

Spin-dependent phase shift at ferromagnetic film interfaces from the reflectivity of slow electrons

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We report on spin-dependent phase shift of the electron wave reflected at the film/vacuum and film/substrate interfaces for electrons with energies above the vacuum level. The phase shift is determined within the Fabry-Pérot interferometer model of an absorbing medium, which takes into account the band structure of film and substrate. The results obtained for the model system Fe/W(110) indicate that the phase shift has nonzero values in the entire investigated energy range (up to 20 eV), including the energies of the allowed states in the substrate. This is in contrast to the available models which give zero phase shift for those energy ranges. It is also shown that the spin dependence is much stronger for the Fe/vacuum than for the Fe/W interface.

DOI: [10.1103/PhysRevB.96.205408](https://doi.org/10.1103/PhysRevB.96.205408)**I. INTRODUCTION**

Spin-dependent reflectivity of electrons at surfaces of nanostructures is of great importance in both basic research and application fields. It plays a pivotal role in scattering of electrons at interfaces of ferromagnetic crystals of numerous spintronic devices and nowadays, it becomes an important factor in studies of nonmagnetic interfaces of new topologically nontrivial phases of matter.

In general, the complex amplitude of a reflected electron wave has a form of $r \exp(i\phi)$, where $r(E)$ is the amplitude and $\phi(E)$ is the phase shift. It is accepted that the reflectivity of electrons is spin dependent and that the amplitude $r(E)$ is responsible for the observed spin dependence. However, as the phase shift is energy dependent also its spin dependence should be expected. Surprisingly, there are no reports in which this has been observed. This very fundamental property of the phase shift is demonstrated in the present study.

Here, we propose a method for the determination of the electron phase shift at both interfaces of an ultrathin film in the energy range above the vacuum level. The method is related to the Fabry-Pérot interferometer model of an absorbing medium, which takes into account the band structure of film and substrate [1]. It makes use of the inelastic mean-free path (IMFP) in the film determined from the reflectivity of low-energy electrons [2]. The obtained results reveal that the phase shifts at both interfaces have very small values which cause very small but finite values of the electron reflectivity at both interfaces. The values of the phase shift increase only at the band edges of the film and substrate, revealing a van Hove-type singularity. The phase shift is found to be spin dependent and shows a significant asymmetry between both spin channels.

The studied ultrathin Fe films have a thickness comparable with the electron wavelength and therefore quantum confinement has a strong impact on the reflectivity of electrons revealing quantum size effect (QSE) [3,4]. One of the necessary conditions for the observation of QSE is a finite electron reflectivity at both interfaces of the film, which is associated

with a change of the phase of the electron wave. This phase shift is usually obtained from the analysis of photoemission data using the phase accumulation model. According to this model the total phase shift, when an electron travels through a film meeting two interfaces, has to be an integer multiple of 2π . The phase accumulation model has been successfully used to describe quantum states in metal layers and to obtain the bulk band structure. Its validity has also been tested and confirmed by first-principles calculations [5]. It is described by the Bohr-Sommerfeld rule,

$$2k(E)Nd + \phi_v(E) + \phi_s(E) = n2\pi, \quad (1)$$

where $k(E)$ is the component of the wave vector perpendicular to the film, E is the electron energy, N is a number of monolayers, d is the monolayer thickness, $\phi_v(E)$ and $\phi_s(E)$ are the energy-dependent phase shifts at the film/vacuum and film/substrate interfaces, respectively, and n is an integer. This condition allows the determination of both bound and free electron states, also of spin-dependent states, as well as the total phase shift at both interfaces. However, it does not allow the determination of the phase shift at a particular interface. Therefore, one of the components of the total phase shift, usually the film/vacuum one, is either set to zero [6–9] or obtained from model calculations.

The phase shift at the film/vacuum interface is often modeled using the image potential [10,11] and that at the film/substrate interface by approaches based either on a two-band model [12] or WKB calculations [13]. In the latter two cases the phase shift at the film/substrate interface differs from zero only for energies in a band gap and is set to zero outside of it. This assumption is not necessarily realistic because the electron wave interacts with the potential barrier existing at the interface, which in turn may change its phase upon reflection.

II. MODEL

In order to determine the phase shifts at both film interfaces we apply the method which has already been described and used for the determination of the spin-averaged and spin-resolved IMFP in Fe [2,14]. It is based on the Fabry-Pérot interferometer model of an absorbing medium, which takes

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into account the band structure of film and substrate. The complex reflection coefficient r of light at normal incidence on a thin absorbing film is expressed by the following formula [1,2]:

$$r = \frac{\rho_{12} + \rho_{23} e^{-2v_2\eta} e^{i2u_2\eta}}{1 + \rho_{12}\rho_{23} e^{-2v_2\eta} e^{i2u_2\eta}}, \quad (2)$$

where the indices 1, 2, and 3 denote the vacuum, the absorbing medium, and the substrate, respectively. $\rho_{12(23)}$ is the amplitude of the reflected wave at the film/vacuum (film/substrate) interface, which depends on the real $u_{2(3)}$ and imaginary $v_{2(3)}$ parts of the complex refractive index of the absorbing medium(substrate), respectively; $\eta = 2\pi h/\lambda_1$, where λ_1 is the wavelength in the vacuum, h is the thickness of the absorbing medium. The real parts of the refractive indices of film and substrate are defined by the corresponding wave vectors as $u_2 = k_2/k_1$ and $u_3 = k_3/k_2$, respectively, where $k_{1,2,3} = 2\pi/\lambda_{1,2,3}$. The imaginary parts of the refractive indices of the film and substrate are defined by the corresponding absorption coefficients $\alpha_{2,3} = 4\pi v_{2,3}/\lambda_{2,3}$. Using the model for an electron wave, the IMFP L is defined as the inverse of the absorption coefficient $\alpha = 1/L$.

Having determined the band structures of Fe ($k_2(E)$, Ref. [3]) and W ($k_3(E)$, Ref. [15]) and the IMFP of the film (L_2 , Refs. [2,14]) and substrate (L_3 , Ref. [2]) the real and imaginary parts of the complex refractive index of the Fe film are calculated. (We have chosen the band structure of tungsten given by Willis and Christensen [15], as their first-principles calculations have been confirmed later by other authors who used different exchange and correlation potentials [16–21] and by experiments [22–24].) The phase shift at the vacuum/film ϕ_v and film/substrate ϕ_s interfaces are then determined as the arctan of the ratio of imaginary and real parts of the corresponding complex refractive indices, which after basic algebra operations have the following forms:

$$\phi_v = \arctan \frac{2v_2}{u_2^2 + v_2^2 - 1}, \quad (3a)$$

$$\phi_s = \arctan \frac{2u_3v_2 - 2u_2v_3}{u_2^2 + v_2^2 - u_3^2 - v_3^2}. \quad (3b)$$

The reflectivity of slow spin-polarized electrons (with polarization $P \approx 20\%$) from Fe islands with well-defined thickness and atomically flat surfaces [\mathbf{P} parallel/antiparallel to Fe easy axis ([1 $\bar{1}$ 0])] has been published in Refs. [3] and [4]. It reveals oscillations whose period and amplitude depend on the Fe film thickness and incident electron energy indicating size quantization. From the energy position of the maxima and minima of the QSE oscillations, the band structure of Fe above the vacuum level has been determined [3]. From the decrease of the amplitude of the QSE oscillations with electron energy the spin-averaged [2] and spin-resolved [14] IMFP of electrons in Fe have been determined according to the model presented in Ref. [2].

III. RESULTS AND DISCUSSION

The phase shifts at both film interfaces were determined using the IMFP and the band structure of Fe and W. The spin-averaged results are shown in Fig. 1(a) together with

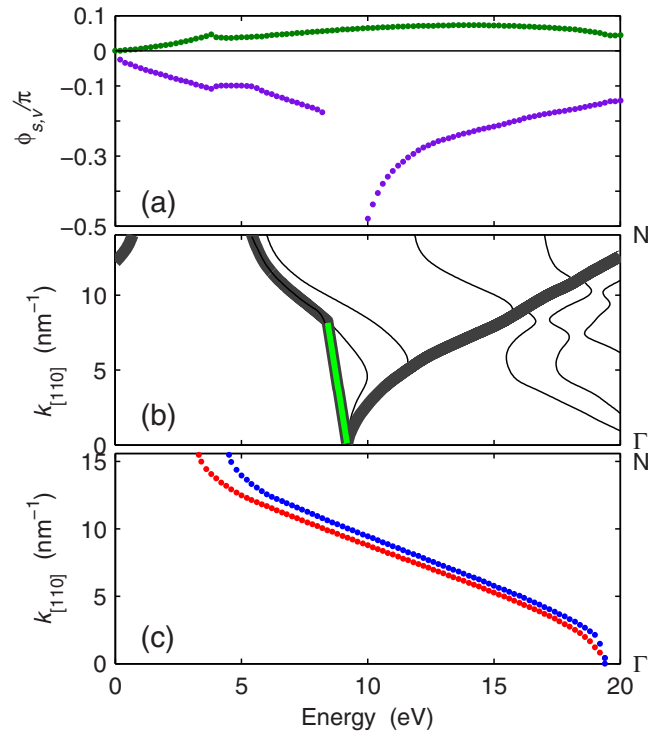


FIG. 1. (a) Spin-averaged phase shift at the film/vacuum (upper curve) and film/substrate (lower curve) interfaces. (b, c) Band structure of W and Fe [3], respectively, along [110]. For W the bands marked by the heavy lines were used. The lower band was interpolated linearly, as indicated by the green line, in order to get the injective function of $k(E)$. Figure 1(b) adapted with permission from Ref. [15]. Copyrighted by the American Physical Society.

the band structures of W (b) and Fe (c) in the corresponding energy ranges. The characteristic feature of the phase shift is that it is much smaller, with values between 0 and 0.1π , at the film/vacuum interface than at the film/substrate interface. The phase shift at the film/vacuum interface reveals a small bump located at about 4 eV and a much weaker one at 19 eV. They are associated with the edges of the Fe s - p band located at the N and Γ points of the Brillouin zone, respectively, Fig. 1(c). In addition, at the film/substrate interface a huge phase change is seen at the Γ point of the band structure of the W substrate near 10 eV, Fig. 1(b). The characteristic changes of the phase reveal a van Hove-type singularity, as already reported in other studies [13,25].

The spin-resolved phase shifts at both interfaces are shown in Fig. 2. It is worthwhile to note the stronger spin dependence of the phase shift at the film/vacuum interface, Fig. 2(a), than at the film/substrate interface, Fig. 2(c). The corresponding asymmetry curves of the spin-dependent phase shifts are shown in Figs. 2(b) and 2(d). The asymmetry of the phase shift is defined as $A = (\phi_{\text{up}} - \phi_{\text{down}})/(\phi_{\text{up}} + \phi_{\text{down}})$, where ϕ_{up} and ϕ_{down} are the phase shifts for the spin-up and spin-down electrons, respectively. The largest asymmetry in the phase shift of about 0.45 is observed for the film/vacuum interface between 4 eV and 5 eV. It is associated with the exchange-split bands which have the edges at the N point around these energies with the exchange splitting of 1.3 eV [3]. A subsequent decrease of the asymmetry is due to the

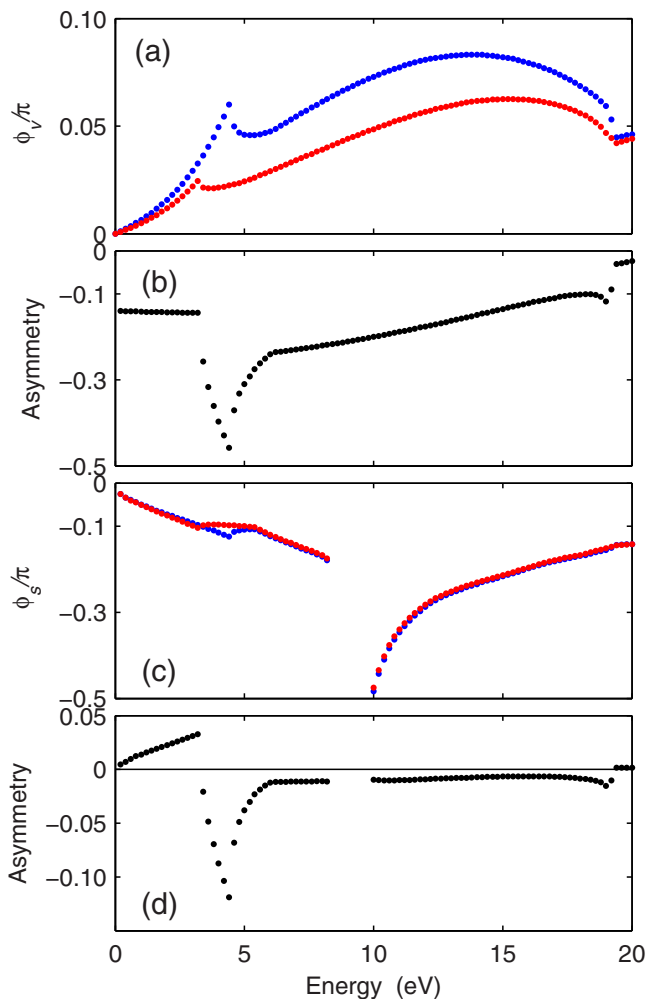


FIG. 2. Spin-resolved phase shift at the Fe film/vacuum (a) and film/substrate (c) interface. Red (blue) color denotes the phase shift for spin-up (spin-down) electrons. Asymmetry (for definition see text) at the Fe film/vacuum (b) and film/substrate (d) interface.

decreasing splitting of the majority and minority bands of Fe with increasing energy. The small increase of the asymmetry at about 19 eV is a fitting artefact due to a limited number of experimental points in the $E(k)$ dependence. In the case of the film/substrate interface, the asymmetry of the phase shift is close to zero except for the energies associated with the edges of the Fe s - p band, similar to that at the film/vacuum interface. In a narrow energy range below 10 eV, no reliable values for the asymmetry can be given because of the uncertainty of the phase shift in this region caused by the interpolation of the W band structure seen in Fig. 1(b).

The most frequently used model of the phase shift at the film/vacuum interface ϕ_v utilizes the image potential and is described by the following formula [10,11]:

$$\phi_v/\pi = [3.4 \text{ eV}/(E_V - E)]^{1/2} - 1, \quad (4)$$

where E_V denotes the vacuum energy. On the other hand, the phase change at the film/substrate interface has been described, within a two-band model, by a purely empirical formula [12]:

$$\phi_s = 2 \arcsin [(E - E_L)/(E_U - E_L)]^{1/2} - \pi, \quad (5)$$

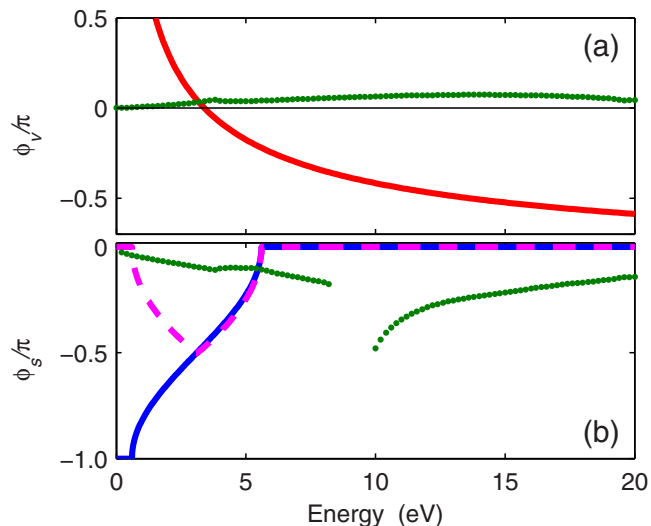


FIG. 3. Phase shifts at the Fe film/vacuum (a) and Fe film/W(110) (b) substrate interfaces. Green, dotted lines denote phase shifts obtained from the spin-averaged IMFP data. Red line in (a) obtained according to Eq. (4), blue and pink curves in (b) according to Eqs. (5) and (6), respectively. In the case of Eq. (6) the proportionality constant is set to 1.

where E_U and E_L are, respectively, the energies of the upper and lower edges of the band gap in a substrate. An alternate description of the phase shift at the film/substrate interface [13] is

$$\phi_s \propto (E - E_0)^{1/2} \theta(E - E_0), \quad (6)$$

where E_0 is the substrate band-edge energy and θ is the unit step function. The phase shifts according to those models for the Fe/W(110) case are plotted in Fig. 3 together with our results. The values of E_U , E_L , and E_0 have been taken from Ref. [15]. In the case of the film/vacuum interface the phase shift obtained according to our approach is much smaller than that obtained on the basis of Eq. (4).

The main idea behind the models describing the film/substrate interface [Eqs. (5) and (6)] is that the phase is constant and equals either 0 or 2π in the energy regions of the allowed bands (electrons from a film couple with the allowed states of a substrate) and it changes smoothly in between. However, this is in disagreement with results of many experiments. A simple test of the validity of models describing phase shifts can be made by comparison of the total phase determined within the phase accumulation model with the one calculated according to Eqs. (4) and (5). [Equations (5) and (6) give the same phase shift (zero) for the allowed bands, see Fig. 3.] As shown in Ref. [26], the sum of the calculated phases described by Eqs. (4) and (5) does not agree with the one determined with the phase accumulation model. The difference is in the range of 100° – 150° . One of the reasons is that Eq. (5) sets the phase to zero outside a band gap, where usually resonance coupling appears between the film and substrate states [26].

According to our approach, the phase shift differs from zero also in the energy regions of the allowed bands, Fig. 1. The observed changes are smooth with the exception of the

band-gap edges where density of states is large [$\nabla_k E(k) \rightarrow 0$], which causes van Hove-type singularities. Such singularities in the phase shifts have been observed also in photoemission studies of quantum-well films [13,25].

Our results show a large difference between the phase shifts at the vacuum/film and film/substrate interfaces and a significant spin dependence at both interfaces. These differences can be understood qualitatively easily by keeping in mind the meaning of these phase shifts. As seen in Eq. (1), they account for the fact that the quantum well does not have infinitely high walls, so that the wave can leak out somewhat from the quantum well, which is accounted for by the phase shifts. At the vacuum/film interface there is a significant potential jump (inner potential); at the film/substrate interface the potential jump is small (contact potential). As a consequence, the wave can leak out very little into the vacuum, causing a small but finite phase shift, while the potential wall at the film/substrate interface is small, so that the wave can penetrate significantly beyond the quantum well, causing a large phase shift. This is particularly pronounced at energies at which the density of states is high in the substrate, which produces the large van Hove singularity at the film/substrate interface. That the van Hove singularities in the Fe film are less pronounced at the vacuum/film interface can be attributed to the suppression of the wave function leaking at the higher potential wall.

As expected from the spin-split band structure [Fig. 1(c)] and shown in Fig. 2, the phase shift at the paramagnetic/ferromagnetic interface is spin dependent. One of the first suggestions that the phase shift at the ferromagnetic/paramagnetic interface can be spin dependent has been given by Egger *et al.* [27]. The authors attributed the observed energy shift between the reflectivity of electrons with the opposite spin polarization, which is due to the spin dependence of the band structure [3], to a spin dependence of the phase shift at the paramagnetic/ferromagnetic interface. Similarly, data presented by Wu and co-workers [6] also exhibit energy shift and intensity difference between reflectivity curves measured for oppositely polarized incident electron beams. The observed shifts in the position of extrema of the reflectivity are due to the exchange-split bands at the Cu/Co interface. The difference between the amplitude of both curves can be easily explained by the difference in IMFP of spin-up and -down electrons, as recently demonstrated for Fe [14]. Thus, these results are not a direct experimental proof of a spin-dependent phase shift.

Calculations of the spin dependence of the interfacial phase shift have been reported by Xu *et al.* [28] for the system Cu/Co(100) and compared with experimental data. The agreement between theory and experiment of the sum of the interfacial reflection phase shifts (vacuum/film + film/substrate) is poor. In particular, the calculated van Hove singularity is absent in the experiment, which was tentatively attributed to the neglect of electron-phonon coupling. Nevertheless, the calculations illustrate the spin dependence of the interfacial phase shift.

Yet two more points should be addressed in the discussion. The first is a possible impact of spin-orbit coupling in the Fe layer on the spin-dependent reflectivity. In experiment, Ref. [4], it is possible to distinguish between asymmetry associated with the exchange interaction and spin-orbit coupling (see, e.g., Ref. [29]). Within the error bar we did not observe a contribution to the asymmetry due to the spin-orbit coupling in the investigated Fe layers. Second, because the crystallographic structure influences electronic structure and therefore also the phase change at the interfaces of the film, it is important to know whether the film is homogenous. In the case of Fe on W(110) it is known that depending on the substrate temperature, one or two iron layers are pseudomorphic [30,31]. However, it is also known that when the third layer grows, the misfit between the film and substrate is relaxed by the appearance of a network of dislocations. As a result the crystallographic and electronic structures become bulklike. The situation is different at the film/vacuum interface. Here, the distance between the topmost layers of Fe is slightly smaller and the magnetic moments of the atoms are larger than in the bulk [32,33]. In general, both factors may slightly modify the band structure of the surface layers of the Fe film and consequently, the phase shift at the film/vacuum interface. Despite possible changes in the magnitude, the results reported here clearly show that the phase shift is spin dependent and that the asymmetry of the phase shift is larger at the film/vacuum than at the film/substrate interface. The observed difference between the phase-shift asymmetries at the two interfaces is apparently due to the coupling of the spin-polarized electrons in the ferromagnetic film with the unpolarized electron states in the tungsten substrate. This leads to the depolarization of the reflected electrons, resulting in a decrease of the polarization.

IV. SUMMARY

Summarizing, we have demonstrated that the phase shift which is gained by an electron wave upon reflections at the ferromagnetic film/vacuum and film/substrate interfaces is spin dependent. The clearest difference between both spin channels is observed at the film/vacuum interface in the energy range where the exchange spin-split bands merge the Brillouin zone boundary. The phase shift is determined within a new approach based on the Fabry-Pérot interferometer model of an absorbing medium, which takes into account the band structure and IMFP of film and substrate. The proposed approach gives results which are different from the phase shifts obtained from presently available models. The main difference is a finite value of the phase shift in the allowed band energy ranges for the film/substrate interface.

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