

Photoemission from Stepped W(110): Initial or Final State Effect?

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The electronic structure of the (110)-oriented terraces of stepped W(331) and W(551) is compared to the one of flat W(110) using angle-resolved photoemission. We identify a surface-localized state which develops perpendicular to the steps into a repeated band structure with the periodicity of the step superlattices. It is shown that a final-state diffraction process rather than an initial-state superlattice effect is the origin of the observed behavior and why it does not affect the entire band structure.

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Single-crystal surfaces that bear, due to a controlled surface miscut, a regular superlattice of steps are envisaged as adequate templates for the growth of a large number of identical nanostructures like quantum wires and quantum stripes [1]. In this context, vicinal W(110) has developed into an important stepped substrate and controlled growth of nanostructures like stripes formed of Fe has led to novel magnetic anisotropy and coupling phenomena like perpendicular magnetization and dipolar antiferromagnetism [2,3]. These new accomplishments in nanomagnetism occur on a relatively large length scale (e.g., 8 nm) and make use of magnetic dipole interactions. They have not, so far, utilized size quantization of electronic states as were found essential for the magnetic interlayer coupling in two-dimensional films [4]. Whether or not such effects will in the future be used depends on the ability of the stepped substrate to support quantization effects in the electronic structure. The method of choice to test this is angle-resolved photoemission since it provides the electronic structure including band dispersions [5]. The stepped surfaces most extensively studied to date are the (111) vicinals of fcc Cu and Au where surface state electrons in a narrow bulk band gap at the L point are probed [6–11]. Different types of behavior have been obtained: the wave functions were either spread out over the macrosurface [6,10,11] or localized at the microsurface [9], and a switch over from two-dimensional to one-dimensional behavior was observed at a step width of ~ 17 Å [8]. Most recently, we have analyzed data from carbon nanostripes on stepped Ni(771) and observed intense final-state effects due to so-called umklapp scattering at the superlattice of steps and stripes [12] and similar effects occur also in the data from vicinal Au(111) [13]. In the umklapp picture, the photoelectron can undergo scattering at the surface lattice when leaving the crystal and acquire an extra reciprocal surface lattice vector \mathbf{G}_{\parallel} according to $\mathbf{k}_{\parallel,\text{vac}} = \mathbf{k}_{\parallel,\text{crystal}} + \mathbf{G}$ which leads to observable effects especially when the surface periodicity differs from the one of the bulk [14].

The aim of the present work is to clarify the presence of one- or two-dimensional behavior and initial- or final-state effects on stepped W(110). W is in principle well suited since theory predicts a substantial portion of the W density of states surface-localized [15] although the identification of surface states in an experiment is often complicated and has on W been achieved only for a small number of cases. While experimental data are still outstanding, theory went ahead predicting, e. g., that W surface states will be responsible for the observed oscillatory stability conditions of W steps [16].

In this Letter, we will demonstrate strong superlattice effects fulfilling the periodicity of the step superlattice. By angle- and energy-dependent measurements in photoemission and in low-energy electron diffraction (LEED), we show that the observed behavior is due to a final-state diffraction effect at the step superlattice instead of a superlattice bandstructure effect on the initial states. We have chosen the (331) and (551) surfaces characterized by terrace widths of three and five W lattice constants, respectively, or 0.948 and 1.58 nm. Figure 1 shows the surface atom arrangement for these surfaces and a comparison of the surface Brillouin zones of flat and stepped W(110). The miscut angle amounts to 13° for W(331) and 8° for W(551) along the [001] direction leading to step edges oriented along $[1\bar{1}0]$. Our samples have been prepared as usual [17] until LEED showed sharp 6×1 and 10×1 superstructures for W(331) and W(551), respectively. Photoelectrons were excited with linearly polarized synchrotron light at the U125/1 and UE56/1 PGM beamlines [18] at BESSY. A spherical electron analyzer has been used at 1° angle resolution and 50–150 meV combined (photon and electron) energy resolution in a vacuum of 2×10^{-10} mbar.

Figure 2 displays angle-resolved photoemission spectra of W(331) for \mathbf{k}_{\parallel} vectors parallel (a) and perpendicular (b) to the step direction at 62.5 eV photon energy for different polar angles. (We relate all polar angles θ to the [110] microsurface normal.) At the bottom, Fig. 2 shows for the sake of comparison normal-emission spectra from

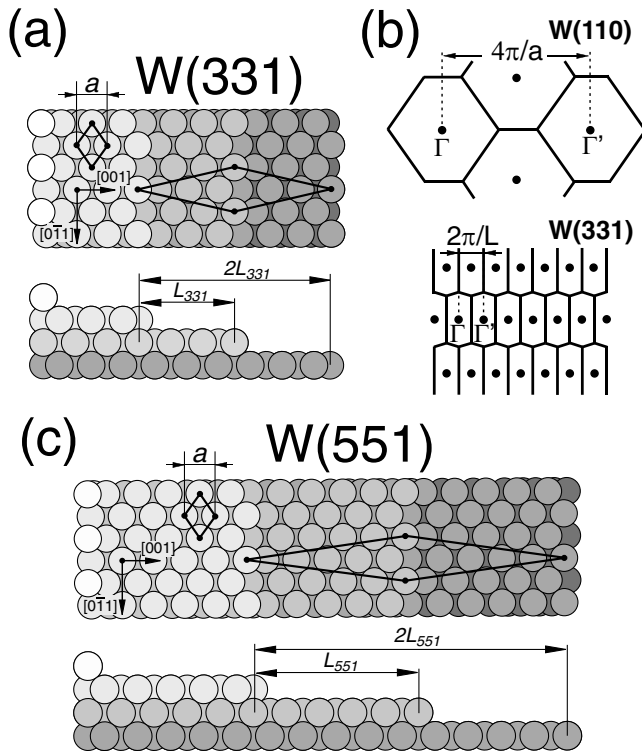


FIG. 1. Geometry of W(331) (a) and relation of its surface Brillouin zone to the one of flat W(110) (b). The steps run perpendicular to the [001] or Γ -H direction and are by a factor of 5/3 wider on W(551) (c).

flat W(110) measured in such a way as to ensure the same measurement conditions as for the vicinals [i. e., the \mathbf{E} -vector of the light points along $[0\bar{1}1]$ in Fig. 2(a) and along [001] in Fig. 2(b)]. Prominent features identified earlier on W(110) are an intense surface-resonance (SR) at 1.2 eV for $\bar{\Gamma}$ [17,19] and bulk emission from the top of the Σ_1 band which is sampled near the N_1 point of the bulk Brillouin zone when excited around 63 eV photon energy. The comparable spectrum in the emission-angle series from W(331) in Fig. 2, i. e., the one for emission along the microsurface normal [110], is colored red. For \mathbf{k}_{\parallel} parallel to the steps [Fig. 2(a)], peak S is seen to disperse symmetrically about the red spectrum. Surface-resonance emission at 1.2 eV can in principle be assigned by comparison to W(110) [19], but the small intensity on the stepped surface suggests that bulk emission may actually dominate the peak.

The [110]-normal-emission spectra display strong differences: For W(331), the Σ_1 band does not appear at 6 eV as for W(110) but at 5.2 eV where states are *forbidden in bulk W* due to the large gap (6.2 to 3.3 eV) for emission along [110] [17,19].

The top panel of Fig. 4 indicates the presence of a surface-resonance which has not yet been reported inside or near this gap: The dispersion measured at 62.5 eV gives only an indication for this since it comprises degenerate

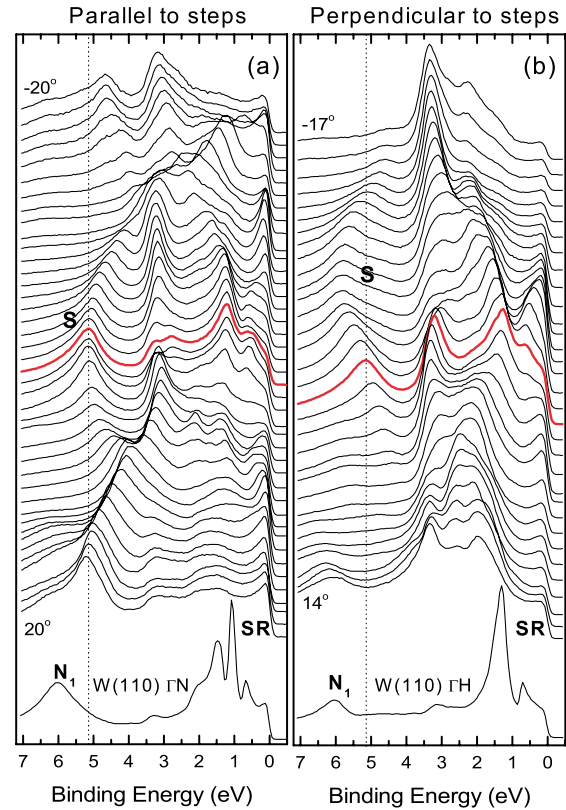


FIG. 2 (color online). Angle-dependent photoemission spectra of W(331) parallel (a) and perpendicular (b) to the step direction for 62.5 eV photon energy. The bottom of the band labeled S appears 6° away from the microsurface normal.

bulk and surface bands. At 110 eV photon energy, however, the Γ point is reached and the bulk-derived emission corresponds to the band bottom at 9 eV. No other emission from W bulk states is expected in normal-emission in the whole energy range from 9 eV up to the border of the band gap at 3.3 eV. Nevertheless, a second dispersion has remained at 5.8 eV and is identified with surface emission. Its dispersion with \mathbf{k}_{\parallel} follows closely the shape of the bulk band gap according to the surface projection of the bulk bands (solid line) calculated in Ref. [20].

Returning to Fig. 2, we note that in the direction perpendicular to the steps, the 5.2 eV peak disperses downwards and assumes a minimum energy of 6 eV up the stairs at about 6° off normal, i. e., at this angle the same energy is reached as for normal-emission on flat W(110). Figure 3 allows for a better judgement of this behavior: Figs. 3(a)–3(c) are a color-scale representation of photoemission spectra at 50, 62.5, and 125 eV photon energy for different polar emission angles perpendicular to the steps. When changing the photon energy, we move in terms of the W bulk band structure along the [110]- or Σ -direction away from the N point (~ 63 eV) towards Γ [19]. At 62.5 eV photon energy, we observe one branch S with binding energy between 4 and 6 eV, shifted away from the microsurface normal. At 50 eV, we observe addi-

tionally a second branch at positive emission angles, S^* . At 125 eV, both branches S and S^* are equally pronounced and shifted approximately symmetrically with respect to the microsurface normal.

Figure 4 allows us to determine the shift between the two dispersions as 0.66 \AA^{-1} . For W(551) we observe even more than two branches with a period of 0.40 \AA^{-1} . These values correspond exactly to the periods of the superlattice Brillouin zones of the two surfaces or, following a simple reasoning, to the distance from $\bar{\Gamma}$ to $\bar{\Gamma}'$ along [001] for W (4.0 \AA^{-1}) divided by the six-fold and ten-fold surface periodicities measured by LEED. In the Brillouin zone picture, the electron is assumed to hop across the steps and form Bloch waves along the macrosurface. This is the simplest interpretation in which the repetition of the band dispersion is due to band periodicity in the repeated zone scheme of the step superlattice. Such behavior would very well be compatible with our identification of the feature S as surface-resonance. This requires the wave function to be oriented parallel to the macrosurface [6,8], which means that in order to be an initial-state effect, the repeated $E(\mathbf{k}_{\parallel})$ dispersions for various photon energies must coincide when \mathbf{k}_{\parallel} is related to the macrosurface (i. e., $\mathbf{k}_{\parallel} = 0$ corresponds to the macrosurface normal) and deviate when \mathbf{k}_{\parallel} is related to the microsurface ($\mathbf{k}_{\parallel} = 0$ at the microsurface normal). Because of the large angles between macro- and microsurface of 13° for W(331) and 8° for W(551) the present experiment can unambiguously distinguish between the two cases. Both cases can be compared in a single figure [see the W(331) data in Fig. 4(b)]: Good agreement is reached for \mathbf{k}_{\parallel} related to the microsurface. That a plot relative to the macrosurface fails can be seen from the macrosurface normals for photon energies of 50, 62.5, and 105 eV which were inserted into Fig. 4 and are clearly separated. The one for 125 eV would appear outside of the figure frame. This shows that in its initial-state the electron occupying S remains localized at the one-dimensional microsurface and that the repetition of band S is not due to an initial-state effect.

In the LEED picture, the electron undergoes scattering in the final-state of the photoemission process. As this concept gives the same periodicity as the initial-state concept, we seek for further support for a final-state interpretation. This is found in the intensities of the two bands which vary strongly with photon energy so that at 62.5 eV the S^* branch even vanishes (Fig. 2). We have recorded LEED patterns *in situ* for comparable primary electron energies and display them in Fig. 3(d)–3(f) *vis-à-vis* the measured dispersions. The LEED spots (dispersions) are split for 44 eV (50 eV) and 125 eV (125 eV), and for 62.5 eV (62.5 eV) almost all of the intensity is concentrated in a single spot (band).

We want to discuss which model can appropriately describe this effect. The present experiment allows us to

relate the measured dispersions to the direction of the [110] microsurface normal given by the bulk-derived Σ_1 band in Fig. 3(c) and in Fig. 4. This is also very clearly seen in the original data of Fig. 2(b) where the peak at $\sim 1.5 \text{ eV}$ disperses around the microsurface normal. On the basis of a description by umklapp scattering at the step superlattice we expect a process according to $\mathbf{k}_{\parallel, \text{vac}} = \mathbf{k}_{\parallel, \text{crystal}} + \mathbf{G}$ with $\mathbf{G}_{331} = 0.66 \text{ \AA}^{-1}$ for W(331) and $\mathbf{G}_{551} = 0.40 \text{ \AA}^{-1}$ for W(551). Instead, if we identify the dispersion of band S on flat W(110) with $\mathbf{k}_{\parallel, \text{crystal}}$ and compare it with the dispersions on W(331) and W(551), we observe $\mathbf{k}_{\parallel, \text{vac}} = \mathbf{k}_{\parallel, \text{crystal}} \pm \mathbf{G}/2$ with the dispersion for $\mathbf{k}_{\parallel} = 0$ completely missing.

More appropriate is the description given for LEED from stepped surfaces [21]. Figure 3(b) of Ref. [21] shows how a single LEED spots splits up into two when the \mathbf{k} vector moves from the in-phase to the antiphase condition: The reciprocal lattice rods for scattering from a stepped surface are perpendicular to the macrosurface and therefore tilted with respect to the microsurface. Because they pass through the Γ point of W , scattering conditions remain unchanged there, but they separate at the N point (antiphase). This explains why in our experiment the dispersion S , which is located at the N point, appears split and the band bottom at 9 eV, which is located at the Γ point, remains single. As the photoemission transition determines the primary beam for electron scattering, it restricts the dispersions which appear repeated to those at the N point. This applies to bulk and surface bands but surface bands dominate the emission from the N point in the present case. We believe that the location at the N point in momentum space is the main cause of the

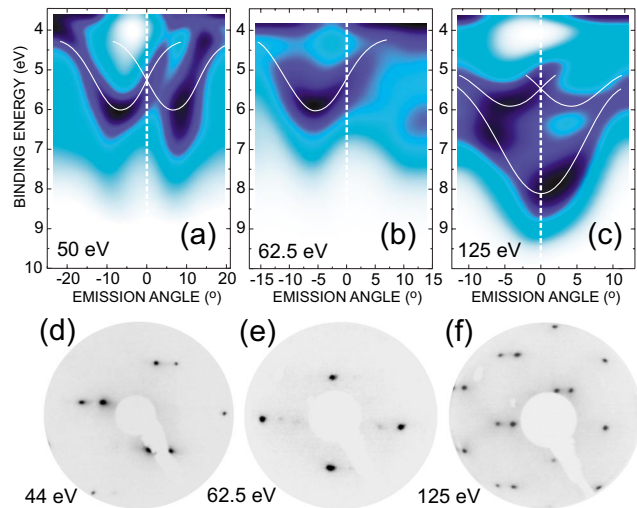


FIG. 3 (color online). Extraction of dispersions from photoemission spectra for W(331) (a–c) together with LEED patterns of similar intensity behavior (d–f). The emission angle is given with respect to the [110] microsurface normal.

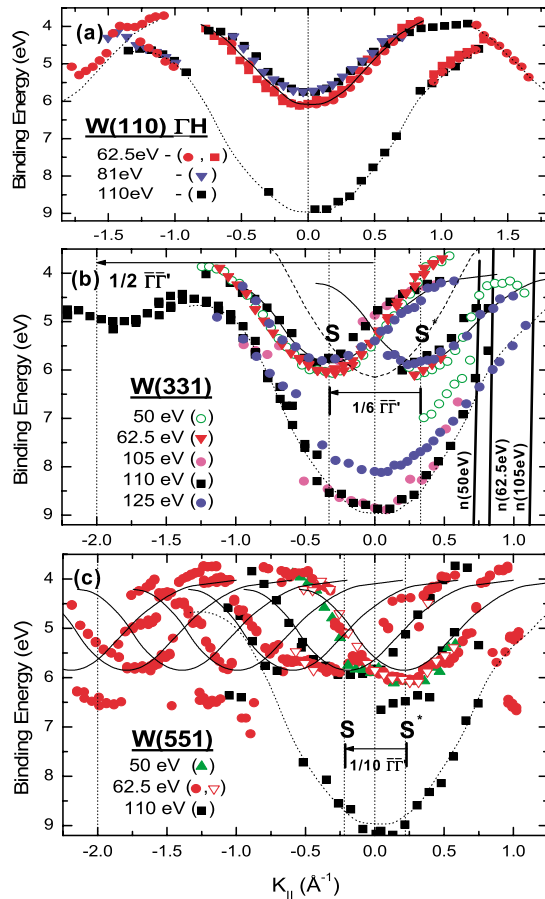


FIG. 4 (color online). Dispersion for \mathbf{k}_{\parallel} -vectors along 001 for W(110) (a) and for W(331) (b) and W(551) (c) i. e., perpendicular to the steps. \mathbf{k}_{\parallel} is given relative to the [110] microsurface normal. The macrosurface normal $n(h\nu)$ shifts strongly between 50 and 105 eV (125 eV outside of the frame) and is therefore not a reference for the repeated surface band S.

repetition of the band structure and that the surface localization is of secondary importance.

In conclusion, we identify a surface-localized feature on W(110) and stepped W(331) and W(551) that shows diffraction effects on a large intensity, energy, and momentum scale in agreement with the lateral superlattice translation vector. The data do not reconcile with an initial-state bandstructure effect of the superlattice but can be explained on the basis of electron scattering from stepped surfaces taking into account where in \mathbf{k} space the photoemission transition takes place. The result that

the repetition is a final-state effect and that the electron wave function is localized at the microsurface is an important prerequisite for tailoring electronic properties in one-dimensional nanostructures grown on stepped W substrates.

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