

Influence of Ni buffer layer on spin-related electronic properties of Co film on W(110) studied by spin-polarized single- and two-electron spectroscopy

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A thin Ni buffer layer between a W(110) substrate and a Co three-monolayer (ML) film was found to improve the crystallinity of the Co film and enhance its magnetic properties. The use of spin-polarized single- and two-electron spectroscopies allowed the identification of energy- and momentum-resolved spin-related modifications of the electronic structure of 3-ML cobalt film due to an Ni buffer layer on W(110) substrate. The presence of a thin Ni layer (1 to 4 MLs) increased the spin asymmetry of the Co density of states in the center of the Brillouin zone just below the Fermi level. Analysis of the spin asymmetry in the $(e,2e)$ spectrum of Co film showed the spin-orbit component of asymmetry was readily observable even with 2 and 3 MLs of Ni buffer layers. The shape and magnitude of the asymmetry are similar to those observed in a 3-ML Co film on W(110) without an Ni buffer. This rules out the earlier suggestion that the spin-orbit interaction in the Co film is induced by W(110) substrate via the proximity effect.

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I. INTRODUCTION

Multilayered structures of ferromagnetic metals attract attention in the field of nanomagnetism because of a peculiar and yet undefined interaction between the constituents of such structures. This interaction determines the overall magnetic anisotropy of the system and is very sensitive to the crystallinity and the thickness of layers. One striking example is the spin-reorientation transition (SRT) discovered in the three-layer Ni(1 ML)/Fe(1 ML)/Ni(8 ML)/W(110) system,¹⁻⁵ where ML means a monolayer. In this system, at room temperature, the easy magnetization axis of the entire system changes from in-plane with one-layer [8-ML Ni/W(110)] to out-of-plane two-layer [at 1-ML Fe/8-ML Ni/W(110)] and back to in-plane three-layer [at 1-ML Ni/1-ML Fe/8-ML Ni/W(110)] with successive deposition of additional MLs of Fe and Ni. The microscopic origin of such a complicated behavior of the multilayered system is not yet completely understood. Therefore, a study of the geometric structure and spin-related electronic properties of a bilayer ferromagnetic system, such as Co/Ni/W(110), is of a particular interest.

Our previous study of Co films on W(110) using spin-polarized two-electron spectroscopy⁶ confirmed the uniaxial anisotropy of the Co film with the easy magnetization axis along [110] direction of the substrate and visualized exchange and spin-orbit interaction (SOI) in the Co film. The spin-orbit interaction was observed in the valence band of the Co film just below the Fermi level. Since the spin-orbit interaction in the ferromagnetic film often is responsible for magnetic anisotropy of the film (magneto-crystalline anisotropy), it was important to try to understand the origin of such spin-orbit interaction and the role of the substrate. A large spin-orbit effect observed in the substrate [high- z W(110) surface] suggested that some proximity effect might be responsible for translation of the spin-orbit interaction from the substrate to the Co film.⁷⁻⁹ One of the objectives of the present investigation was to check this suggestion by introducing a thin spacer (buffer layer) between the substrate and the Co film.

The structural and magnetic properties of a Co film on W(110)¹⁰⁻¹⁴ and of an Ni film on W(110)^{4,15-20} have been studied extensively. It was established that a Co film grows on a W(110) surface above 2 MLs in *hcp* structure with the *c* axis perpendicular to the surface and the easy magnetization axis in the plane of the film along the $[1\bar{1}0]$ direction of the substrate. An Ni film grows in *fcc* structure [above one monolayer (ML)] with the $[111]$ direction perpendicular to the surface. The easy magnetization axis lies in the plane of the film and is parallel to the $[001]$ direction of the substrate.¹⁵

Now, we briefly explain the technique we used for studying this multilayered system.

It is well known that a more sensitive approach to study the spin-related properties of surfaces and thin films is to explore the interaction of spin-polarized electrons with a ferromagnetic surface and to detect the secondary electrons. In the present work, we used two spectroscopies based on such interaction: (i) spin-polarized electron energy loss spectroscopy (SPEELS),²¹ and (ii) spin-polarized two-electron spectroscopy, SP $(e,2e)$.⁸

The first technique is a single-electron spectroscopy in which an energy distribution of secondary (inelastically scattered and ejected) electrons is measured. This distribution is dependent on the mutual orientation of the incident beam polarization and the magnetic moment of the surface. At least two characteristic spin-dependent excitations on a ferromagnetic surface have been studied in this way: Stoner excitations²¹ and spin waves.²²

The second technique is two-electron spectroscopy in reflection for which the geometry is shown in Fig. 1. The additional information comes from the detection of the time correlated electron pairs excited by an incident electron and the measurement of the distribution of such pairs as a function of the momenta of both electrons of the pair. We call such a distribution $(e,2e)$ spectrum. Assuming that energy and momenta-parallel-to-the-surface conservation holds for these scattering events, we determine the distribution of the

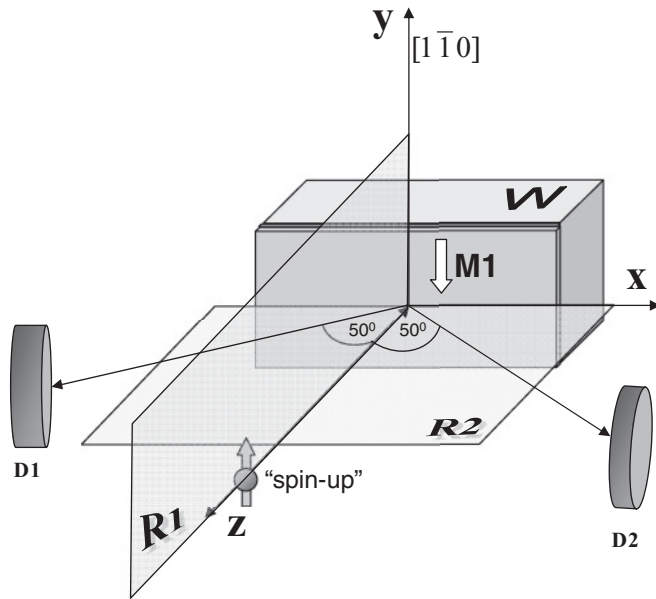


FIG. 1. Geometry of experiment.

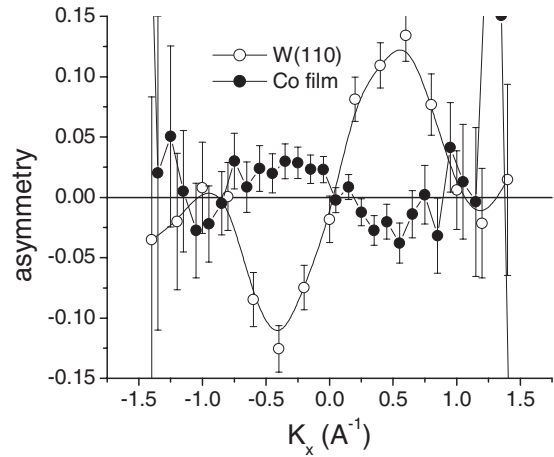
correlated electron pairs as a function of the valence electron wave vector parallel to the surface, i.e. a K_x distribution, at a certain binding energy.^{7,8} Thus, energy and momentum conservations in the $(e,2e)$ reaction allow the valence electron involved in the collision to be localized in energy-momentum space in contrast to a single-electron spectroscopy (EELS), where the result of a scattering event is integrated over unresolved second electron state.

Here, K_x distributions are measured for two orientations of the polarization vector of the incident beam: parallel and antiparallel to the magnetic moment of the ferromagnetic film. Both of these vectors are perpendicular to the scattering plane, which comprises the normal to the surface and two detectors. Two other analogous distributions (for spin-up and spin-down incident beam) are measured for reversed magnetization of the film. Analyses of these four distributions allow the extraction of information about the exchange and spin-orbit interaction in the film and the location of these interactions in energy and momentum space of the valence band of the sample.^{8,9}

Figure 2 shows the spin-orbit asymmetry in the K_x distributions of the W(110) substrate and of the Co film at a binding energy within 0.5 eV just below the Fermi level. The shape of both curves is compatible with the symmetry property of the spin-orbit interaction (SOI). Indeed, mirror reflection in the symmetry plane R1 (Fig. 1) perpendicular to the scattering plane R2 reverses the sign of the incident beam polarization and interchanges the detectors (E1 and E2). Note that the sign of asymmetry of the substrate is opposite to the sign of asymmetry of the Co film.

We intended in this work to test the suggested proximity effect by introducing a buffer layer between the W(110) surface and the Co film. This buffer layer had to preserve the *hcp* structure of the Co film and to reduce the influence of the high- z W substrate on the Co film. A thin Ni layer was chosen to be such a buffer layer.

As mentioned above, we used the spin-polarized electron energy loss spectroscopy²¹ and spin-polarized two-electron

FIG. 2. Spin-orbit asymmetry of K_x distributions of the substrate [W(110)] and 3-ML Co film on W(110). $E_p = 22$ eV.

spectroscopy^{6,7} for the observation of spin-related (magnetic) properties of the Co film. We studied this bilayered system with a fixed thickness of 3-ML Co film and varied the thickness of the Ni buffer layer from 0 to 9 MLs.

II. EXPERIMENTAL DETAILS

Experiments were carried out in vacuum with a base pressure in the low 10^{-11} Torr range. The substrate W(1 $\bar{1}$ 0) crystal was mounted on a rotatable manipulator such that the [1 $\bar{1}$ 0] direction was along the rotational axis and perpendicular to the scattering plane that contains the normal to the sample surface and the detectors (Fig. 1).

The substrate was cleaned in vacuum prior to a film deposition using a standard procedure,^{22,23} including oxygen treatment at 10^{-7} Torr oxygen pressure and 1,400 K sample temperature followed by a few high-temperature flashes up to 2,300 K. The cleanliness of the surface was monitored by Auger electron spectroscopy and low-energy electron diffraction (LEED) as well as by two-electron coincidence spectroscopy, which is much more sensitive to the presence of oxygen.²⁴

The Ni and Co films were deposited using EFM-3 (OMICRON) evaporators with the deposition rate of 0.5 ML per minute. Ni films were deposited in field-free conditions with the Earth's magnetic field compensated by Helmholtz coils down to 10 Oe. Co films were deposited in the same conditions as in our previous work,^{8,9} namely a weak magnetic field (about 600 Oe), generated by a coil around the vacuum chamber, was applied parallel or antiparallel to the [1 $\bar{1}$ 0] direction along the surface of the tungsten substrate. The direction of magnetization was defined by the direction of applied magnetic field. This weak magnetic field is sufficient to magnetize a very thin Co film at an initial stage of growth because the Curie temperature of such a film apparently is very low, near room temperature or even lower.¹⁴ The thickness of the Co film was always 3 MLs, whereas the thickness of the Ni layer varied from 0 to 9 MLs. The quality of the prepared films was monitored by low-energy electron diffraction and Auger electron spectroscopy.

The experimental setup of the $(e,2e)$ spectrometer in back-reflection mode is described elsewhere.^{25,26} In brief, the incident electron impinges onto the sample surface, and two outgoing (back) electrons are detected in coincidence, and their momenta are measured using a time-of-flight (TOF) technique.²⁵ This TOF technique requires a reference point on the timescale which is provided by pulsing the incident beam with a pulse width of less than 1 ns and repetition rate of 4 MHz. The coincidence electronics allows the detection of two time-correlated electrons within a time window of 200 ns. To increase the relative number of correlated electron pairs resulting from an interaction of a single incident electron with the sample (compared to accidental pairs resulting from two incident electrons), a very low incident current ($\sim 10^{-14}$ A) was used providing less than one electron per pulse on average. For the electron detection, we used two position-sensitive detectors based on 75-mm diameter microchannel plates (MCP). Position sensitivity allows the measurement of the electron angular distribution and the correction of the electron flight distance for the time-of-flight energy measurement.²⁵ A spin-polarized electron source is based on photoemission from a strained GaAs photocathode. Photoelectrons excited by circularly polarized light with the wavelength 836 nm are initially longitudinally polarized, and they pass through a 90° deflector such that the emergent beam is transversely polarized. The degree of polarization P of the electron beam is measured in a separate experiment and is estimated to be $(66 \pm 2)\%$. Beam polarization can be reversed by changing the sense of circular polarization of the laser light incident on the GaAs photocathode. The polarization vector of the incident beam is chosen to be perpendicular to the scattering plane that comprises the normal to the sample surface and the two detectors.

The measured $(e,2e)$ spectrum is a six-dimensional array, where each detected correlated electron pair is represented by arrival times T_1 and T_2 and coordinates on the detectors X_1, Y_1 and X_2, Y_2 and can be projected on various two-dimensional or three-dimensional distributions. For example, it can be presented in the form of a number of pairs as a function of energy E_1 and E_2 of each electron of the pair.

With a spin-polarized incident beam, two $(e,2e)$ spectra were measured: one for the beam polarization spin-up (see Fig. 1) and another for the beam polarization spin-down (opposite to spin-up). Then spin effects are analyzed by comparing these two spectra.

When the coincidence conditions between the two arms of the $(e,2e)$ spectrometer are switched off, each of these arms can be used as a SPEELS spectrometer for recording electron energy loss spectra for normal incidence and two complementary detection angles.

III. RESULTS AND DISCUSSION

A. Spin-polarized electron energy loss spectroscopy

Prior to the $(e,2e)$ measurements, we performed SPEELS measurements on the deposited ferromagnetic layers. Electron energy loss spectra were recorded for spin-up polarization (that we define as polarization vector being parallel to the Y axis) of the primary electron beam and denoted as I^+ and

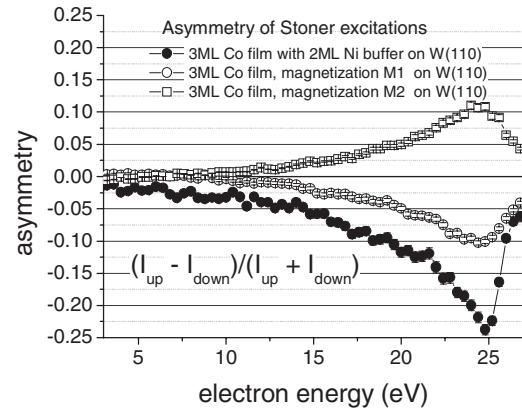


FIG. 3. Stoner excitation asymmetry with (closed circles) and without (open circles) Ni buffer layer. $E_p = 27$ eV, normal incidence, 50° detection angle.

for spin-down polarization of the incident beam (opposite to spin-up) denoted as I^- . The spin-dependent features of a SPEEL spectrum are visualized by the asymmetry A defined as $A = (I^+ - I^-)/(I^+ + I^-)$.

For a ferromagnetic sample, the main spin-dependent feature of a SPEEL spectrum is Stoner excitations asymmetry,²⁷ which we used as an indication of the magnetization of the film along the quantization axis, which is chosen to be the Y axis of the coordinate system (see Fig. 1). An example of the Stoner excitation (SE) asymmetries for two opposite magnetizations $M1$ and $M2$ of the Co film on W(110) is shown in Fig. 3 by open circles and squares, respectively. The sign of the asymmetry changes when the sign of magnetization changes, and this clearly indicates the magnetic origin of the asymmetry.

Ni layers in the thickness range from 1 to 9 MLs deposited on the W(110) surface show no SE asymmetry, i.e. no magnetization along the chosen quantization axis (Y axis). This is not a surprise because, as mentioned above, the Ni film on a W(110) substrate has its easy magnetization axis in the plane of the film and parallel to the $[001]$ direction of the substrate, i.e. perpendicular to the polarization of the incident beam. We note here also that, because of the thickness dependence of the Curie temperature of the Ni film, it is paramagnetic at room temperature up to a thickness of 3–4 MLs.

When a Co film is deposited on a 2-ML Ni buffer layer, it shows a Stoner excitation asymmetry that is then about two times larger than in the case of no buffer layer (Fig. 3). This indicates the change of the Stoner density of states (DOS). Indeed, the Stoner DOS is a sort of joint density of states with the condition that occupied and unoccupied states have opposite spins, and they are separated by a definite momentum transfer.^{27–30} The sign and the magnitude of the asymmetry of SE are determined by the combination of majority and minority density of states and their energy distribution. We note here that majority and minority density of states are defined as density of electronic states with spin quantum number (spin projection on quantization axis Y) $-1/2$ and $+1/2$, respectively. The presence of the Ni buffer layer changes this combination and increases the asymmetry of SE. The reason might be an improved crystallinity of the Co film and, as a consequence, transformation of the multidomain Co film with

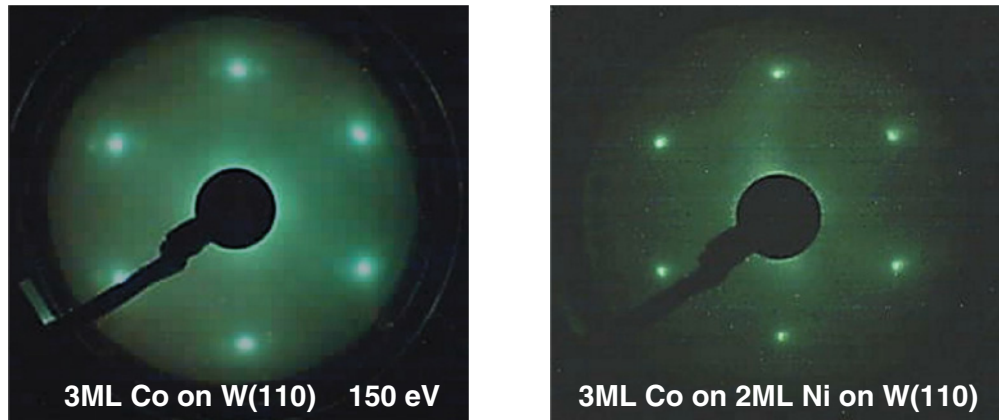


FIG. 4. (Color online) LEED patterns from 3-ML Co film on W(110) without and with Ni buffer layer.

slightly tilted magnetic moments with respect to each other into a single-domain film. This suggestion is supported by Fig. 4: it seems that an Ni buffer layer serves as a good template for the cobalt film. Comparison of the LEED patterns of the 3-ML Co film on clean W(110) and on Ni buffer layer shows that Ni template improves the crystallinity of the layer that shows up in much sharper LEED patterns (see Fig. 4).

On the microscopic level, the change of the Stoner DOS might be caused by the hybridization of the electronic states of Ni and Co.

B. Spin-polarized two-electron spectroscopy

Two-electron spectroscopy relies on the fact that the incident electron undergoes a single-step individual electron-electron scattering with the valence electron of the target. The energy conservation law in the $(e,2e)$ reaction implies that $E_b = (E_1 + E_2) - E_o$, where E_b is the binding energy of the valence electron, E_o the primary electron energy, and E_1 and E_2 the energies of two detected electrons.

This energy conservation only defines the binding energy of the valence electron if the detected electrons result from

a single-step electron-electron collision. Earlier experimental results²⁶ have demonstrated that the dominant contribution of the single-step process can be ensured for correlated pairs excited from a metal crystal within about 4 eV below the Fermi level. This conclusion has been made on the basis of the measurements at normal and off-normal incidence. It was shown that the correlated electron pairs, excited from within this binding energy range, remember the tangential component of the incident electron at off-normal incidence, i.e. they obey the momentum conservation law for the component parallel to the surface, i.e. $K_{b\parallel} = K_{1\parallel} + K_{2\parallel} - K_{0\parallel}$, where indices $b, 1, 2$, and 0 denote parallel to the surface components of the valence electron, first and second detected electrons, and incident electron, respectively. This is only possible if the incident electron undergoes single-step electron-electron collision (i.e. a collision between two electrons) and no additional electron-electron or electron-phonon collisions occur.

Another argument in favor of the individual electron-electron collision in the $(e,2e)$ experiment on metals is the observation of the exchange effects in the scattering of spin-polarized electrons on a ferromagnetic surface.⁶ Figure 5 shows the spin-resolved binding energy spectra of 3-ML Co

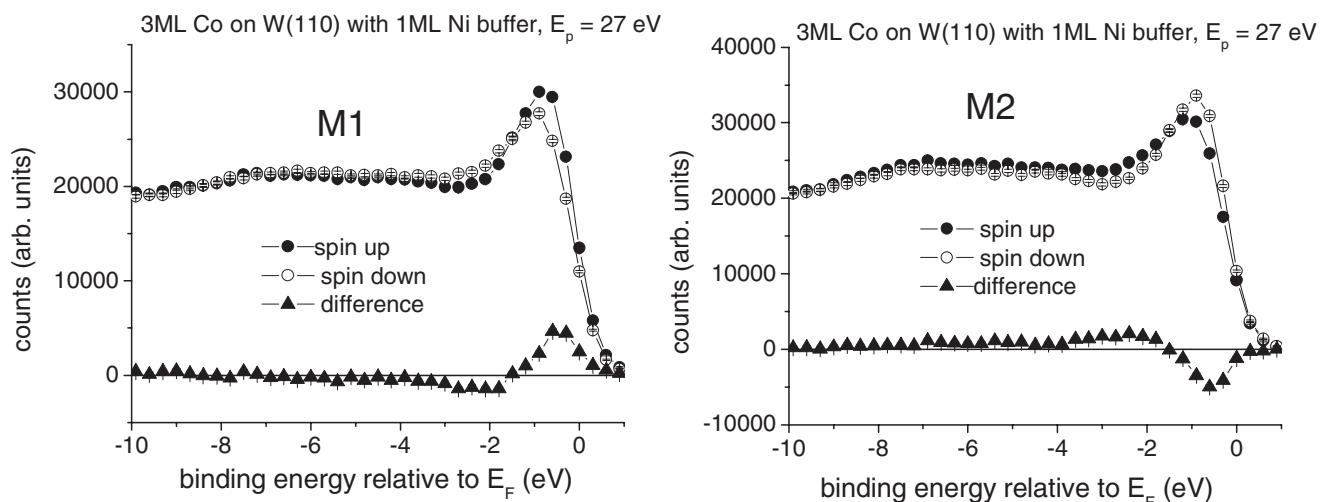


FIG. 5. Spin-resolved binding energy spectra for two magnetizations of 3-ML Co film on W(110) with 1-ML Ni buffer layer.

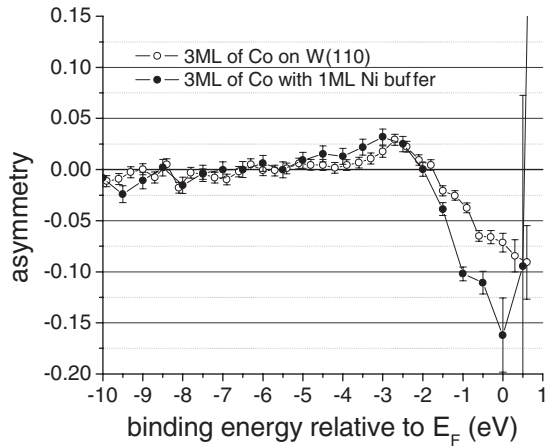


FIG. 6. Spin asymmetry of the spectral density function of Co film with and without Ni buffer layer at $k_x = 0$.

film on W(110) with 1-ML Ni buffer layer for two opposite magnetizations of the sample. One can see that the difference between spin-up and spin-down spectra changes sign when the magnetization of the film changes sign. This indicates that the origin of the spin-related effect is the exchange interaction, and it occurs at the binding energy where the imbalance between spin-up and spin-down DOS exists.

Spin effects in the $(e,2e)$ scattering on a ferromagnetic sample have been analyzed by Samarin *et al.*,⁶ Morozov *et al.*,⁷ and Berakdar.³¹ One of the outcomes of this analysis concerns the particular kinematical arrangement of normal incidence and detection of two electrons with equal energies and at equal angles. Then the asymmetry of the binding energy spectrum represents the asymmetry of the spectral density function of valence electrons in the center of the Brillouin zone: $S(E, K_x = 0) = (D^+ - D^-)/(D^+ + D^-)$, where D^+ and D^- are density of states for majority and minority electrons at an energy E and a wave vector $K_x = 0$. This result provides an access to the spin-dependent electronic structure through the measured $(e,2e)$ spectra. Indeed, if we measure the $(e,2e)$ spectra for spin-up and spin-down polarization of the incident beam, we can select from the measured set of data the electron pairs which meet these indicated requirements. The measured spin asymmetry A of the binding energy spectrum for these pairs would represent spin asymmetry of the Bloch spectral density function (SDF) in the center of the Brillouin zone $S(E, k_{\parallel} = 0) = -(1/P) A$, where P is the degree of polarization of the incident beam. We measured SDF of Co film on W(110) with and without Ni buffer layer. Figure 6 shows spin asymmetry of the SDF of 3-ML Co film for two cases: (i) 3-ML Co film on W(110) surface and (ii) 3-ML Co film on 1-ML Ni buffer layer. It is clearly seen that Ni buffer layer substantially enhances asymmetry of SDF just below the Fermi level.

We measured the asymmetry of the spectral density function of the Co film on W(110) with a Ni buffer layer as a function of the thickness of the buffer layer (Fig. 7). One can see that few atomic layers of Ni increases the absolute value of asymmetry of $S(E, k_{\parallel} = 0)$, but starting from the thickness of 2 MLs of Ni, the absolute value of asymmetry decreases and reaches zero at about 6 MLs.

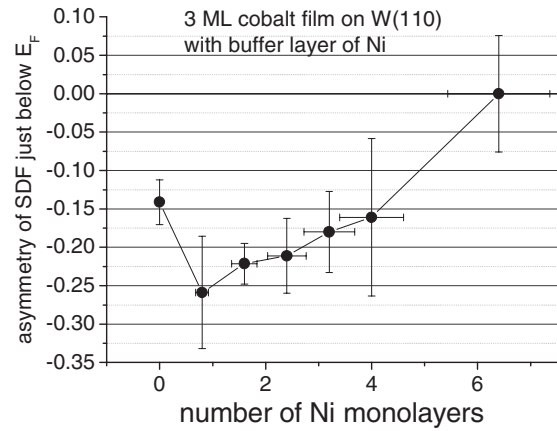


FIG. 7. Asymmetry of spectral density function at $k_x = 0$ just below the Fermi level as a function of the thicknesses of Ni buffer layer.

The nonvanishing value of the asymmetry $S(E, k_{\parallel} = 0)$ indicates an imbalance of spin-up and spin-down density of states in the film. A thin (1- to 4-ML) Ni buffer layer increases the relative minority density of states compared to the Co/W(110) system, as shown by the asymmetry of SDF. On the other hand, the thick-enough layer of Ni (about 5–6 MLs), which is already in a ferromagnetic state at room temperature and magnetized in-plane along [100] direction of the substrate (X axis of the coordinate system), drives the Co layer to be magnetized along the same direction and hence to be seen as nonmagnetic by the incident electrons polarized along the Y direction. That is why on Fig. 6 the asymmetry of SDF is zero at 6 MLs of Ni buffer layer. The transition of the SDF asymmetry from its maximum absolute value at 1 ML of Ni buffer layer to zero at about 6 MLs most likely reflects the transition of the Ni layer from a paramagnetic state to a ferromagnetic state due to the thickness dependence of the Curie temperature of the thin Ni layer. Indeed, for very thin Ni films (in the range of 1–4 MLs), the Curie temperature is close to the room temperature (or even below it).^{16–20} It means, in this thickness range, the Ni buffer layer does not have a preferred magnetization direction and is magnetized by the applied magnetic field along the Y axis. The Co film grows magnetized in the direction of the applied magnetic field. After 3-ML Co film is grown, the external field is removed, and all measurements are done at remanent magnetization.

To test the influence of the W(110) substrate on the spin-orbit interaction in the Co film, we used spin-polarized $(e,2e)$ spectroscopy and measured spectra for two opposite magnetizations $M1$ and $M2$ (both are perpendicular to the scattering plane) of the bilayered system with various thicknesses of the Ni buffer layer.

The exchange and spin-orbit interactions are visualized as in Ref. 9 by so-called K_x distributions of correlated pairs. These distributions represent the number of pairs as a function of the K_x component of the valence electron wave vector. The binding energy of the valence electron is selected to be within 1 eV below the Fermi level. If the asymmetries of K_x distributions for nonreversed and reversed sample magnetization are denoted as A^+ and A^- , the spin-orbit (A_{SO})

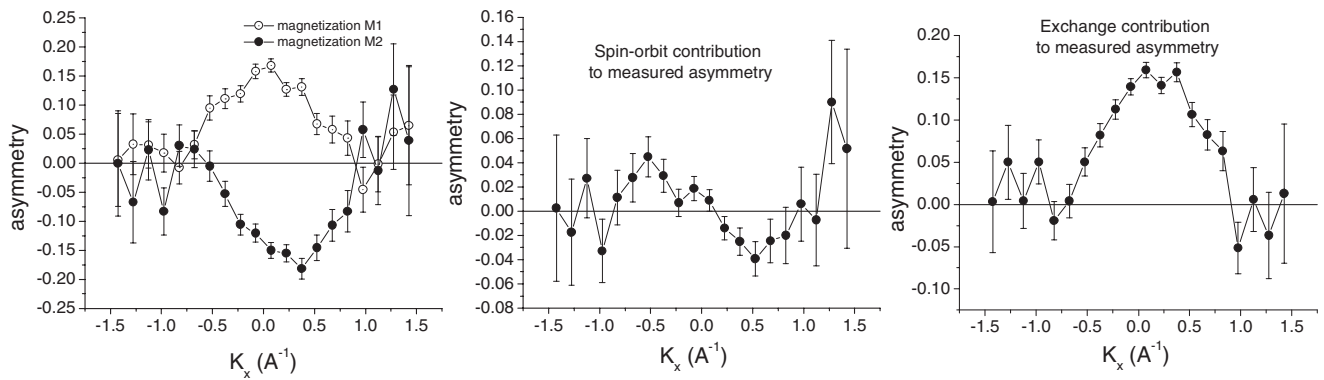
3 ML of Co film on 2 ML Ni buffer, $E_p = 27$ eV

FIG. 8. Spin-orbit and exchange components of the asymmetry of K_x distributions of 3-ML Co film on W(110) with 2-ML Ni buffer layer; spectra are integrated over 1 eV binding energy just below the Fermi level.

and exchange (A_{ex}) contributions are given to leading order by:²⁷

$$A_{ex} = 1/2(A^+ - A^-)$$

$$A_{SO} = 1/2(A^+ + A^-).$$

One can see from Fig. 8 that 3-ML Co film on 2-ML Ni buffer layer exhibits a substantial spin-orbit component in the K_x distribution asymmetry for binding energy within 1 eV below the Fermi level. The shape and magnitude of the asymmetry are very similar to those observed in a Co film on W(110) substrate without a buffer layer.⁸ This finding seems to rule out our earlier suggestion about a proximity effect between the W substrate and the Co film that induces the spin-orbit interaction in the Co film. On the other hand, it may happen that a very thin Ni layer still translates the spin-orbit interaction into the Co film from the W substrate.

We would like to note here that the probing depth of the ($e,2e$) technique is equal to the effective ($e,2e$) decay length λ that is determined by inelastic mean free paths λ_1 , λ_2 , and λ_3 of the incident and two outgoing electrons: $\lambda^{-1} = (\lambda_1)^{-1} + (\lambda_2)^{-1} + (\lambda_3)^{-1}$. Using realistic values of mean free paths of electrons for primary energy of about 20 eV and secondary electrons of 6.5 eV, the value of λ was estimated to be 2.55 Å.³² It means that all experimental results presented here are assigned to the Co layer, and the contribution from the substrate is negligible.

IV. CONCLUSIONS

We investigated the influence of an Ni buffer layer on the spin-related electronic properties of 3-ML Co film on W(110) substrate. A thin (1- to 4-ML) Ni layer on W(110) is a very good template for growing a Co film giving sharp LEED patterns of the *hcp* crystal structure of the film.

Although the Ni buffer layer in the thickness range of 1 to 4 MLs does not seem to show a ferromagnetic order, it exhibits a substantial influence on the spin-dependent electronic structure of the 3-ML Co layer. The observed increase of the spin asymmetry of the cobalt DOS in the presence of the thin buffer layer of Ni might be due to the improved crystallinity of the Co layer. Indeed the improved crystallinity would reduce the formation of domains in the film and disorientation of them with respect to each other. From a microscopic viewpoint, this would mean the hybridization of the *d* states of the Ni and Co and rearrangement of spin-up and spin-down states of the Co layer in favor of minority DOS just below the Fermi level.

Analysis of the spin asymmetry in the ($e,2e$) spectrum of the Co film on W(110) with an Ni buffer layer shows the spin-orbit component of asymmetry is readily observable even with 2 and 3 MLs of Ni buffer layers. The shape and magnitude of the asymmetry are similar to those observed in a 3-ML Co film on W(110) without an Ni buffer. This seems to rule out the earlier suggestion that the W(110) substrate induces this spin-orbit interaction in the Co film via the proximity effect. Other possible mechanisms must be considered to explain the observed spin-orbit component of the asymmetry of ($e,2e$) spectra and magnetic anisotropy of the Co film on W(110), such as an epitaxial misfit strain, magnetoelastic coupling, and magnetostriction.

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