Magnetic dichroism in darkfield UV photoemission electron microscopy

— Supplemental Material —

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ELECTRONIC-STRUCTURE AND PHOTOEMISSION CALCULATIONS

For the calculations of the electronic structure and the photoemission intensities we used our computer program package OMNI, which is based on the spin-polarized relativistic layer Korringa-Kohn-Rostoker method (KKR). Solving the Dirac equation for a magnetic material, exchange splitting and spin-orbit coupling are treated on equal footing. OMNI has proven its quality in a number of publications, see for example Ref. 1 and references therein.

The sample is taken as semi-infinite Fe(100), for which the muffin-tin potentials of the first > Insert number < layers deviate from that of bulk layers. > Relaxation? < For the image-potential barrier we take the > Add form < form > Add reference < .

Self-consistent electronic-structure calculations have been performed using the local spin-density approximation for the exchange-correlation functional in density-functional theory. For solving the single-site scattering problem, the layer-KKR method uses an expansion of the scattering solutions with respect to angular momentum, in this work up to $l_{\max} =?$> Add number < . The interlayer scattering relies on a plane-wave expansion, here with an energy cut-off of $\hbar^2 \boldsymbol{k}_{\parallel}^2/2m \leq ?$ H.> Add number < . The calculated bulk and the surface electronic structures as well as the layer-resolved magnetic moments agree with those computed and published elsewhere.

The results of the electronic-structure calculations serve as basis for the spin- and angle-resolved photoemission calculations, for which the same potentials as for the electronic-structure calculations are used. The spin-polarized photocurrent is calculated within the one-step model of photoemission, taking a time-reversed LEED state as final state. Photoelectrons excited within the topmost 50 layers are considered in order to obtain converged intensities and spin polarizations. The calculated spin-density matrix of the photoelectron allows to derive all components of the photoelectron's spin polarization vector.

In order to mimic many-particle effects, we use an energy-dependent self-energy $\Sigma(E)$ whose real part shifts electronic states in energy and its imaginary part broadens the photoemission spectra. This energy dependence is modelled by smooth functions while the true self-energy is more complicated and, on top of this, depends on the wave vector $\boldsymbol{k}_{\parallel}$ (see for example Ref. 1). This means that one can expect deviations between experimental and theoretical data taken at constant binding energy (confer Figs. 2 and 3 of the main text).

TABLE I. Effect of symmetry operations on the photoelectron wavevector $\boldsymbol{k}_\parallel = (k_x, k_y)$, the helicity of the incident light $\sigma_\pm$, and the in-plane magnetization $\boldsymbol{M} = (M_x, M_y)$. For details see text.

operation	wavevector		helicity	magnetization	
$\hat{1}$	k_x	k_y	$\sigma_\pm$	M_x	M_y
$\hat{m}_{xz}$	k_x	$-k_y$	$\sigma_\mp$	$-M_x$	M_y

SYMMETRY ANALYSIS

The point group of a nonmagnetic bcc(100) surface is $C_{4\mathrm{v}}$ $(4mm)$, which in the case of photoemission from ferromagnetic Fe is reduced to C_s (m), if the light impinges within a mirror plane of the lattice (here: the xz-plane; confer Fig. 1 of the main text). The two symmetry operations are thus the identity $\hat{1}$ and the reflection $\hat{m}_{xz}$ at the xz-plane. The effect of these operations on the photoelectron's wavevector $\boldsymbol{k}_\parallel = (k_x, k_y)$, on the helicity $\sigma_\pm$ of the circular polarized light, and on the in-plane magnetization $\boldsymbol{M} = (M_x, M_y)$ is given in Table I.

Table I allows to derive relations between photoemission intensities $I(k_x, k_y; \sigma_\pm; M_x, M_y)$ for equivalent setups. For example, photoemission from a ferromagnetic domain with magnetization along the x-axis ($M_y = 0$) yields

$$I(k_x, k_y; \sigma_+; M_x, 0) = I(k_x, -k_y; \sigma_-; -M_x, 0). \tag{1}$$

If $k_y = 0$ (detection in the xz-plane) one obtains the established relation that the intensity is not changed if both magnetization and helicity are reversed. Likewise, the relation

$$I(k_x, k_y; \sigma_+; 0, M_y) = I(k_x, -k_y; \sigma_-; 0, M_y) \tag{2}$$

tells that for a domain with magnetization along the y-axis reversing both the azimuth ($k_y \to -k_y$) and the helicity yield the same intensity. Moreover, generalizing these examples allows to derive the symmetry of the patterns shown in Figs. 2 and 3 of the main text.

The above considerations yield relations between intensities for photoelectrons emitted with opposite azimuths, that is with $\boldsymbol{k}_\parallel = (k_x, k_y)$ and $\boldsymbol{k}_\parallel = (k_x, -k_y)$. Since photoelectrons from both azimuths are detected in a PEEM, the exchange asymmetry can in general be increased if only photoelectrons of one chosen azimuth $\boldsymbol{k}_\parallel$ are considered; the others are ignored by placing an aperture within the PEEM's beam path.

For a fixed $\boldsymbol{k}_\parallel$ we define asymmetries that disentangle the contrast mechanisms: light polarization (A_{pol}) and magnetization orientation ((A_{ex})). With the four intensities $I(\boldsymbol{k}_\parallel, \sigma_\pm, \pm M)$, in the following shortened as $I_{\pm\pm}$, we define the total intensity

$$I \equiv I_{++} + I_{+-} + I_{-+} + I_{--} \tag{3}$$

and the respective asymmetries

$$A_{\mathrm{pol}} \equiv \left[(I_{++} + I_{+-}) - (I_{-+} + I_{--})\right]/I, \tag{4a}$$

$$A_{\mathrm{ex}} \equiv \left[(I_{++} + I_{--}) - (I_{+-} + I_{-+})\right]/I. \tag{4b}$$

A third asymmetry,

$$A_{\mathrm{soc}} \equiv \left[(I_{++} + I_{-+}) - (I_{+-} + I_{--})\right]/I, \tag{5}$$

quantifies the spin polarization produced by optical orientation, aka Fano effect, which requires spin-orbit coupling (SOC).

[1] C. Tusche, M. Ellguth, V. Feyer, A. Krasyuk, C. Wiemann, J. Henk, C. M. Schneider, and J. Kirschner, Nonlocal electron correlations in an itinerant ferromagnet, Nature Communications **9**, 3727 (2018).