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# Electronic and magnetic properties of ultrathin films and interfaces investigated by dichroic core-level photoelectron spectroscopy

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## Abstract

The electronic and magnetic structures of ultrathin films made of a ferromagnetic and a nonmagnetic material are theoretically investigated by means of magnetic dichroism in spin- and angle-resolved core-level photoelectron spectroscopy. How these properties manifest themselves in the photoemission intensities is analyzed with a focus on the interface between film and substrate. The dependence on both exchange and spin–orbit splitting, magnetic ordering, core-level shift and on the thickness of the covering layer are investigated in detail. Ultrathin films of Fe and Pd serve as prototypical systems because of their large exchange and spin–orbit splittings, respectively.

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## 1. Introduction

Transport properties of electrons through planar junctions of magnetic and nonmagnetic materials depend to a great extent on the geometric, electronic, and magnetic structures at the interfaces, i.e. the common boundaries of the constituents. Therefore, it is necessary to obtain detailed knowledge of the interfacial properties to understand completely spin-dependent transport [1]. To gain access to these properties experimentally is still a challenge.

Experimentally, an interface can be realized by the common boundary of a thin film and the substrate on which it is deposited. Since the interface is ‘hidden’ by the covering film, one needs a local probe, preferably with both energy and spin resolution. One of the most successful techniques with the required properties is spin- and angle-resolved photoelectron spectroscopy (PES; for an overview, see Ref. [2]). First, through emission from core levels one can discriminate between the different elements that make up the system. Second, spin-resolved PES provides direct access to the magnetic properties (although one has to keep in mind that the measured spin polarization of the photoelectron is in general not that of the initial state). Third, magnetic dichroism (MD) supplies additional information about the

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electronic and magnetic structure. One drawback of PES in this context has to be mentioned: due to the comparably small photoelectron escape depth [3,4], the thickness of the covering films has to be restricted to a few monolayers (ML). In a recent publication, Le Cann et al. investigated the magnetism of Fe/Pd(001) by means of dichroism in both X-ray absorption spectroscopy (XAS) and core-level photoemission [5]. Although using two well-established spectroscopies they pointed out that “these experiments show that the MCD (magnetic circular dichroism) and MLD (magnetic linear dichroism) in core-level photoemission are far from a simple experimental fingerprint of magnetic thin films, surfaces, and interfaces.” Hence unfortunately, there seems to exist no published analysis that systematically reveals how the electronic and magnetic properties are coded in spin-resolved magnetic dichroism in core-level photoemission. In this paper, we hope to give some answers on this topic.

Another powerful tool for the analysis of magnetic systems is X-ray absorption spectroscopy (XAS; see e.g. Refs. [6–9]). For example, the local spin and orbital magnetic moments can be obtained via sum rules (Ref. [10] and references therein). However, XAS can be a rather indirect way to investigate the electronic and magnetic properties since it does not provide direct access to the electronic spin, in contrast to spin-resolved photoemission. Further, (M)EXAFS [(magnetic) extended X-ray absorption fine structure] requires support by theoretical calculations: [11] the oscillations in the experimental spectra are typically analyzed by means of (spin-polarized) multiple-scattering theory [12]. We would like to mention that interface properties can also be obtained with ferromagnetic resonance (FMR) and Brillouin light scattering (BLS). But compared to XAS and PES these methods can be viewed as even more ‘indirect’, in particular since they rely on reference systems (see e.g. Ref. [13]).

In a previous publication [14], we described successfully spin-resolved dichroic photoemission from Fe-3p and Ni-3p core levels (see also Ref. [15]). In that work, we focused on effects of photoelectron diffraction in emission from surface systems. In the present paper, we concentrate on how properties of films and interfaces manifest themselves in the PE spectra. All aspects of the systems

chosen can be analyzed theoretically in terms of layer-resolved magnetic and electronic quantities (for example, magnetic moments and spectral densities). These allow a detailed understanding of the key features that appear in the dichroic PE spectra.

Paramagnetic films on a ferromagnetic substrate [Pd/Fe(001)] and ferromagnetic films on a paramagnetic substrate [face-centered-cubic Fe/Pd(001)] serve as prototypical systems. The two constituents, Fe and Pd, possess characteristic properties. Iron—as the typical ferromagnet—shows a large magnetic moment, and magnetic dichroism in both core-level and valence-band photoemission is sizable despite the rather weak spin–orbit interaction [16–18]. A particularly interesting element is Pd which can be regarded as ‘close to being magnetic’ [13]. Although usually nonmagnetic, bulk Pd turns ferromagnetic if the lattice spacing is enhanced [19–21]. Further, Pd films can become magnetic due to quantum-size effects [21,22]. Hence, one can expect that due to the Fe-3d–Pd-4d hybridization Fe acts as trigger for turning Pd layers magnetic (at least those Pd layers close to the Fe–Pd interface should show a measurable magnetic moment). A question arises whether these magnetic moments, although small, can be accessed via magnetic dichroism.

In this paper, we focus on the Pd-3d core levels as initial states in PES. With binding energies of 340.5 eV ( $3d_{3/2}$ ) and 335.2 eV ( $3d_{5/2}$ ), spin–orbit splitting is much larger than the induced exchange splitting for these states. Less bound initial states, like the Fe-3p and the Pd-4p states, would lend themselves support for a theoretical investigation. For Fe-3p (about 53 eV binding energy), spin–orbit and exchange splitting are about the same size. But the rather short photohole lifetimes would complicate access to the interface properties. The Pd-4p states show binding energies close to those of the Fe-3p states. But due to super Coster–Kronig Auger transitions the PES maxima are too broad to yield any quantitative information (cf. Fig. 3 in Ref. [23]). Nevertheless, we shall briefly address the core-level shifts of these states.

As mentioned earlier, element-specific properties can be distinguished by sampling from a selected core level. Interface and surface core-level shifts allow a further resolution with respect to layers. The magnetism induced by the ferromagnetic part in the

‘nonmagnetic’ one manifests itself in two ways: in a nonzero spin polarization of the photoelectron and in magnetic dichroism. For Fe/Pd(001), the spin polarization of the excited electrons in the Pd substrate is due to spin–orbit coupling or due to the magnetic moment induced by the Fe film. These spin-polarized electrons are transferred through the Fe film towards the detector in a spin-dependent manner. An interesting question arises how the PE spectra are modified by this Fe spin filter [24] and whether one can conclude on the properties of the Pd substrate. In summary, we expect a variety of effects to be investigated by means of magnetic dichroism in core-level photoemission (for reviews relevant to the subject, see Refs. [25] and [26]).

This paper is organized as follows. Numerical and theoretical aspects are addressed in Section 2. Section 3 presents and discusses the results for Pd films on Fe(001) (3.1) and for Fe films on Pd(001) (3.2). Concluding remarks are given in Section 4.

## 2. Theoretical aspects

The electronic and magnetic structures of Pd films on Fe(001) and Fe films on Pd(001) were obtained by ab initio calculations in the local spin-density approximation of density-functional theory. For the multiple-scattering computations with Perdew–Wang exchange–correlation potential [27] we applied the screened Korringa–Kohn–Rostoker (KKR) method for semi-infinite systems. In the first step, the bulk potentials were calculated self-consistently in the atomic-sphere approximation. In the subsequent calculations for the films, the bulk potentials were kept fixed. Six substrate layers, the potentials of which were varied, served as buffer between the film layers and the bulk. The energy integration was performed by a 23-point Gaussian method on a semi-circle in the complex energy plane. For the Brillouin-zone integration 400 special points were used and the maximum angular momentum was  $l_{\max} = 3$ . More details on our computational method can be found in Refs. [28] and [29].

The photoemission calculations were performed within the one-step model of photoelectron spectroscopy, as formulated within the spin-polarized relativistic layer-KKR method. By this means, photo-

electron diffraction was fully taken into account. All film layers and the subsequent twenty substrate layers contributed to the photocurrent. Convergence in both the intensity and the photoelectron spin polarization was achieved for maximum angular momentum  $l_{\max} = 6$  and about 50 in-plane reciprocal lattice vectors for kinetic energies of about 35 eV. We would like to emphasize that our method worked well for magnetic dichroism from valence bands of transition metals (Fe, Co, Ni; see, for example Ref. [30]). In addition, photoemission from Fe-3p states was described successfully [14]. Of course, one has to keep in mind that within the single-particle picture no multiplet structure is present. However, the main lines (with low binding energy) should be described correctly. To include lifetime broadening, we employed self-energy corrections modeled by a spin-independent imaginary part of the optical potential [14]. Hence, the spin-dependence of the transmission through the covering film is due its spin-split electronic structure. Reasonable values were chosen for the free parameters (e.g. photohole and -electron lifetimes), but these were not optimized because in this paper we are concerned with elaborating the main effects rather than with ‘recalculating’ experimental spectra.

The geometric structures of the semi-infinite systems were taken from Ref. [31]. In the case of Pd/Fe(001), we are concerned with tetragonally distorted body-centered adlayers (bct) with interlayer distances enhanced by 12.7% with respect to the Fe bulk. Depending on the preparation conditions, Fe grows in two distinct ways on Pd(001): either body-centered tetragonal or in continuation of the ideal Pd fcc lattice. In a recent LEED investigation, Lee et al. [32] found capping by Pd and surface alloying for ultrathin Fe films grown on Pd at room temperature. Both effects certainly affect the photoemission spectra with respect to the ideal case (fcc-Fe). In this paper, however, only results for the ideal case are presented. We shall compare the magnetic and electronic structures of the different phases in a future publication [33].

In the forthcoming sections, magnetic linear dichroism (MLD) in spin-resolved photoemission with detection normal to the surface ( $\vartheta_e = 0^\circ$ , that is,  $\vec{k}_{\parallel} = 0$ ) is investigated. For MLD in standard set-up, p-polarized light impinges within a mirror-plane of

the lattice off-normally onto the surface ( $\vartheta_{\text{ph}} = 45^\circ$ ) and perpendicular to the in-plane magnetization  $\vec{M}$ . Hence, the photoelectron spin polarization  $\vec{P}$  is parallel to  $\vec{M}$ . Note that magnetic circular dichroism (MCD) requires also off-normally incident light which leads to all Cartesian components of  $\vec{P}$  being non-zero [34]. Thus, in view of the analysis (Section 3) we prefer MLD to MCD in the case of in-plane magnetization (for Fe/Pd(001) the magnetization was found to be in-plane [5]).

From the preceding it is obviously sufficient to work with the orientations of  $\vec{P}$  and  $\vec{M}$ . The spin polarization  $P$  of the photoelectron can be decomposed into parts odd and even in  $M$ ,  $P(M) = P_{\text{ex}}(M) + P_{\text{so}}(M)$ , with  $P_{\text{ex}}(M) = -P_{\text{ex}}(-M)$  and  $P_{\text{so}}(M) = P_{\text{so}}(-M)$ . Therefore, the spin-resolved dichroic intensities can be expressed as

$$I_{\sigma\tau} = \frac{1}{2} [1 + \sigma(P_{\text{so}} + \tau P_{\text{ex}})] I_{\tau}, \quad \sigma, \tau = \pm \quad (1)$$

with  $I_{\tau} = \sum_{\sigma} I_{\sigma\tau}$ . The first and the second index indicate the orientation of the spin-quantization axis (up/down) and the magnetization direction ( $\pm M$ ), respectively. The three asymmetries can be written as

$$A_{\text{av}} = \frac{I_+ - I_-}{I_+ + I_-} \quad (2a)$$

$$A_{\text{so}} = P_{\text{so}} + P_{\text{ex}} A_{\text{av}} \quad (2b)$$

$$A_{\text{ex}} = P_{\text{ex}} + P_{\text{so}} A_{\text{av}} \quad (2b)$$

For vanishing spin-orbit coupling, there is no dichroism ( $I_+ = I_-$ ,  $A_{\text{av}} = 0$ ) and  $I_{\pm\pm} = I_{\mp\mp}$ . Thus,  $P_{\text{so}} = A_{\text{so}} = 0$ , but  $A_{\text{ex}} = P_{\text{ex}}$ . For vanishing exchange splitting there is also no dichroism, and  $I_{\pm\pm} = I_{\pm\mp}$ . This results in  $P_{\text{ex}} = A_{\text{ex}} = 0$ , but  $A_{\text{so}} = P_{\text{so}}$ . In conclusion,  $A_{\text{av}}$  is the commonly used spin-averaged asymmetry, whereas  $A_{\text{so}}$  and  $A_{\text{ex}}$  probe asymmetries (spin polarizations) which can be attributed mainly to spin-orbit coupling and exchange splitting, respectively [35].

### 3. Results and discussion

#### 3.1. Pd/Fe(001)

##### 3.1.1. Magnetic structure

The layer-resolved magnetic moments (magnetic profiles) for Pd films on Fe(001) are shown in Fig. 1.

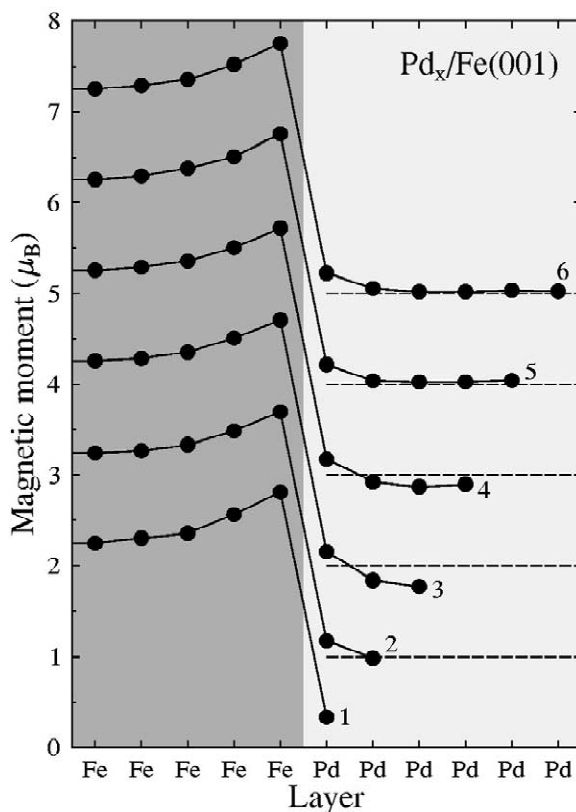


Fig. 1. Layer-resolved magnetic moments of Pd films on Fe(001) with  $x$  ML thickness [Pd $_x$ /Fe(001)],  $x = 1, \dots, 6$ . The curves, each of them indicated by  $x$ , are shifted for clarity, with associated zero lines given at the right (dashed).

We find a strong increase of the magnetic moment of the outermost Fe layer (of about 22% compared to the bulk value) which was already observed for the uncovered Fe surface [36].

Hybridization of the valence d-states of Fe and Pd leads to a sizable magnetic moment at Pd layers close to the Fe substrate. The adjacent Pd layer couples ferromagnetically to Fe, independent of the Pd-film thickness [37]. Its magnetic moment is largest for the 1-ML film ( $0.33 \mu_{\text{B}}$ ) and decreases with film thickness towards an average value of about  $0.2 \mu_{\text{B}}$  [31]. Interestingly, the remaining Pd layers of 2-, 3-, and 4-ML films couple anti-ferromagnetically to Fe. In particular for the 3-ML film, the surface magnetic moment is as large as  $0.22 \mu_{\text{B}}$ . For thicker films, all layers couple ferromagnetically to Fe and the magnetic moment tends to zero, as expected in the limit towards bulk Pd.

In summary, we find a variety of magnetic profiles in the Pd films. (Because these appear interesting in their own right, we shall investigate them in more detail in a future publication [33]). The next question that arises is how these profiles manifest themselves in the Fe and Pd core levels.

### 3.1.2. Electronic structure

Before turning to the spectral densities in the core-level regime, we would like to note that in the presence of both exchange and spin–orbit splitting the nomenclature ( $j, m_j$ ) for the core levels is inaccurate because the spin–orbit interaction couples states with different total angular momentum  $j$  but with the same  $z$ -projection  $m_j$  of  $j$ . However, we shall use this labeling since there is no crossing of levels with interaction strength and each state can still be unambiguously identified by ( $j, m_j$ ) [14].

As typical examples, we address the layer- and spin-resolved spectral densities of Fe-3p and Pd-4p states of a 5-ML Pd film (Fig. 2). Fuggle and Mårtensson give a binding energy of 52.7 eV for Fe-3p (Ref. [38]) which compares nicely with the theoretical mean binding energy of 52.5 eV. Spin–orbit and exchange splitting are approximately of the same size [14]. For the Pd-4p levels, Fuggle and Mårtensson give 55.7 eV ( $4p_{1/2}$ ) and 50.9 eV ( $4p_{3/2}$ ) which agrees well with the data of Nagel et al. [23]. Evidently, spin–orbit splitting is much stronger than the induced exchange splitting. For example, the central layers of a 6-ML Pd film show binding energies of 54.7 and 50.2 eV in theory.

The surface and interface core-level shifts allow one to distinguish between states located in different layers. The Fe states tend to higher binding energies at the Pd–Fe interface, whereas the Pd states show the opposite behavior. At the Pd surface, the 4p states show an increase in binding energy. These findings can be transferred qualitatively to systems with different film thickness or to other core states. Note that the layer-dependent core-level shifts are obtained within the initial-state picture and not within the more sophisticated screened impurity model [39]. Therefore, experimental core-level shifts might differ from our values.

The magnetic ordering and the magnetic profiles (Fig. 1) are reflected in the spin-resolved density of states and the level splittings. The  $4p_{1/2}$  and the  $4p_{3/2}$  levels of the Pd layer adjacent to Fe (labeled

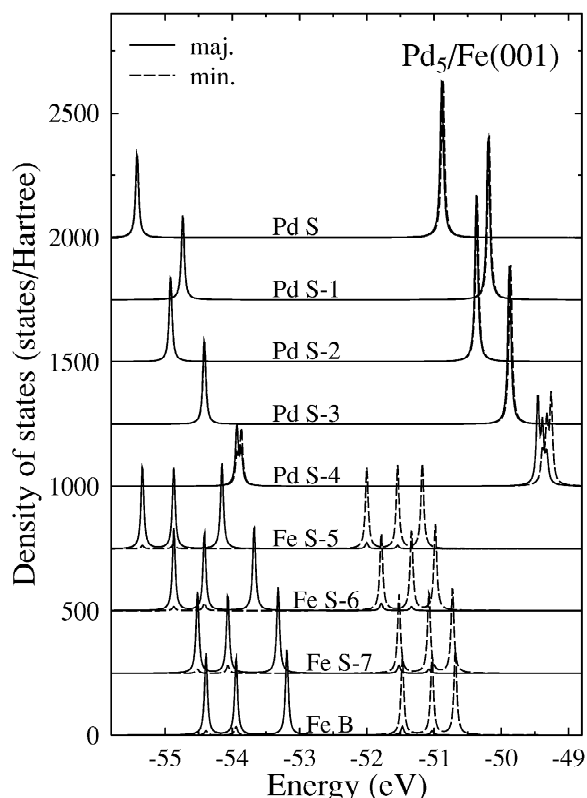


Fig. 2. Layer- and spin-resolved density of states of Fe-3p and Pd-4p states of a 5-ML Pd film on Fe(001). The energy is relative to the Fermi level. Solid and dashed lines indicate majority and minority spin orientation (with respect to the Fe bulk), respectively.

‘Pd S-4’; magnetic moment  $0.22 \mu_B$ ) are clearly split into the  $m_j$  sublevels. The ferromagnetic coupling to the Fe substrate is evident from the fact that the states with minority orientation (with respect to the Fe bulk) are highest in energy. The comparably small magnetic moment of the remaining Pd layers (less than  $0.04 \mu_B$ ) is accompanied by the very small sublevel splitting.

### 3.1.3. Magnetic linear dichroism

In Fig. 3a, results of MLD calculations for Pd-3d states in the standard set-up (see inset in panel (b)) are shown. The spin–orbit splitting of 5.6 eV agrees well with that given by Fuggle and Mårtensson (5.3 eV) and that found experimentally by Nagel et al. [23,38]. Because the exchange splitting (for example, 0.03 and 0.06 eV for the  $3d_{3/2}$  and  $3d_{5/2}$  levels,

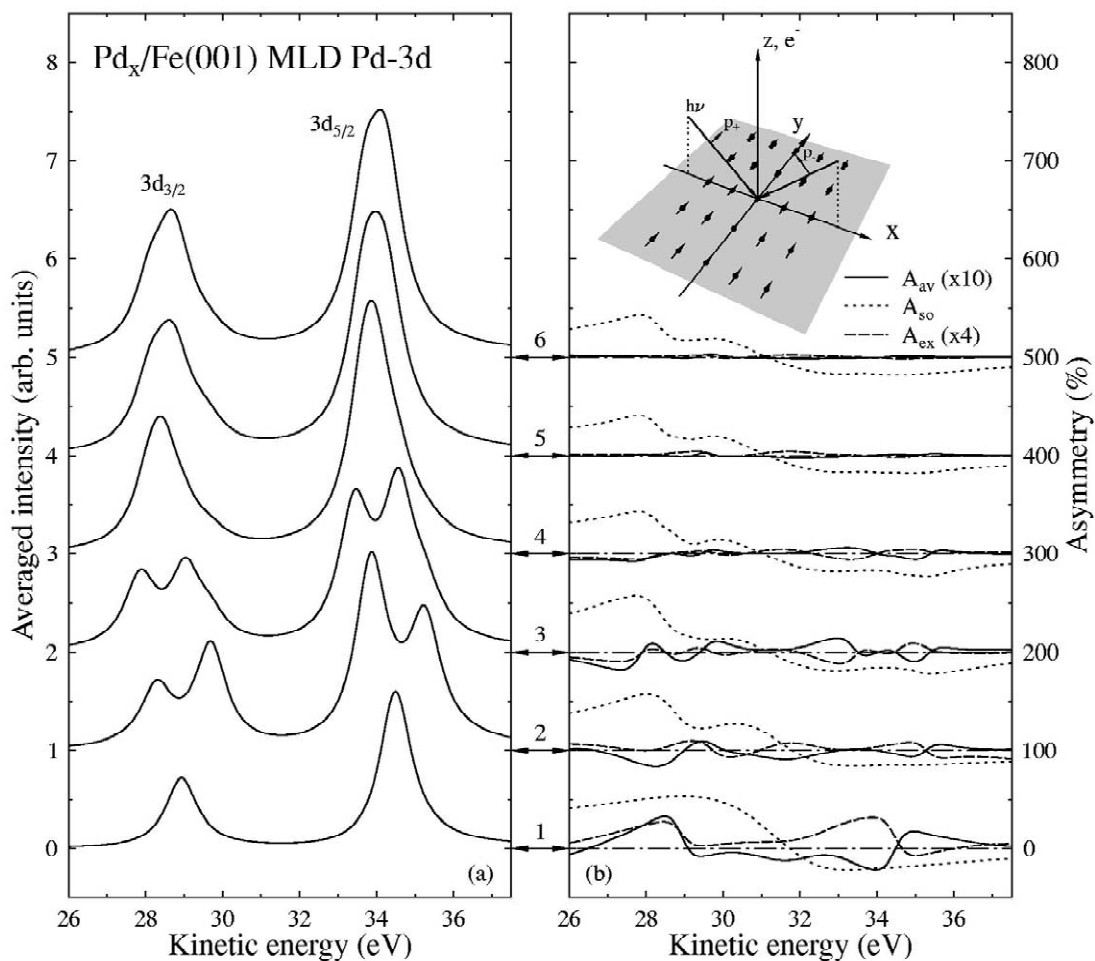


Fig. 3. Magnetic linear dichroism (MLD) from Pd-3d states of Pd films on Fe(001) with  $x$  ML thickness,  $x = 1, \dots, 6$ , as indicated between the panels. Left (a): Averaged intensities [ $I_+ + I_-$ , Eq. (1)] are evenly shifted for clarity. Arrows on the right indicate the respective zero intensity. Right (b): The spin-averaged ( $A_{av}$ ), the spin-orbit ( $A_{so}$ ), and the exchange ( $A_{ex}$ ) related asymmetries [Eq. (2)] are represented by solid, dotted, and dashed lines, respectively, and are shifted for clarity. The respective zero lines are displayed dash-dotted. The inset shows schematically the standard MLD set-up.

respectively, at Pd layers adjacent to the Fe substrate) is much less than the lifetime broadening, the exchange-split sublevels cannot be resolved (cf. Figs. 1 and 2 in Ref. [23]). Hence, we find for the 1-ML film only a single pair of spin-orbit split maxima. For the 2-ML film, these are further split due to core-level shifts, the maxima higher in energy belonging to the subsurface Pd-layer S-1 (cf. Fig. 2). With increasing film thickness, the spectra evolve towards those of semi-infinite Pd(001). However, even for the 6-ML film, faint hints of core-level

shifts can be found (Note that the maxima are significantly broader than for the 1-ML film which indicates that they are built-up by several overlapping peaks).

Fig. 3b displays the asymmetries defined in Eq. (2). As expected for the 3d states, the spin-orbit asymmetry  $A_{so}$  (dotted) is prominent for all film thicknesses and shows a typical broad  $+/-$  shape, indicating that the  $d_{3/2}$  ( $d_{5/2}$ ) states produce a positive (negative) spin polarization  $P_{so}$ . Because of the small magnetic moment in the Pd layers, the

magnetic dichroism as reflected by  $A_{av}$  is small, too. This asymmetry (solid in Fig. 3b) decreases rapidly with film thickness due to the small photoelectron escape depth [3,4]. For the 1-ML film, one finds the typical  $+ / - / - / +$  shape. For the 2-ML case, however,  $A_{av}$  shows additional oscillations due to occurrence of level-shifted maxima. The  $3d_{3/2}$  maximum lowest in energy (at 28.3 eV) produces a  $- / +$  feature which might indicate anti-ferromagnetic coupling to the Fe moments. Indeed, inspection of Fig. 1 shows that the surface layer couples antiferromagnetically to Fe. Further, reverting the Pd magnetic moments produces a  $+ / -$  feature. The  $3d_{5/2}$  maximum highest in energy (at 35.3 eV) shows a  $- / +$  shape, indicating ferromagnetic coupling. Evidently, this maximum is due to the interface (subsurface) Pd layer S-1. The exchange-related spin polarization is reflected by  $A_{ex}$  which for the 1-ML film is positive except for the least bound sublevels (with energy larger than 34.5 eV) [14].

A similar analysis is apparently possible for the thicker films. In particular,  $A_{ex}$  in conjunction with  $A_{av}$  appears helpful in retrieving information on the magnetic structure from the spectra. However, in experiments one is practically limited to Pd films a few ML thick. Otherwise  $A_{ex}$  and  $A_{av}$  become too small to be reliably detected.

### 3.2. Fe/Pd(001)

#### 3.2.1. Magnetic structure

The layer-resolved magnetic moments of fcc-Fe films on Pd(001) (Fig. 4) are significantly larger than that of bulk bcc-Fe ( $2.24 \mu_B$ ). The largest moment is found for the 1-ML film ( $3.14 \mu_B$ ), which agrees almost perfectly with a value calculated by Lee et al. ( $3.13 \mu_B$ ; Ref. [32]). The least moments are found in the interior of the 5- and 6-ML films ( $2.80 \mu_B$ ). In addition, we observe the typical increase at the two interfaces (Pd–Fe and Fe–vacuum).

In clear contrast to the rather complex behavior of Pd/Fe(001), the magnetic structure of the Pd layers close to the Fe films is almost independent of the Fe-film thickness. For all film thicknesses, Pd couples ferromagnetically to the Fe film, with an induced magnetic moment of about  $0.24 \mu_B$  at the interface. Due to the slightly enhanced Fe-moment, the induced moment is  $0.28 \mu_B$  in the 1-ML case

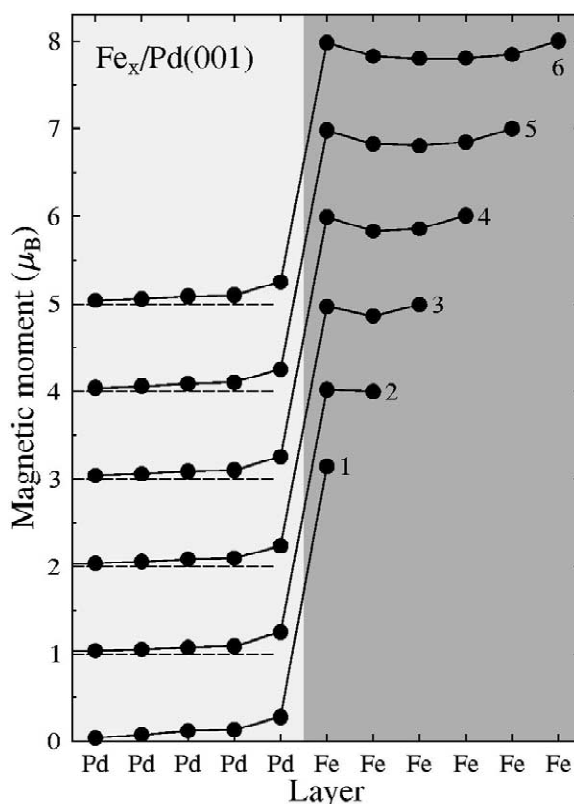


Fig. 4. Same as Fig. 1, but for layer-resolved magnetic moments of fcc-Fe films on Pd(001).

which agrees well with that found by Blügel et al. ( $0.32 \mu_B$ ) [40].

#### 3.2.2. Electronic structure

As a representative of all investigated Fe films, we present in Fig. 5 the Fe-3p and Pd-4p density of states of a 5-ML film. Comparing with Fig. 2, the core-level shifts are rather moderate. The most striking facts are a surface core-level shift to higher binding energies (Fe layer S) and the  $m_j$ -level splitting of the Pd interface layers (Pd layer S-5 with 0.2 eV exchange splitting).

#### 3.2.3. Magnetic linear dichroism

The dichroic PE intensities from the Pd-3d states are shown in Fig. 6a for various thicknesses of the covering Fe film (photon energy 355 eV). The intensities decrease with increasing film thickness due to the mean free path of the photoelectrons. A

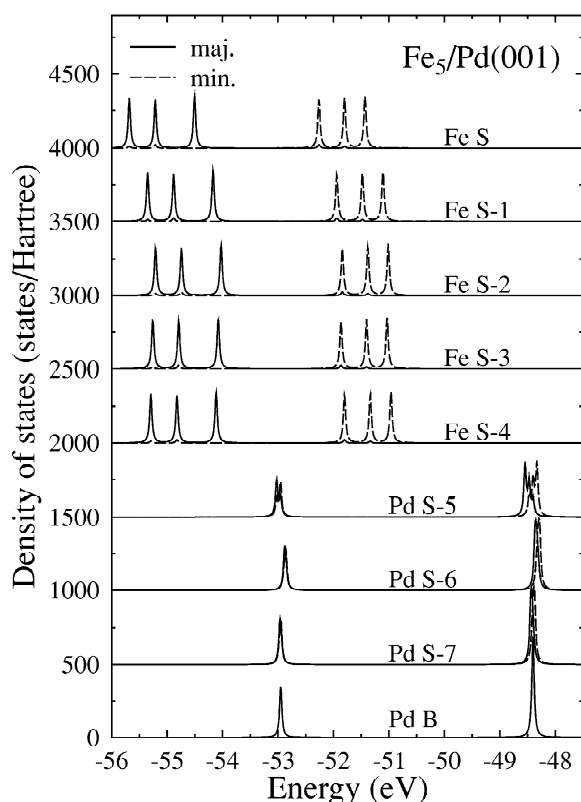


Fig. 5. Same as Fig. 2, but for Pd-4p and Fe-3p states of a 5-ML fcc-Fe film on Pd(001).

quantitative analysis for the  $3d_{3/2}$  maxima confirms the exponential decay and yields an escape depth  $\lambda$  of 2.86 ML for the averaged intensities (i.e.  $I_+ + I_-$ ). For the  $3d_{5/2}$  maxima, we obtain  $\lambda = 2.78$  ML. Hence, the escape depth decreases slightly with kinetic energy. (Calculations for a few ten eV larger kinetic energies show the same trend; cf. also Ref. [3]). Our data correspond very well with those determined by Tamura et al. [41].

An analysis of the dichroic intensities  $I_{\pm}$  establishes escape depths of  $\lambda_+ = 2.88$  ML and  $\lambda_- = 2.85$  ML for the  $3d_{3/2}$  maxima, i.e.  $\lambda_+ > \lambda_-$ . For the  $3d_{5/2}$  maxima we find  $\lambda_+ < \lambda_-$ , in particular  $\lambda_+ = 2.76$  ML and  $\lambda_- = 2.80$  ML. Since the photoelectron spin polarization is dominated by the spin-orbit related one (cf.  $A_{so}$  in Fig. 6b), we can conclude that in this range of kinetic energies Fe films are generally slightly more transparent for majority electrons than for minority electrons (see also Refs. [42] and

[43]). A naive interpretation (without knowing the main origin of the photoelectron spin polarization) as the  $3d_{3/2}$  initial states being prominently ‘majority’ and the  $3d_{5/2}$  initial states being ‘minority’ states would of course be erroneous. This is immediately proven by inspection of  $A_{ex}$  in Fig. 6b which for the thinner films shows  $+/-$  shapes at both the  $3d_{3/2}$  and the  $3d_{5/2}$  maxima. Evidently, both PE peaks are made-up of both ‘majority’ and ‘minority’ states. With increasing film thickness, however,  $A_{ex}$  shifts towards larger values, a fact which can be explained by the spin-filter effect.

A description of the spin-filter effect [24] can be achieved by simply modeling the attenuation via spin-dependent escape depths. The ‘internal’ photocurrent  $I_{\sigma\tau}$  that is present in the substrate is then damped exponentially to yield the ‘external’ one  $I_{\sigma\tau}(d)$  at the detector:  $I_{\sigma\tau}(d) = I_{\sigma\tau} \exp(-d/\lambda_{\sigma\tau})$ , with  $d$  the Fe-film thickness. This simple model is applicable only if there is no emission from the film at that particular binding energy and if there is no significant spin-flip scattering in the ferromagnetic film. Whereas the former requirement is obviously fulfilled, the latter should be fulfilled to a very good approximation for Fe due to its comparably small strength of the spin-orbit interaction and due to the comparably large kinetic energy. (A slightly more sophisticated description of spin-dependent electron scattering would follow the outline given in Chapter 3 of Ref. [44].)

To elucidate the spin-filter effect in more detail, we have calculated MLD for two additional model systems. With setting the Fe layers artificially non-magnetic and keeping the Pd layers magnetic, the spin-filter effect is switched off and only the ‘native Pd dichroism’ remains. On the other hand, turning off the Pd magnetism while keeping the Fe magnetism reveals the dichroism that is exclusively due to the spin-filter effect. The asymmetries which result from these spectra are shown in Fig. 7 for 3 ML Fe on Pd(001). In all three cases, the asymmetry  $A_{so}$  reflects almost exclusively the spin-orbit induced spin polarization  $P_{so}$  because of the small  $A_{av}$ : positive for the  $3d_{3/2}$  initial states, negative for the  $3d_{5/2}$  initial states. Since  $A_{so}$  is almost identical in all three calculations, we can conclude that it represents the spin polarization of the photoelectrons in the Pd substrate. The exchange-related asymmetry



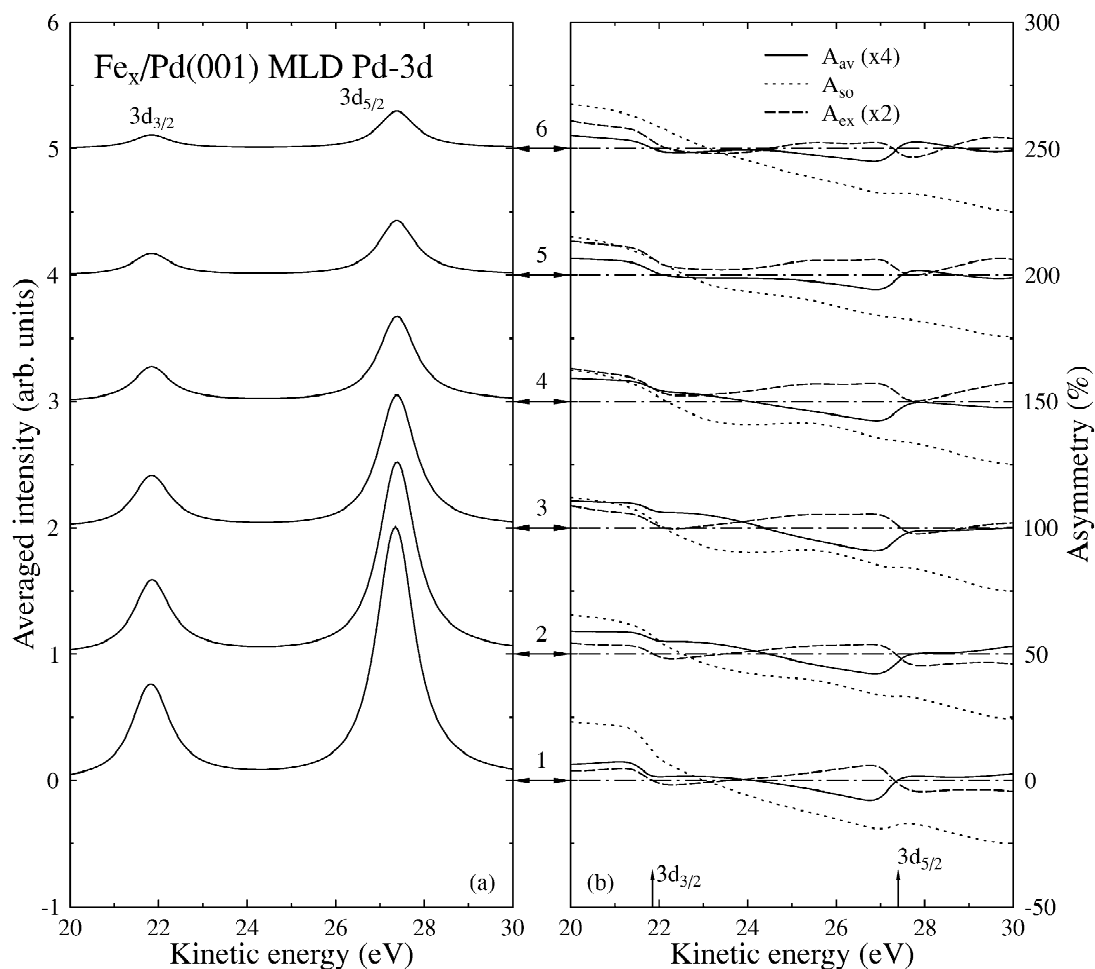


Fig. 6. Magnetic linear dichroism (MLD) from Pd-3d states of fcc-Fe films on Pd(001) with  $x$  ML thickness,  $x = 1, \dots, 6$ . Left (a): Averaged intensities  $[I_+ + I_-]$ , Eq. (1). Right (b): Associated asymmetries ( $A_{av}$  solid,  $A_{so}$  dotted, and  $A_{ex}$  dashed). Vertical arrows indicate the peak positions.

$A_{ex}$  for the pure spin-filter case (dashed in Fig. 7b) is positive and, hence, the 3-ML Fe film is more transparent for majority photoelectrons than for minority ones in this range of kinetic energies. For the ‘native Pd dichroism’,  $A_{ex}$  displays the  $+/-$  shape at the peak positions, indicating the  $m_j$ -level splitting although this is not resolved in the spin-averaged intensities.

Noticing that the magnetic structures of the two artificial model systems add up to that of the ‘fully magnetic’ Fe–Pd system, one obtains a ‘sum rule’ for  $A_{av}$  and  $A_{ex}$ : the asymmetries of the two cases ‘native Pd dichroism’ (dotted) and ‘pure spin-filter

effect’ (dashed) sum up to those of the ‘fully magnetic’ case (solid). This rule is immediately evidenced by inspection of the zeroes in panels b and c: if either the dotted or the dashed line is zero, the other two asymmetries are equal. This feature explains the shifted shape of  $A_{av}$  in the ‘fully magnetic’ case. Typically, for instance in MLD from semi-infinite ferromagnetic systems,  $A_{av}$  shows a  $+/-/-/+$  shape, as it is represented by the dotted line (‘native Pd dichroism’) in panel c. The spin-filter effect shifts  $A_{av}$  to larger values at the  $3d_{3/2}$  maximum and to lower values near the  $3d_{5/2}$  maximum. This transformation retains the overall

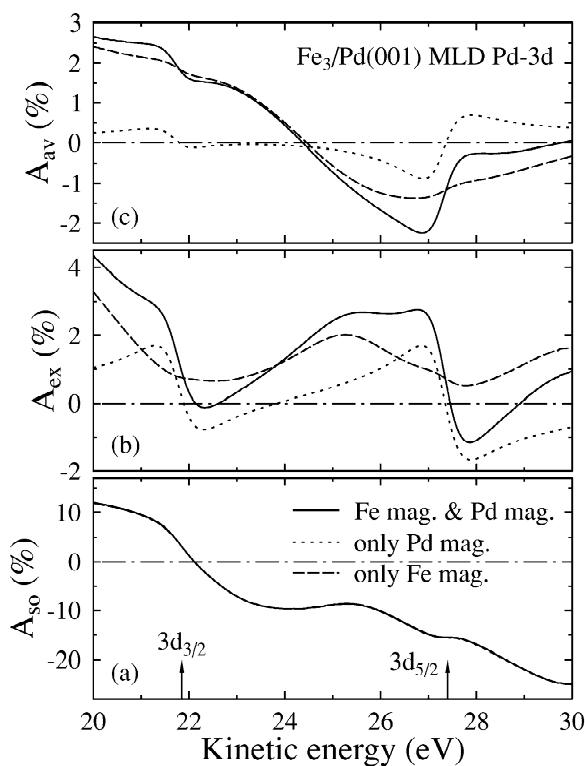


Fig. 7. Asymmetries of magnetic linear dichroism from Pd-3d states of a 3-ML fcc-Fe films on Pd(001). The spin-orbit ( $A_{so}$ , a), the exchange ( $A_{ex}$ , b), and the spin-averaged asymmetries ( $A_{av}$ , c) are displayed for three model systems: both Fe and Pd magnetic (solid), only Pd magnetic (dashed), and only Fe magnetic (dash-dotted). Vertical arrows in (a) indicate the peak positions (cf. Fig. 6).

shape of  $A_{av}$  with its two distinct wiggles at the peak positions.

These findings imply that PES from core levels in principle allows the determination of the spin- and energy-dependent electron escape depth. Further, it provides access to the magnetism of the substrate layers although it is covered by the ferromagnetic film and, hence, obscured by the spin-filter effect. The asymmetries defined in Eq. (2) proved to be essential for a detailed analysis, as is evidenced by the preceding discussion. It appears particularly helpful that they retrieve the main origin of the photoelectron spin polarization.

#### 4. Concluding remarks

With the present investigation we hope to have shown that spin-resolved dichroic PES provides a tool for detailed studies of the electronic and magnetic properties of film and interfaces. Since the calculated intensities serve exclusively (or at least to a great deal) as ‘input’, such an analysis could possibly be performed successfully without theoretical aid. Although practical limitations may arise from a small electron escape depth, too small induced magnetic moments in the ‘nonmagnetic’ constituent (here: Pd), and too small photohole lifetimes, we would like to encourage experimentalists to consider dichroic core-level PES for interface investigations.

The film-substrate systems chosen were built from Fe and Pd. In particular for Pd/Fe(001), these show rich geometric, electronic, and magnetic structures that are worth investigating in their own right. Hence, we shall focus on their properties for various substrate orientations [(001), (110)] and film structures (fcc, bct; ordered, alloyed) in a future publication [33].

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#### References

- [1] S.S.P. Parkin, J. Appl. Phys. 79 (1996) 6078.
- [2] M. Campagna, R. Rosei (Eds.), Photoemission and Absorption Spectroscopy of Solids and Interfaces With Synchrotron Radiation, North-Holland, Amsterdam, 1990.
- [3] M.P. Seah, W.A. Dench, Surf. Interf. Anal. 1 (1979) 2.
- [4] W.S.M. Werner, C. Tomastik, T. Cabela, G. Richter, H. Störi, Surf. Sci. 470 (2000) L123.
- [5] X. Le Cann, C. Boeglin, B. Carrière, K. Hricovini, Phys. Rev. B 54 (1996) 373.
- [6] H. Ebert, S. Ruegg, G. Schütz, R. Wienke, W.B. Zeper, J. Magn. Mater. 93 (1991) 601.
- [7] A. Nilsson, J. Stöhr, T. Wiell, M. Aldén, P. Bennich, N. Wassdahl, M.G. Samant, S.S.P. Parkin, N. Mårtensson, J.

- Nordgren, B. Johansson, H.L. Skriver, *Phys. Rev. B* 54 (1996) 2917.
- [8] S.S. Dhesi, E. Dudzik, H.A. Dürr, N.B. Brookes, G. van der Laan, *Surf. Sci.* 454–456 (1999) 930.
- [9] S.S. Dhesi, H.A. Dürr, E. Dudzik, G. van der Laan, N.B. Brookes, *Phys. Rev. B* 61 (2000) 6866.
- [10] P. Carra, *J. Electron Spectrosc. Relat. Phenom.* 86 (1997) 139.
- [11] P. Srivastava, F. Wilhelm, A. Ney, N. Haack, H. Wende, K. Baberschke, *Surf. Sci.* 402–404 (1998) 818.
- [12] A.L. Ankudinov, J.J. Rehr, *Phys. Rev. B* 56 (1997) R1712.
- [13] Z. Celinski, B. Heinrich, J.F. Cochran, W.B. Muir, A.S. Arrott, J. Kirschner, *Phys. Rev. Lett.* 65 (1990) 1156.
- [14] J. Henk, A.M.N. Niklasson, B. Johansson, *Phys. Rev. B* 59 (1999) 13986.
- [15] H. Ebert, G.-Y. Guo, *J. Magn. Magn. Mater.* 148 (1995) 178.
- [16] F.U. Hillebrecht, H.B. Rose, C. Roth, E. Kisker, *J. Magn. Magn. Mater.* 148 (1995) 49.
- [17] F.U. Hillebrecht, C. Roth, H.B. Rose, E. Kisker, M. Finazzi, L. Braicovich, *Phys. Rev. B* 51 (1995) 9333.
- [18] A. Rampe, G. Güntherodt, D. Hartmann, J. Henk, T. Scheunemann, R. Feder, *Phys. Rev. B* 57 (1998) 14370.
- [19] V.L. Moruzzi, P.M. Marcus, *Phys. Rev. B* 39 (1989) 471.
- [20] H. Chen, N.E. Brener, J. Callaway, *Phys. Rev. B* 40 (1989) 1443.
- [21] S. Mirbt, H.L. Skriver, B. Johansson, *Phys. Rev. B* 53 (1996) R13310.
- [22] A.M.N. Niklasson, S. Mirbt, H.L. Skriver, B. Johansson, *Phys. Rev. B* 56 (1997) 3276.
- [23] S.R. Nagel, G.B. Fisher, J. Tauc, B.G. Bagley, *Phys. Rev. B* 13 (1976) 3284.
- [24] W. Kuch, M.-T. Lin, K. Meinel, C.M. Schneider, J. Noffke, J. Kirschner, *Phys. Rev. B* 51 (1995) 12627.
- [25] M. Alouani, H. Dreyse, *Curr. Opin. Solid State Mater. Sci.* 5 (1999) 499.
- [26] R. Wu, A.J. Freeman, *J. Magn. Magn. Mater.* 200 (1999) 498.
- [27] J.P. Perdew, Y. Wang, *Phys. Rev. B* 45 (1992) 13244.
- [28] A. Ernst, G. van der Laan, W.M. Temmerman, S.S. Dhesi, Z. Szotek, *Phys. Rev. B* 62 (2000) 9543.
- [29] A. Ernst, M. Lüders, W.M. Temmerman, Z. Szotek, G. van der Laan, *J. Phys.: Condens. Matt.* 12 (2000) 5599.
- [30] R. Feder, J. Henk, in: H. Ebert, G. Schütz (Eds.), *Spin–Orbit Influenced Spectroscopies of Magnetic Solids*, Springer, Berlin, 1996, Number 466 in *Lecture Notes in Physics*, 85.
- [31] E.E. Fullerton, D. Stoeffler, K. Ounadjela, B. Heinrich, Z. Celinski, J.A.C. Bland, *Phys. Rev. B* 51 (1995) 6364.
- [32] S.-K. Lee, J.-S. Kim, B. Kim, Y. Cha, W.K. Han, H.G. Min, J. Seo, S.C. Hong, *Phys. Rev. B* 65 (2002) 014423.
- [33] A. Ernst, J. Henk, Electronic and magnetic structures of Pd and Fe films, in preparation.
- [34] J. Henk, T. Scheunemann, S.V. Halilov, R. Feder, *J. Phys.: Condens. Matt.* 8 (1996) 47.
- [35] J. Henk, B. Johansson, *J. Electron Spectrosc. Relat. Phenom.* 94 (1998) 259.
- [36] A.J. Freeman, C.L. Fu, S. Ohnishi, M. Weinert, in: R. Feder (Ed.), *Polarized Electrons in Surface Physics*, *Advanced Series in Surface Science*, World Scientific, Singapore, 1985, Chapter 1, 3.
- [37] H. Huang, J. Hermanson, J.G. Gay, R. Richter, J.R. Smith, *Surf. Sci.* 172 (1986) 363.
- [38] J.C. Fuggle, N. Mårtensson, *J. Electron Spectrosc. Relat. Phenom.* 21 (1980) 275.
- [39] M. Aldén, I.A. Abrikosov, B. Johansson, N.M. Rosengaard, H.L. Skriver, *Phys. Rev. B* 50 (1994) 5131.
- [40] S. Blügel, B. Drittler, R. Zeller, P.H. Dederichs, *Appl. Phys. A* 49 (1989) 547.
- [41] E. Tamura, R. Feder, J. Krewer, R.E. Kirby, E. Kisker, E.L. Garwin, F.K. King, *Sol. State Commun.* 55 (1985) 543.
- [42] D.P. Pappas, K.-P. Kämper, B.P. Miller, H. Hopster, D.E. Fowler, C.R. Brundle, A.C. Luntz, Z.-X. Shen, *Phys. Rev. Lett.* 66 (1991) 504.
- [43] M.P. Gokhale, D.L. Mills, *Phys. Rev. Lett.* 66 (1991) 2251.
- [44] J. Kessler, *Polarized Electrons*, 2nd Edition, Vol. 1 in *Springer Series on Atoms and Plasmas*. Springer, Berlin, 1985.