Spin-polarized standing waves at an electronically matched interface detected by Fermi-surface photoemission

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Highly spin-polarized reflection at an interface of a ferromagnetic thin film is made visible by photoelectron spectroscopy. The technique of *k*-space mapping of the exchange-split Fermi surface is employed to detect standing waves confined to the ferromagnetic layer. A drastic spin asymmetry of this effect is achieved for a specific matching of the Fermi-surface topologies of film and substrate, respectively. For Fe(110) films on a W substrate, intense standing waves are obtained exclusively for majority states, while minority states are virtually unaffected by the boundary.

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In the pursuit of designing magnetoelectronics, it is mandatory to enhance the conductivity for one spin orientation over the other, thereby encoding information. Spin-dependent transmission can be affected by scattering processes in the bulk such as structural imperfections.[1] Even without defects, spin-dependent scattering in the bulk depends on empty states for electron-hole pair formation that lead to a spin asymmetry in the mean free path.[2] In layered magnetic nanostructures, however, one must specifically acknowledge the presence of interfaces. It is thus desirable to account for the propagation of spin-polarized electron wave functions across a single interface. Magnetic tunneling barriers are a key class of devices that directly depend on the interfacial discontinuity.[2–4] For giant magnetoresistance devices based on multilayers, the relative benefits of the various scattering processes are a matter of debate.[5,6]

However, it has not been clearly demonstrated to date to which degree the interfacial reflection can be optimized for spin filtering. Spectroscopic data for the conduction states of the ferromagnetic side are lacking. Previous work is focused on the *nonmagnetic side* of the interface. By spin-polarized photoemission, extension of electron states from bulk Co into a Cu film has been reported,[7] yet with rather limited spin polarization. For thin layers, the formation of standing waves (SWs) or "quantum well states" (in the low thickness limit) can be observed as indication of the interface reflectivity. Demonstrations of their feasibility are limited to nonmagnetic films, as observed in Cu,[8] Ag,[9,10] and Al.[11] In Ag quantum well films on Fe(110) (Ref. 9) using spin-polarized photoemission at normal emission, the Ag spectra for spin down and spin up coincide up to 1 eV below the Fermi level E_F, thus falling short of experimentally demonstrating a spin asymmetry near E_F. Concerning the ferromagnetic side of the interface, in a study of Co films on Cu(100),[12] quantum well states of *both* spin signs in Co were reported. This points at the difficulty to achieve the desired spin asymmetry, resulting in a reflectivity for both spin signs.

In this Brief Report, we describe a spectroscopic technique to observe the reflective properties of a ferromagnetic-paramagnetic interface. It is tuned for high spin asymmetry based on metals with selectively corresponding Fermi surfaces (FS). For these, a realization is found in Fe films on a W(110) substrate. The efficiency is probed by *k*-space mapping of the ferromagnetic FS with angle-resolved photoemission. SWs at the Fermi level restricted to one spin sign are thereby observed. In contrast, the minority Fermi level states exhibit a bulklike dispersion largely unaffected by this interface.

As theoretical prerequisite, the transmission properties depend on how the respective wave functions are continued across the interface. For transport, the wave functions at E_F are relevant. A selective match of the FS sheets of one spin sign in the ferromagnet with the FS of the paramagnet hints at the desired spin polarization. Among elemental ferromagnets and paramagnets within the same crystal lattice, we have considered various combinations and conclude that bcc Fe/W with a (110) interface provides the desired properties. The FS of bcc Fe as well as W have been derived with a modern density-functional theory (DFT) algorithm employing the computer code WIEN2K,[13] which includes the generalized gradient approximation. This delivers more accurate results for 3*d* metals than the conventional local density approximation, as discussed in Ref. 14.

For electron propagation perpendicular to the interface, the (110) FS projections are relevant, as shown in Fig. 1(a) for Fe and in Fig. 1(b) for W. Ferromagnetic Fe exhibits *minority* and *majority* sheets, the latter being clearly distinguishable by their comparatively large size and by forming extra sheets, as discussed in our earlier study of bulk iron.[14] The relevant fact is that the minority FS of Fe has a high resemblance to that of W, in stark contrast to the majority FS. One must therefore conclude that, at least close to E_F, only minority states find an extension in the W substrate, whereas most of the majority electrons are confined to the ferromagnetic layer, as illustrated in Fig. 1(c).

Experimentally, bcc Fe(110) films were grown by electron-beam evaporation onto a W(110) substrate and annealed at 500 °C. The thickness range determined with W 4*f* core level attenuation was typically $d \sim 10-20$ ML, as desired for SW formation discussed below. Angle-resolved

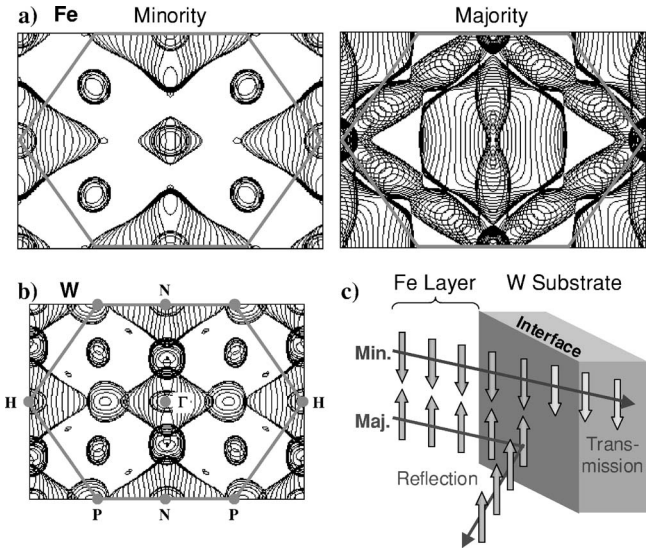

FIG. 1. Spin-selective matching of wave functions across the interface. (a) Calculated spin-resolved FS for Fe, projected on the (110) interface plane. (b) The FS projection of the paramagnetic W substrate is very similar to the minority states of Fe. (c) Schematic of spin-polarized transmission at E_F expected for the match of Fe minority states with those of W.

photoelectron spectroscopy (ARPES) was used to map FS cross sections. It was performed at the Advanced Light Source in Berkeley (beamline 7.0.1, ESF end station) at $T=25$ K with a total energy resolution of ~35 meV.

As an advantage of this technique, spin character can be assigned to the FS sheets after accurate comparison to bulk FS data available from theory and experiment. This has been done using the recent study with ARPES and DFT in Ref. 14. The FS data for an ~15 ML film in Fig. 2(a) show a section through the Brillouin zone (BZ) close to the Γ point ($h\nu=128$ eV). At $T=25$ K, the FS is fully exchange split into minority and majority FS sheets (small and large, respectively). The thin-film FS data have generally close correspondence to the bulk FS.[14] However, an additional observation is the splitting of the majority contour into two subcontours, corresponding to SW states.

The SW formation results from the quantized values for the momentum $k_\perp$ perpendicular to the surface. In a one-dimensional simplification, their number is given by the number N of unit cells (lattice constant a), resulting in a spacing of $2\pi/Na=2\pi/d$. Due to the uncertainty relation, they are also broadened by the same amount $\Delta k_\perp \sim 2\pi/d$. The picture in Fig. 2(b) assumes that the films are thick enough so that the bulk dispersion remains essentially intact, consistent with the present ARPES data and earlier findings.[10] An additional phase factor may occur due to the film boundaries.[9] Regarding the FS, the dispersion parallel to the interface is unaffected. The perpendicular quantization slices through this topology, as in Fig. 2(c) for the large majority FS sheet. The simultaneous observation of FS contours at given perpendicular momentum $k_\perp^0$ results from the limited electron escape depth $\lambda \sim 5$ ML. Thus, the Lorentzian sampling window $\Delta k_{\text{sampling}} \sim 2\pi/\lambda$ is larger than the $k_\perp$ raster. Accordingly, for $d \sim 15$ ML, one can detect approximately two to four FS contours simultaneously.

As for the remaining FS sheets, data closer to the Brillouin zone boundary in Fig. 3(a) show elliptical cross sections of the tubular majority FS sheets that surround the H points (labeled "A," guidelines based on symmetry arguments). A large rectangle around H is a minority sheet (labeled "B"), while a circular surface resonance is closely centered at H. Only majority sheets exhibit formation of SW states. A typical band map relating to a line cut through A in Fig. 3(b) shows multiples of the hole pocket bands. The rather parallel dispersion behavior is characteristic of SW states. To ensure that minority SW states have not been overlooked, we varied photon energy and film thickness, yet with negative result. A close-up data set in Fig. 3(c) for $d \sim 15$ ML shows the minority FS sheet B, recorded exactly at the Brillouin zone boundary ($h\nu=97$ eV).[14] Any SW states inside the hole pocket are absent within our detection limit.

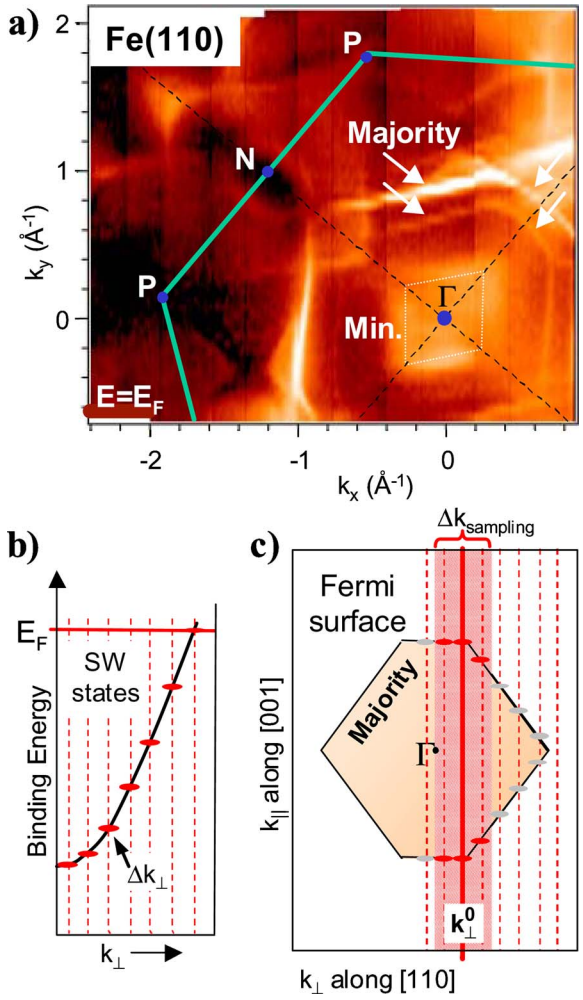

FIG. 2. (Color online) SW formation in the majority FS sheets of the Fe film. (a) ARPES data of ~15 ML film near the Γ point ($h\nu=128$ eV). It shows minority (small) and majority (large) sheets; the latter split due to SWs. (b) SWs are due to momentum quantization perpendicular to the interface, with energy levels derived from the bulk band structure. (c) Effect of SW formation on the FS of Fe, shown for the majority sheet at Γ. Multiple contours are seen simultaneously due to the sampling depth.

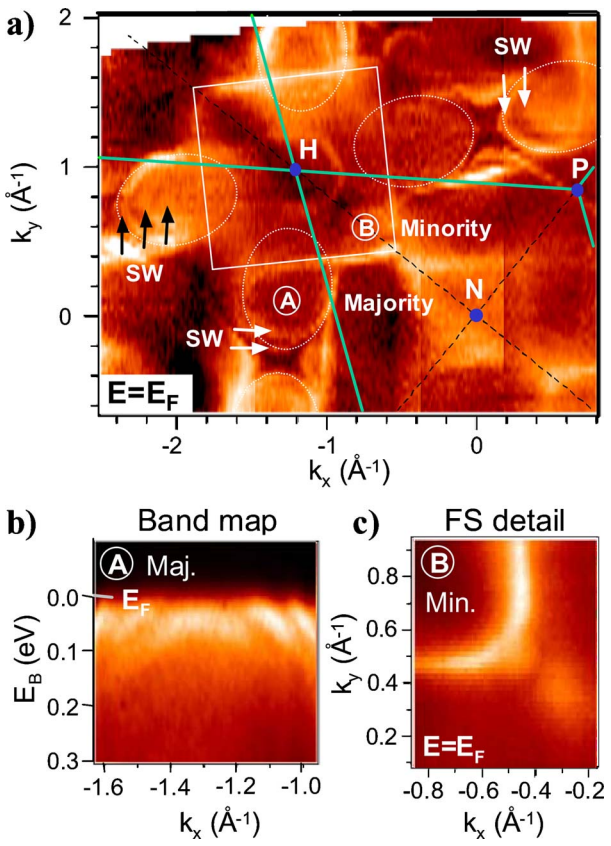

FIG. 3. (Color online) SWs near the boundary plane of the Fe Brillouin zone. (a) FS data of ~15 ML Fe film ($h\nu$=112 eV). The numerous tubular majority sheets are intersected as ellipses (A) and exhibit SW multiplets. In contrast, the square minority sheet (B) does not show SWs. (b) Band map ($h\nu$=97 eV) through majority sheet (A) showing multiple SW electron bands. (c) Closeup of minority sheet (B) measured at the zone boundary ($h\nu$=97 eV) with high statistics, confirming lack of SW states.

The film thickness chosen avoids the initial regime of Fe growth, as the lattice constant of Fe is smaller than W by nominally ~9%. The strain is accommodated in the first layers by periodic elongations of the Fe lattice positions.[15,16] Fortunately, in the interfacial registry, 37 Fe atoms coincide with 34 W atoms.[15] Consequently, the effective strain is reduced to ~1.2% laterally, while normal to the interface it is only 0.2%. In subsequent layers, the strain is relaxed until, at ~5 ML, the Fe lattice is assumed. The one-to-one correspondence of all minority FS contours with bulk data[14] assures us that parallel momentum is conserved. Intense majority SWs require that the confinement causes hardly any loss of amplitude, thus a reasonably smooth interface must be concluded. As additional test of the SW origin, thickness-dependent scans in the FS contours have been performed, as in Fig. 4(a). With increasing thickness, more discrete slices of the majority Fermi surface are detected. This evolution supports the interpretation as SW states.

The picture of high spin asymmetry receives confirmation from a DFT calculation of the layered structure. It is modeled by 9 ML Fe on 9 ML W (indefinitely repeated in order to obtain a band structure), see Fig. 4(b). The Fe lattice constant was used throughout, thereby admitting an approximation for the paramagnetic W side. Qualitatively similar results can be obtained with the W lattice constant. Already near the interface, Fe is ferromagnetic, with the magnetic moment assuming the bulk value of $\sim 2.2\mu_B$.

The spin-resolved band structure perpendicular to the interface along *N-H* is plotted in Fig. 4(c). Majority electrons clearly exhibit SW states, characterized by discrete and dispersionless energy levels. This band structure is back-folded due to the supercell, so that the sequence of bulk levels in Fig. 2(b) now appears as a ladder of energies. The population of these states stems only from Fe orbitals (indicated by circle size), and is evenly distributed over the film without localization directly at the interface. The confinement for the two majority bands in this direction known from bulk Fe is particularly well expressed for the upper band (in a 1.5 eV window below E_F). In contrast, the minority states in Fig. 4(c) do *not* show quantization and follow a steep dispersion instead. Moreover, these minority bands comprise both Fe and W orbitals to roughly equal extent. This indicates that the minority wave functions extend well across the layer boundaries.

These results may be contrasted with findings in spin-resolved experiments. A spin-dependent electron transmission through ultrathin ferromagnetic films has been observed,[17] and is explained by spin-dependent elastic

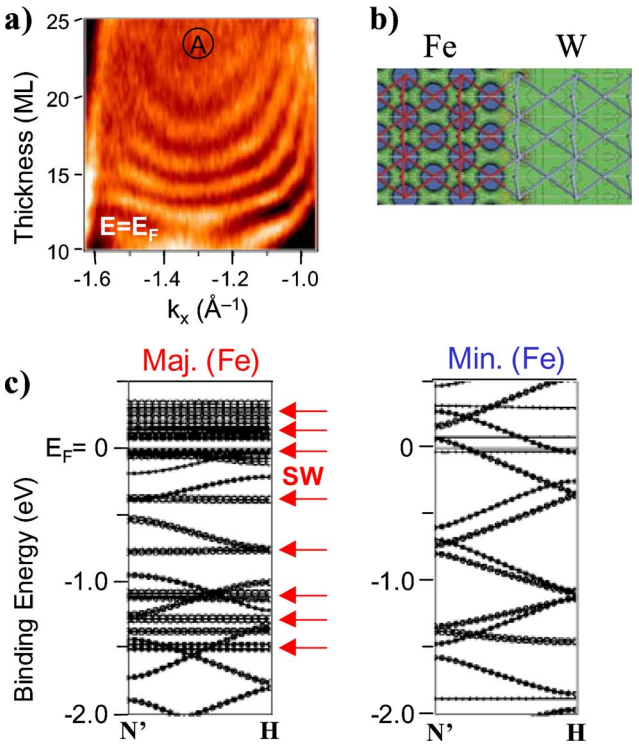

FIG. 4. (Color online) (a) Thickness dependence of SW states along a line scan ($h\nu$=112 eV) through majority contour A of Fig. 3. (b) Fe-W system used for DFT calculation with 9 ML each. The magnetization density (blue) shows Fe magnetized up to the interface. (c) Back-folded band structure perpendicular to the interface (contribution of Fe orbitals indicated by circle size). Majority states form dispersionless SW states exclusively populated by Fe electrons. Minority states disperse freely, with contributions from both Fe and W orbitals.

scattering.[18] Asymmetries in photoemission spectra can be derived from first-principles calculations.[19] A strong spin asymmetry like in our data is consistent with electron-beam transmission experiments.[20]

A consequence of the spin-selective confinement is the redistribution of the spin density of states near the E_F. The ARPES data indicate that it oscillates with thickness, and magnetic properties should thus be modulated. Such oscillations are indeed found for Fe films in the monolayer regime for the magnetic moments[21] and derived properties such as T_C.[22] The present experiment also bears relation to tunneling experiments with Fe contact layers on oxide barriers (which exclude exchange coupling).[3,23] Oscillations in the tunneling magnetoresistance occur as a function of bias and Fe film thickness.[3] It has been suggested that this may be due to quantum well states in the magnetic film.[4] Our results provide direct spectroscopic evidence for such states.

In comparing Fe/W to other commonly used interfaces, the most closely related system is Fe/Cr.[24] The FS of Cr closely resembles that of W, therefore one can expect similar results for the Fe/Cr interface. Also, Mo is a candidate for a matched Fermi surface. In all these cases, the band filling of one spin of the ferromagnet equals the band filling of the paramagnet, although due to varying bandwidths the match need not be optimal at higher binding energies. Considering other elemental ferromagnets, fcc Co or Ni is usually combined with Cu layers. The FS of fcc Cu resembles the majority FS of both these ferromagnets. However, their minority FS is not much different in extension, and therefore the selective advantage for spin transmission cannot be large. Accordingly, SW states of *both* signs have been detected in Co on Cu(100) by inverse photoemission.[12] This is underlined by calculations[5,6] deriving a magnetoresistance in Co/Cu multilayers that is only about half the value compared to Fe/Cr,[6] in support of the FS viewpoint.

In conclusion, by using *k*-space mapping with ARPES, one can probe directly how the spin states of a ferromagnet are altered by interfacial reflection. A single electronically matched interface, based on FS topology, can represent a significant spin-selective factor. SWs at the Fermi level restricted to one spin sign can thereby be achieved. As an outlook, it would be interesting to evaluate the degree of spin filtering in the conductance. Likewise the standing wave evolution with thickness should modulate the transmission properties.

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[1] E. E. Fullerton, D. M. Kelly, J. Guimpel, I. K. Schuller, and Y. Bruynseraede, Phys. Rev. Lett. **68**, 859 (1992).
[2] T. Banerjee, E. Haq, M. H. Siekman, J. C. Lodder, and R. Jansen, Phys. Rev. Lett. **94**, 027204 (2005).
[3] T. Nagahama, S. Yuasa, Y. Suzuki, and E. Tamura, J. Appl. Phys. **91**, 7035 (2002).
[4] Z.-Y. Lu, X.-G. Zhang, and S. T. Pantelides, Phys. Rev. Lett. **94**, 207210 (2005).
[5] P. Zahn, I. Mertig, M. Richter, and H. Eschrig, Phys. Rev. Lett. **75**, 2996 (1995).
[6] K. M. Schep, P. J. Kelly, and G. E. W. Bauer, Phys. Rev. B **57**, 8907 (1998).
[7] J. E. Ortega, A. Narmann, K. N. Altmann, W. O'Brien, D. J. Seo, F. J. Himpsel, P. Segovia, A. Mascaraque, and E. G. Michel, J. Magn. Magn. Mater. **203**, 126 (1999).
[8] R. K. Kawakami, E. Rotenberg, H. J. Choi, E. J. Escorcia-Aparicio, M. O. Bowen, J. H. Wolfe, E. Arenholz, Z. D. Zhang, N. V. Smith, and Z. Q. Qiu, Nature (London) **398**, 132 (1999).
[9] N. V. Smith, N. B. Brookes, Y. Chang, and P. D. Johnson, Phys. Rev. B **49**, 332 (1994).
[10] D.-A. Luh, J. J. Paggel, T. Miller, and T.-C. Chiang, Phys. Rev. Lett. **84**, 3410 (2000).
[11] L. Aballe, C. Rogero, P. Kratzer, S. Gokhale, and K. Horn, Phys. Rev. Lett. **87**, 156801 (2001).
[12] D. H. Yu, M. Donath, J. Braun, and G. Rangelov, Phys. Rev. B **68**, 155415 (2003).
[13] P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka, and J. Luitz, Computer Code WIEN2K, Vienna, 2001.
[14] J. Schäfer, M. Hoinkis, E. Rotenberg, P. Blaha, and R. Claessen, Phys. Rev. B **72**, 155115 (2005).
[15] R. Popescu, H. L. Meyerheim, D. Sander, J. Kirschner, P. Steadman, O. Robach, and S. Ferrer, Phys. Rev. B **68**, 155421 (2003).
[16] H. Bethge, D. Heuer, C. Jensen, K. Reshoft, and U. Kohler, Surf. Sci. **333**, 878 (1995).
[17] D. P. Pappas, K.-P. Kämper, B. P. Miller, H. Hopster, D. E. Fowler, C. R. Brundle, A. C. Luntz, and Z.-X. Shen, Phys. Rev. Lett. **66**, 504 (1991).
[18] M. P. Gokhale and D. L. Mills, Phys. Rev. Lett. **66**, 2251 (1991).
[19] J. Henk, T. Michael, P. Bose, and P. Bruno, Surf. Sci. **566**, 252 (2004).
[20] D. Oberli, R. Burgermeister, S. Riesen, W. Weber, and H. C. Siegmann, Phys. Rev. Lett. **81**, 4228 (1998).
[21] C. M. Fang, R. A. de Groot, M. M. J. Bischoff, and H. van Kempen, Phys. Rev. B **58**, 6772 (1998).
[22] M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Phys. Rev. Lett. **85**, 5424 (2000).
[23] C. Tiusan, J. Faure-Vincent, C. Bellouard, M. Hehn, E. Jouguelet, and A. Schuhl, Phys. Rev. Lett. **93**, 106602 (2004).
[24] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).