

# Magnetic dichroism in darkfield UV photoemission electron microscopy

Maximilian Paleschke,<sup>1</sup> David Huber,<sup>1</sup> Friederike Wühl,<sup>1</sup> Cheng-Tien Chiang,<sup>2</sup> Frank O. Schumann,<sup>3</sup> Jürgen Henk,<sup>1</sup> and Wolf Widdra<sup>1</sup>

<sup>1</sup>*Institute of Physics, Martin Luther University Halle-Wittenberg, D-06099 Halle (Saale), Germany*

<sup>2</sup>*Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei, Taiwan*

<sup>3</sup>*Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany*

(Dated: August 9, 2024)

Photoemission electron microscopy (PEEM) has evolved into an indispensable tool for structural and magnetic characterization of surfaces at the nanometer scale. Particularly, synchrotron-radiation-based X-ray PEEM has emerged as a leading method for probing element-specific magnetic properties via magnetic circular dichroism (MCD) in core-level photoemission. In laboratory settings, ultraviolet (UV) radiation is utilized for near-threshold PEEM, which, when combined with femtosecond lasers, offers the potential for ultrafast time resolution. However, the characterization of magnetic properties, such as local magnetic domain structures, has seen limited application in UV PEEM, with studies reporting poor magnetic dichroism effects for in-plane magnetization. Here we introduce the concept of darkfield PEEM for MCD in threshold photoemission. This method enables efficient MCD imaging with a significantly enhanced MCD contrast —by an order of magnitude—for in-plane magnetization, as demonstrated for Fe(001). This advancement paves the way for MCD imaging on femtosecond timescales using modern lasers. Darkfield PEEM imaging employs an aperture for photoelectron momentum selection in the back focal plane of the electron imaging column before forming the real-space image. While the general momentum dependence of the MCD contrast will be explained through symmetry considerations, the experimental results for Fe(001) will be quantitatively compared with state-of-the-art relativistic photoemission calculations.

**Introduction.** Ultrafast spin and magnetization dynamics are exciting and rapidly growing fields in condensed matter physics with promising implications for both future research and device applications. Ultrafast imaging of magnetic domains on the micrometer scale is well established utilizing all-optical methods, as e.g. Kerr microscopy. On the nanometer scale, however, electron microscopy is the method of choice due to the electrons' short de Broglie wavelength.

Magnetic circular dichroism (MCD) provides the contrast mechanism used for imaging magnetic domains in photoelectron emission microscopy (PEEM). The intensity recorded for a particular domain changes with the helicity of the incident radiation, thereby producing magnetic contrast without need for explicitly detecting the electron spin. By tuning the incident X-ray radiation to a magnetic core level absorption edge, substantial and element-specific MCD asymmetries have been reported. With the wide availability of tunable synchrotron radiation, this technique of XMCD-PEEM is well established for magnetic domain imaging on the nanometer scale [1]. However, the pulse length of synchrotron radiation of typically 30 – 50 ps renders XMCD-PEEM unsuitable on ultrafast timescales. Replacing the incident X-ray radiation by ultrashort laser pulses would solve this issue straightforwardly and allow for pump-probe experiments on the timescale of a few femtoseconds. In addition, experiments can be performed in the laboratory with UV laser sources, which excite electrons close to the Fermi level to energies slightly above the escape threshold. However, reported MCD contrasts are quite small in threshold photoemission [2], especially for in-plane mag-

netization. Obviously, magnetic contrast needs to be increased for domain imaging with ultrashort laser pulses.

As we demonstrate here, the concept of darkfield PEEM in threshold photoemission allows efficient MCD imaging with an order-of-magnitude enhanced MCD contrast for in-plane magnetization. It paves the way for MCD imaging on femtosecond timescales with modern UV laser sources. Darkfield PEEM imaging uses an aperture for photoelectron momentum selection in the back focal plane of the electron imaging column prior to forming the real space image. We will demonstrate this for the in-plane magnetic structure at the Fe(001) surface and compare quantitatively the experimental results with those of fully relativistic photoemission calculations.

Following initial reports of magnetic dichroism in UV photoemission and its theoretical description in the 1990s [3–5], Marx *et al.* reported the first observation of magnetic dichroism in threshold PEEM in 2000 [2]. This study on polycrystalline Fe revealed an asymmetry in magnetic linear dichroism of 0.37 %. Subsequent spectroscopic studies confirmed the presence of both circular and linear dichroism in various ferromagnetic materials. Building on Marx's experimental work, Nakagawa *et al.* examined Ni films adsorbed with Cs, discovering significant asymmetries of up to 12 % in circular dichroism PEEM for out-of-plane magnetized domains [6–8]. This work also demonstrated the feasibility of using pulsed laser light for dichroism imaging. However, due to the limited photon-energy range of common optical laser setups, Cs remains necessary in most photoemission experiments in order to reduce the work function [9, 10]. For instance, investigations of domains on FePt surfaces em-

played a pulsed deep-UV laser with a photon energy of 7 eV [11].

The theoretical framework for valence-band dichroism was primarily developed in the 1990s and early 2000s (cf. Refs. 4, 5, 12–16) and was bolstered by pioneering experiments (cf. Refs. 17–21). It is based on calculating the relativistic electronic structures in conjunction with a theoretical description of the photoemission process

Do we need to cite Getzlaff'22 [22]?

*Conceptual basis.* For a simplified conceptual approach we consider a surface with fourfold symmetry, as e.g. the (001) fcc or bcc surfaces with magnetic easy axes along one of the four [100] directions. Let us assume light incidence along the surface normal (Fig. 1 for  $\theta = 0$ ). The photoemission intensity of electrons detected with off-normal wavevector  $\mathbf{k}$  depends then on the helicity,  $\sigma_+$  or  $\sigma_-$ , of the incident circular polarized laser radiation and on the two orientations  $\pm M$  of the in-plane magnetization in a selected domain, yielding four intensities  $I_{\mathbf{k}}(\sigma_{\pm}, \pm M)$  (shortened  $I_{\pm\pm}$ ). The latter intensities are combined into the total intensity

$$I \equiv I_{++} + I_{+-} + I_{-+} + I_{--}. \quad (1)$$

In order to disentangle the two main contrast mechanisms we define appropriate asymmetries [23],

$$A_{\text{pol}} \equiv [(I_{++} + I_{+-}) - (I_{-+} + I_{--})] / I, \quad (2a)$$

$$A_{\text{ex}} \equiv [(I_{++} + I_{--}) - (I_{+-} + I_{-+})] / I. \quad (2b)$$

In the polarization asymmetry  $A_{\text{pol}}$  the magnetization's orientation is averaged out; it thus encodes contrast due to the light's helicity, as if the domain were nonmagnetic. Contrast due to the exchange splitting is quantified by the exchange asymmetry  $A_{\text{ex}}$ , in which one averages over the mutual orientations of helicity and magnetization. Note that the *chiral geometry* for photoelectrons with off-normal wavevector  $\mathbf{k}$  outside the scattering plane results in magnetic dichroism and, hence, in magnetic contrast.

If the scattering plane is a mirror plane of the lattice, the photoemission intensities for fixed  $\mathbf{k}$  within the scattering plane obey  $I_{++} = I_{--}$  for a magnetization within the scattering plane (Fig. 1(a)). This results in a nonzero  $A_{\text{ex}}$ , but vanishing  $A_{\text{pol}}$ . For perpendicular magnetization,  $I_{++} = I_{-+}$  holds, leading to vanishing  $A_{\text{pol}}$  and vanishing  $A_{\text{ex}}$ .

In the following, we compare theoretical MCD asymmetries based on relativistic photoemission computations for Fe(001), briefly described in the Supplemental Material [24], with corresponding experimental results for a photon energy of 5.2 eV. The photocurrent has been recorded for 60° grazing light incidence within the [100] high-symmetry direction in a standard PEEM setup (Focus GmbH, Hünstetten). As light source either a mercury discharge lamp or the frequency-doubled output of a non-collinear optical amplifier (NOPA) with circular polarization optics is used [25–27].

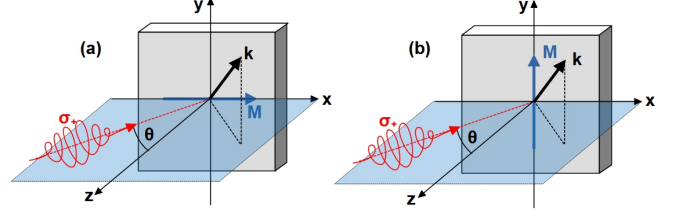


FIG. 1. Symmetry analysis. A circular polarized laser pulse (orange, with helicity  $\sigma_+$  impinges onto a magnetic domain (rectangular solid)). The light incidence direction and the surface normal ( $z$ -axis) span the scattering plane (blue;  $xz$ -plane) with the magnetization direction  $\mathbf{M}$  oriented within (a) or perpendicular (b) to the scattering plane, respectively. The off-normal detection of photoelectrons with wavevector  $\mathbf{k}$  (black arrow) results in a chiral setup.

*Contrast mechanisms.* The  $k_{\parallel}$ -dependent pattern of the polarization asymmetry  $A_{\text{pol}}$ , defined in Eq. (2a) and depicted in Fig. 2, depends on the binding energy of the initial states. Both experimental (left column) and theoretical data (right column) show that this contrast mechanism is sizable with absolute values up to about 20 % in experiment and 40 % in theory; it can thus hardly be ignored.

The theoretical pattern in momentum space (right column in Fig. 2) exhibits a nodal line at  $k_y = 0$  and almost a nodal line at  $k_x = 0$ . Moreover, one finds a change of sign if  $k_y$  is reversed. These features are imposed by the symmetry of the setup. Note that an antisymmetric pattern with respect to the  $k_x = 0$  and  $k_y = 0$  lines follows strictly only for normal light incidence [28]. However, the breaking of the antisymmetric behavior with respect to the  $k_x = 0$  line due to the off-normal light incidence is only hardly visible. The experimental data (left column) display the same features and the overall agreement between experiment and theory is remarkably good, which includes also the sign change for binding energies above and below 0.2 eV. Note that the experimental asymmetries have been determined from two individual 2D momentum maps for magnetization directions  $+\mathbf{M}$  and  $-\mathbf{M}$  oriented along the  $+x$  and  $-x$  directions, respectively, via selection of appropriate individual magnetic domains.

The momentum-dependent exchange asymmetry  $A_{\text{ex}}$ , defined in Eq. (2b) and shown in Fig. 3, exhibits absolute values up to 10 % in theory and 6 % in experiment. An odd symmetry of the momentum-dependent  $A_{\text{ex}}$  pattern with respect to the  $k_x = 0$  line would be expected for normal light incidence [28]. However, for the grazing light incidence here, we find a clear deviation, which results in a curved nodal line between the regions of positive and negative  $A_{\text{ex}}$ . With respect to the  $k_y = 0$  line, experiment as well as theory show a mirror symmetric pattern in contrast to  $A_{\text{pol}}$ . The absolute  $A_{\text{ex}}$  values, including the sign, depend on the initial state binding energy via

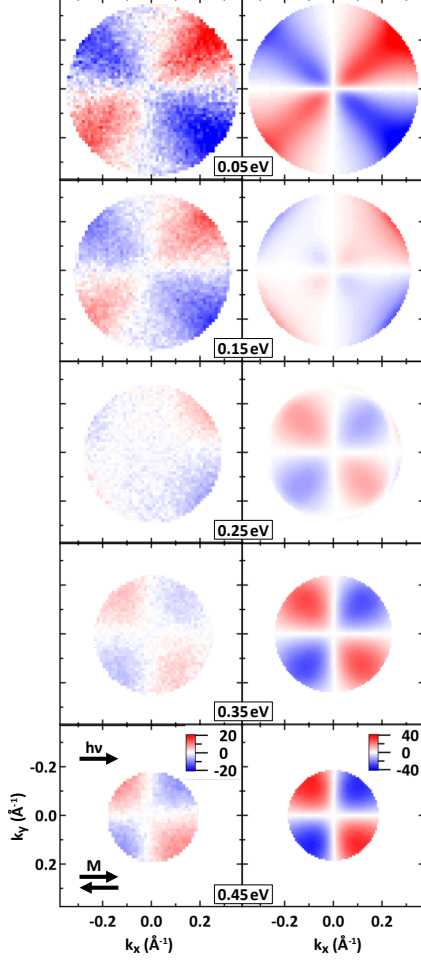


FIG. 2. Momentum-resolved polarization asymmetry  $A_{\text{pol}}$  of Fe(001) at selected binding energies for 60° grazing light incidence. Left column: experimental results. The arrow marked  $h\nu$  indicates the light incidence direction. The arrows  $M$  represent the two magnetization directions considered for  $A_{\text{pol}}$ . Right column: respective theoretical results obtained from photoemission calculations. The binding energy is indicated at each panel. The color scale, showing  $A_{\text{pol}}$  as defined in Eq. (2a) in percent, is identical for all panels in this column.

the band structure and photoemission matrix elements.

The above findings support that both asymmetries  $A_{\text{pol}}$  and  $A_{\text{ex}}$  are suitable tools for disentangling and quantifying the main contrast mechanisms for domain imaging.

*Domain imaging.* From the momentum-resolved  $A_{\text{ex}}$  pattern in Fig. 3, it follows that MCD imaging **might** reveal strong magnetic contrast in case of *off-normal* electron momentum selection. However, without momentum selection or with a momentum selection centered at  $k_x = k_y = 0$ , which has been **widely applied so far**, different momentum contributions **will** largely cancel each other. This cancellation explains the small or vanishing magnetic dichroism for in-plane magnetized domains re-

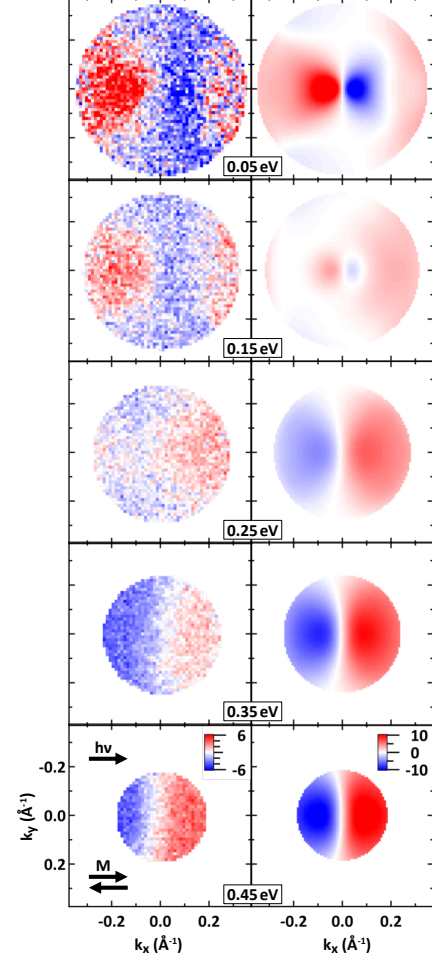


FIG. 3. Momentum-resolved exchange asymmetry  $A_{\text{ex}}$  of Fe(001) at selected binding energies, as in Fig. 2. Left column: experimental PEEM data, right column: respective data from photoemission calculations. Small differences with respect to an odd symmetry upon reversal of  $k_x$  result from grazing light incidence. For normal incidence they are absent.

ported so far [2].

The above suggests to **reduce** the  $k_{\parallel}$  area of interest in order to enhance the magnetic contrast. Hence, we place a circular contrast aperture in a  $k_{\parallel}$  area with high exchange asymmetry, a procedure known as dark-field imaging in optics.

Depending on the position of the aperture the contrast of specific domains is increased, as we show for a Landau-like pattern of four orthogonal magnetic domains at a Fe(001) surface. (Fig. 4). Placing the aperture in **nine** different positions (black circles in the top panel of Fig. 4) results in **nine** corresponding MCD PEEM images of the same surface region (bottom panel).

For the centred aperture, marked in red, the MCD contrast almost vanishes in accordance with **the** above **argument**. However, an aperture centered at  $k_x > 0$

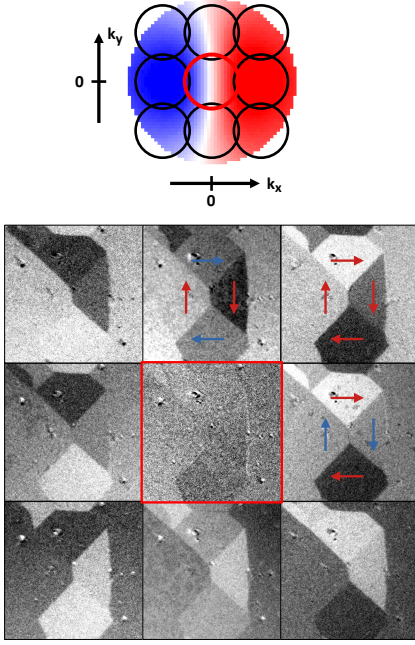


FIG. 4. Darkfield MCD imaging of Fe(100). Top: Schematics of the **nine** aperture positions in the momentum plane, with a momentum-resolved  $A_{\text{ex}}$  pattern as background. Bottom: Domain imaging using the nine aperture positions shown above. ( $\hbar\nu = 5.2$  eV, maximum domain contrast is between 3 and 4%, field of view  $56 \times 56 \mu\text{m}^2$ .)

and  $k_y = 0$  results in a drastically increased contrast of 3 – 4% for magnetic domains oriented in  $+x$  versus  $-x$  direction, whereas the contrast for domains oriented in  $+y$  and  $-y$  directions (blue arrows) vanishes. Both observations match quantitatively the result of the  $\mathbf{k}_{\parallel}$  space measurements in Fig. 3(b).

Positioning the momentum aperture at  $k_x < 0$  and  $k_y = 0$  reverses the contrast of  $+x$  and  $-x$  domains. As expected, the contrast switches from sensitivity in  $x$  direction to  $y$  direction when positioning the aperture at  $k_x = 0$  and  $k_y > 0$  (upper-middle PEEM image in Fig. 4). The upper-right measurement shows a diagonal position with  $k_x > 0$  and  $k_y > 0$ , where the different contributions to the MCD signal **mix**, resulting in four different asymmetry values for the four in-plane magnetization directions.

*Initial-state effects.* The magnitude of  $A_{\text{ex}}$  and, therefore, of the MCD contrast in PEEM for near-threshold photoemission depend on the initial-state energy, as is demonstrated in Fig. 3.  $A_{\text{ex}}$  reverses sign from about +6% slightly below the Fermi level to –6% at  $E_B = 0.45$  eV. As a second, magnetically similar system we studied the oxygen-passivated Fe(001)-(1×1)-O surface with darkfield threshold PEEM, as described above. It yields very similar  $A_{\text{pol}}$  and  $A_{\text{ex}}$  patterns as those for bare Fe(001) (not shown here), which reverse sign at a binding energy of approximately 0.2 eV. Momentum-

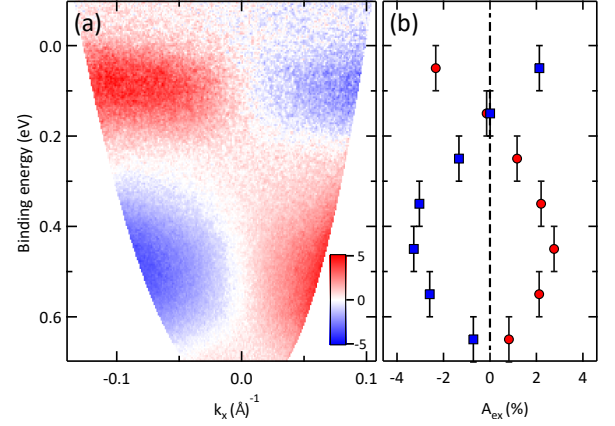


FIG. 5. Binding-energy dependent exchange asymmetry  $A_{\text{ex}}$  for Fe(001)-(1×1)-O at  $\hbar\nu = 5.2$  eV. (a) ARPES data for an oxygen-passivated Fe(001) thin film grown on MgO(001) with sample magnetized in  $+x$  and  $-x$  direction (light incidence at  $70^\circ$ ,  $k_y = 0$ ). (b) Momentum-selected PEEM data for an oxygen-passivated Fe(001) single crystal for positive and negative  $k_x$  momentum selection as marked by blue squares and red circles, respectively. (**selection at  $|k_x| = (0.16 \pm 0.12) \text{ \AA}^{-1}$ ,  $k_y = (0 \pm 0.12) \text{ \AA}^{-1}$ , light incidence at  $60^\circ$ .**)

selected  $A_{\text{ex}}$  data from momentum-resolved PEEM measurements on **two either in  $+x$  or  $-x$**  direction magnetized domains are shown in Fig. 5(b) for positive  $k_x$  as blue squares and for negative  $k_x$  as red circles.

Using an angle-resolved photoelectron spectroscopy (ARPES) setup described **elsewhere** [26, 29], the magnetic circular dichroism is analyzed in an independent experiment with higher energy resolution for **a** 11 nm thick Fe(001)-(1×1)-O thin film grown on MgO(001). For fully in  $+x$  or  $-x$  direction magnetized films, the exchange asymmetry  $A_{\text{ex}}$  is depicted in an energy vs momentum map in Fig. 5(a). Both datasets show large  $A_{\text{ex}}$  values, which switch sign upon reversal of  $k_x$ . At the Fermi level and at  $E_B = 0.45$  eV we find strong contrast of about 5% between  $A_{\text{ex}}$  values of +2 and –3% with a contrast reversal between 0.15 and 0.25 eV. We recall that this observation calls for exact threshold photoexcitation or for an energy-resolved electron detection in order to obtain high MCD signals. **Otherwise positive and negative exchange asymmetries will partly cancel each other.**

While the darkfield scheme of threshold MCD PEEM is broadly applicable, the magnitude of the binding-energy dependent exchange asymmetry  $A_{\text{ex}}$  is a material-specific property. It results from the spin-dependent electronic structure of Fe(001) and the associated ARPES transition matrix elements. Note that for a fixed binding energy these matrix elements depend on the photon energy due to energy conservation.

*Summary and prospects.* The present investigation proves that magnetic domains can be imaged with high



contrast using a threshold PEEM with momentum selection of the detected photoelectrons, thereby introducing the concept of darkfield threshold MCD PEEM. For a proof-of-principle, we applied darkfield UV PEEM to an in-plane magnetized Fe(001) surface. However, the approach is general so that it can easily be applied to other ferromagnets, even to those which include out-of-plane magnetization [9]. Thus, it is well suited for studying magnetic reorientation transitions, as for example observed for Ni/Cu(001) [9, 30–32]. We see its main capabilities, however, in investigations of ultrafast magnetization dynamics using femtosecond laser pulses in a optical pump and threshold UV photoemission probe scheme. For applications it paves the way for imaging the ultrafast motion of domain walls [33] or of large skyrmions on nanometer length scales [34–36].

*Acknowledgments.* This work is funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project-ID 328545488 – TRR 227, projects A06 and B04.

- 
- [1] W. Kuch, R. Schäfer, P. Fischer, and F. Hillebrecht, *Magnetic Microscopy of Layered Structures*, Springer Series in Surface Sciences, Vol. 57 (Springer, Berlin Heidelberg, 2015).
  - [2] G. K. L. Marx, H. J. Elmers, and G. Schönhense, Magneto-optical Linear Dichroism in Threshold Photoemission Electron Microscopy of Polycrystalline Fe Films, *Physical Review Letters* **84**, 5888 (2000).
  - [3] C. M. Schneider, M. S. Hammond, P. Schuster, A. Cebollada, R. Miranda, and J. Kirschner, Observation of magnetic circular dichroism in **uv** photoemission from ferromagnetic fcc cobalt films, *Physical Review B* **44**, 12066 (1991).
  - [4] J. Henk, T. Scheunemann, S. V. Halilov, and R. Feder, Magnetic dichroism and electron spin polarization in photoemission: Analytical results, *Journal of Physics: Condensed Matter* **8**, 47 (1996).
  - [5] R. Feder and J. Henk, Magnetic dichroism and spin polarization in valence band photoemission, in *Spin-Orbit-Influenced Spectroscopies of Magnetic Solids*, Vol. 466, edited by H. Araki, E. Brézin, J. Ehlers, U. Frisch, K. Hepp, R. L. Jaffe, R. Kippenhahn, H. A. Weidenmüller, J. Wess, J. Zittartz, W. Beiglböck, H. Ebert, and G. Schütz (Springer Berlin Heidelberg, Berlin, Heidelberg, 1996) pp. 85–104.
  - [6] T. Nakagawa, T. Yokoyama, M. Hosaka, and M. Kato, Measurements of threshold photoemission magnetic dichroism using ultraviolet lasers and a photoelastic modulator, *Review of Scientific Instruments* **78**, 023907 (2007).
  - [7] T. Nakagawa, K. Watanabe, Y. Matsumoto, and T. Yokoyama, Magnetic circular dichroism photoemission electron microscopy using laser and threshold photoemission, *Journal of Physics: Condensed Matter* **21**, 314010 (2009).
  - [8] T. Nakagawa and T. Yokoyama, Laser induced threshold photoemission magnetic circular dichroism and its application to photoelectron microscope, *Journal of Electron Spectroscopy and Related Phenomena* **185**, 356 (2012).
  - [9] M. Kronseder, J. Minár, J. Braun, S. Günther, G. Woltersdorf, H. Ebert, and C. H. Back, Threshold photoemission magnetic circular dichroism of perpendicularly magnetized Ni films on Cu(001): Theory and experiment, *Phys. Rev. B* **83**, 132404 (2011).
  - [10] T. N. G. Meier, M. Kronseder, and C. H. Back, Domain-width model for perpendicularly magnetized systems with Dzyaloshinskii-Moriya interaction, *Phys. Rev. B* **96**, 144408 (2017).
  - [11] Y. Zhao, H. Lyu, G. Yang, B. Dong, J. Qi, J. Zhang, Z. Zhu, Y. Sun, G. Yu, Y. Jiang, H. Wei, J. Wang, J. Lu, Z. Wang, J. Cai, B. Shen, W. Zhan, F. Yang, S. Zhang, and S. Wang, Direct observation of magnetic contrast obtained by photoemission electron microscopy with deep ultra-violet laser excitation, *Ultramicroscopy* **202**, 156 (2019).
  - [12] E. Tamura, W. Piepke, and R. Feder, New spin-polarization effect in photoemission from nonmagnetic surfaces, *Phys. Rev. Lett.* **59**, 934 (1987).
  - [13] W. Kuch, A. Dittschar, K. Meinel, M. Zharnikov, C. M. Schneider, J. Kirschner, J. Henk, and R. Feder, Magnetic-circular-dichroism study of the valence states of perpendicularly magnetized Ni(001) films, *Phys. Rev. B* **53**, 11621 (1996).
  - [14] W. Kuch and C. M. Schneider, Magnetic dichroism in valence band photoemission, *Reports on Progress in Physics* **64**, 147 (2001).
  - [15] D. Venus, Interrelation of magnetic-dichroism effects seen in the angular distribution of photoelectrons from surfaces, *Phys. Rev. B* **49**, 8821 (1994).
  - [16] D. Venus, Interpretation of magnetic dichroism in angle-resolved UV photoemission from valence bands, *Journal of Magnetism and Magnetic Materials* **170**, 29 (1997).
  - [17] B. Schmiedeskamp, B. Vogt, and U. Heinzmann, Experimental verification of a new spin-polarization effect in photoemission: Polarized photoelectrons from Pt(111) with linearly polarized radiation in normal incidence and normal emission, *Phys. Rev. Lett.* **60**, 651 (1988).
  - [18] D. Venus, W. Kuch, A. Dittschar, M. Zharnikov, C. Schneider, and J. Kirschner, Spin-dependent surface transmission in 3d metals: Implications for magnetic-dichroism measurements of the valence bands, *Phys. Rev. B* **52**, 6174 (1995).
  - [19] K. Hild, J. Maul, T. Meng, M. Kallmayer, G. Schönhense, H. J. Elmers, R. Ramos, S. K. Arora, and I. V. Shvets, Optical magnetic circular dichroism in threshold photoemission from a magnetite thin film, *J. Phys.: Condens. Matter* **20**, 235218 (2008).
  - [20] K. Hild, J. Maul, G. Schönhense, H. J. Elmers, M. Amft, and P. M. Oppeneer, Magnetic Circular Dichroism in Two-Photon Photoemission, *Physical Review Letters* **102**, 057207 (2009).
  - [21] K. Hild, G. Schönhense, H. J. Elmers, T. Nakagawa, T. Yokoyama, K. Tarafder, and P. M. Oppeneer, Energy- and angle-dependent threshold photoemission magnetic circular dichroism from an ultrathin Co/Pt(111) film, *Phys. Rev. B* **82**, 195430 (2010).
  - [22] M. Getzlaff, Mcdad investigation to characterize the magnetic behavior of thin **pd** films on **co**(0001), *Journal of Electron Spectroscopy and Related Phenomena* **261**, 147266 (2022).

- [23] J. Henk and B. Johansson, Magnetic dichroism in off-normal valence-band photoemission, *Journal of Electron Spectroscopy and Related Phenomena* **94**, 259 (1998).
- [24] See Supplemental Material at [URL will be inserted by publisher] for supporting information and additional results.
- [25] K. Duncker, M. Kiel, and W. Widdra, Momentum-resolved lifetimes of image-potential states on Ag(001), *Surface Science* **606**, L87 (2012).
- [26] K. Gillmeister, D. Golež, C.-T. Chiang, N. Bittner, Y. Pavlyukh, J. Berakdar, P. Werner, and W. Widdra, Ultrafast coupled charge and spin dynamics in strongly correlated NiO, *Nat Commun* **11**, 4095 (2020).
- [27] M. Paleschke, C.-T. Chiang, L. Brandt, N. Liebing, G. Woltersdorf, and W. Widdra, Plasmonic spin-Hall effect of propagating surface plasmon polaritons in **Ni<sub>80</sub>Fe<sub>20</sub>** microstructures, *New J. Phys.* **23**, 093006 (2021).
- [28] (2024), f. Schumann et al., in preparation.
- [29] K. Gillmeister, M. Kiel, and W. Widdra, Image potential states at transition metal oxide surfaces: A time-resolved two-photon photoemission study on ultrathin NiO films, *Phys. Rev. B* **97**, 085424 (2018).
- [30] J. Henk, A. M. N. Niklasson, and B. Johansson, Magnetism and anisotropy of ultrathin Ni films on Cu(001), *Phys. Rev. B* **59**, 9332 (1999).
- [31] D. Sander, W. Pan, S. Ouazi, J. Kirschner, W. Meyer, M. Krause, S. Muller, L. Hammer, and K. Heinz, Reversible h-induced switching of the magnetic easy axis in **ni/cu**(001) thin films, *Physical Review Letters* **93**, 247203 (2004).
- [32] T. Nakagawa and T. Yokoyama, Magnetic Circular Dichroism near the Fermi Level, *Physical Review Letters* **96**, 237402 (2006).
- [33] S. S. P. Parkin, M. Hayashi, and L. Thomas, Magnetic domain-wall racetrack memory, *Science* **320**, 190 (2008).
- [34] B. Göbel, A. F. Schäffer, J. Berakdar, I. Mertig, and S. S. P. Parkin, Electrical writing, deleting, reading, and moving of magnetic skyrmioniums in a racetrack device, *Scientific Reports* **9**, 12119 (2019).
- [35] H. Jani, J.-C. Lin, J. Chen, J. Harrison, F. Maccherozzi, J. Schad, S. Prakash, C.-B. Eom, A. Ariando, T. Venkatesan, and P. G. Radaelli, Antiferromagnetic half-skyrmions and bimerons at room temperature, *Nature* **590**, 74 (2021).
- [36] L.-M. Kern, B. Pfau, V. Deinhart, M. Schneider, C. Klose, K. Gerlinger, S. Wittrock, D. Engel, I. Will, C. M. Günther, R. Liefferink, J. H. Mentink, S. Wintz, M. Weigand, M.-J. Huang, R. Battistelli, D. Metternich, F. Büttner, K. Höflich, and S. Eisebitt, Deterministic generation and guided motion of magnetic skyrmions by focused **he+**-ion irradiation, *Nano Letters* **22**, 4028 (2022).

## TODO LIST

Do we need to cite Getzlaff'22 [22]? ..... 2