}) b(\ell', \mathbf{k}) + b^{\dagger}(\ell, \mathbf{k}) b(\ell, \mathbf{k}) + a^{\dagger}(\ell', \mathbf{k}) a(\ell', \mathbf{k}) \right] \\ \left. \sum_{\ell, \mathbf{k}} \left[\left(K_{AFM} + g\mu_B (H_{FC} + H_{dip}^{FM}) \right) a^{\dagger}(\ell, \mathbf{k}) a(\ell, \mathbf{k}) + \left(K_{AFM} - g\mu_B (H_{FC} - H_{dip}^{FM}) \right) b^{\dagger}(\ell, \mathbf{k}) b(\ell, \mathbf{k}) \right],$$

$$(14)$$

with $\gamma_{\mathbf{k}} = \sum_{\boldsymbol{\eta}} e^{i\mathbf{k}\cdot\boldsymbol{\eta}}$, $\boldsymbol{\eta}$ are the lattice vectors. Quantum fluctuations dynamics and symmetry breaking, can be extracted from the Green functions, which are defined by

$$G^{aa}_{\ell,\ell'}(\omega,\mathbf{k}) = -\frac{i}{\hbar} \int dt e^{i\omega t} \left\langle \left[a(\ell,\mathbf{k},t), a^{\dagger}(\ell',\mathbf{k},0) \right] \right\rangle,$$
(15)

$$G^{bb}_{\ell,\ell'}(\omega,\mathbf{k}) = -\frac{i}{\hbar} \int dt e^{i\omega t} \left\langle \left[b(\ell,\mathbf{k},t), b^{\dagger}(\ell',\mathbf{k},0) \right] \right\rangle, \tag{16}$$

In terms of the dimensionless variables $z = \omega/J_{AFM}$, $h = g\mu_B H_{FC}/J_{AFM}$, $t = g\mu_B H_{dip}/J_{AFM}$, $\kappa = K_{AFM}/J_{AFM}$, $g_{\ell,\ell'}^{(aa)} = J_{AFM}G_{\ell,\ell'}^{aa}$, and $g_{\ell,\ell'}^{(bb)} = J_{AFM}G_{\ell,\ell'}^{bb}$, the average magnetic moment can be written as

$$m_{\rm AFM} = \mu_B \sum_{\mathbf{r}} \left\langle S_{\alpha}^z(\mathbf{r}) - S_{\beta}^z(\mathbf{r}) \right\rangle \approx \frac{2\mu_B}{\pi} \Im \sum_{\mathbf{k}} \int dz \left(\frac{g_{0,0}^{aa}(z,\gamma_{\mathbf{k}},h) - g_{0,0}^{bb}(z,\gamma_{\mathbf{k}},h)}{\exp[J_{\rm AFM}z/k_BT] - 1} \right) \tag{17}$$

Due to sub-lattice symmetry of compensated AFM (110) surface, one obtain $g_{0,0}^{aa}(z,\gamma_{\mathbf{k}},h) = g_{0,0}^{bb}(z,\gamma_{\mathbf{k}},-h)$, thus

$$m_{\rm AFM} = -\frac{4\mu_B h}{\pi} \Im \int dz \, \frac{\partial}{\partial h} \left(\frac{\sum_{\mathbf{k}} g_{0,0}^{bb}(z,\gamma_{\mathbf{k}},h)}{1 - \exp[J_{\rm AFM} \, z/k_B T]} \right) \Big|_{h \to 0}.$$
(18)

Using the transfer matrix method, and the solution of the coupled Green function equations we obtain

$$\sum_{\mathbf{k}} g_{0,0}^{bb}(z,\gamma_{\mathbf{k}},h) \approx \frac{1}{z+t+h-\kappa-2},\tag{19}$$

this way

$$m_{\rm AFM} = \left(\frac{4\mu_B h}{\pi}\right) \Im \int dz \frac{1}{1 - \exp[J_{\rm AFM} z/k_B T]} \frac{1}{(z + t - \kappa - 2)^2},$$

= $4\mu_B \left(\frac{2\mu_B H_{\rm FC}}{k_B T}\right) \frac{\exp[(K_{\rm AFM} + 2J_{\rm AFM} - g\mu_B H_{\rm dip})/k_B T]}{(1 - \exp[(K_{\rm AFM} + 2J_{\rm AFM} - g\mu_B H_{\rm dip})/k_B T])^2}.$ (20)