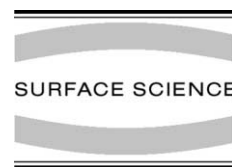




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Modification of the Shockley-type surface state on Ag(1 1 1) by an adsorbed xenon layer

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Abstract

The Shockley-type surface state for the clean and a Xe covered Ag(1 1 1) surface is studied with scanning tunneling microscopy and scanning tunneling spectroscopy at a temperature of 5 K. The minimum of the parabolic dispersion shifts from -67 meV below $E_F = 0$ for the clean Ag(1 1 1) surface to $+52$ meV with one layer of Xe adsorbed, hence the surface state becomes completely unoccupied after Xe adsorption. The dispersion is determined by measuring energy-dependent, locally-resolved, tunneling spectroscopy maps. These show standing wave patterns as local density of states formed by electron scattering at surface steps, which reveal equal effective masses with and without the Xe layer adsorbed. A line width analysis of the energetic onset of the surface state gives a slightly smaller lifetime of the surface state after Xe adsorption, in sharp contrast to the results for image states. We discuss the results and conclude that the wavefunction of the Shockley-type surface state stays close to the metal surface also after Xe adsorption, and that the observed shifts cannot be explained by the simultaneous change of the workfunction, but will be given mainly by the changes of the surface potential on an atomic scale. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Two-dimensional (2D) electron states are formed on several metal surfaces by a confinement of the surface electrons – on the one side by the vacuum barrier and on the other side by a band gap in the bulk states. Especially the surface states

of the (1 1 1) surfaces of noble metals have been investigated in the past as a kind of model system. Besides the general interest in the physics of electrons in low-dimensional systems these surface states can be used to monitor several other phenomena, e.g., surface structure [1], magnetic properties [2] or dynamics of adsorbate motion [3]. Processes on the surface like adatom diffusion or dissociative adsorption can be influenced significantly by the existence of surface states [4,5]. The lifetime of surface states was studied mostly with two photon photoemission for the so-called image states close to the vacuum energy [6–8]. For the Shockley-type surface states which exist some eV

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below the vacuum energy, close to the Fermi energy, the lifetime was studied with scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS) using a line width analysis [9,10] or studying the spatial damping of interference patterns from surface-state electrons scattered at surface steps [11]. The adsorption of atoms or molecules on the metal surface often leads to a quenching of surface states especially if reactive species are chemisorbed, a fact which can be used to distinguish between bulk and surface states [12]. If more weakly bound species are adsorbed, e.g., rare gas atoms, surface states often persist, however with modified energy and lifetime. Such adsorbate induced modifications were studied thoroughly for the image states [13,14] but there exist only a few studies of the corresponding changes for the Shockley-type surface states [6,15]. In this work we present a study for the modification of the Shockley-type surface state on Ag(1 1 1) by an adsorbed Xe layer using low-temperature STS at $T = 5$ K.

2. Experimental

The experiments were performed in a low-temperature surface-science facility, which has been described previously [16]. It consists of a preparation chamber equipped with a high-resolution hemispherical energy analyzer for ultraviolet photoelectron spectroscopy (UPS) and an analysis chamber containing a low-temperature STM operating at $T = 5$ K.

The Ag(1 1 1) sample was prepared by repeated cycles of ion sputtering (1 keV Ar^+ -ions) and heating for one hour at $T = 700$ K. We checked its cleanliness by low-energy electron diffraction, which finally displayed a brilliant diffraction pattern. In a next step we used ultraviolet photoemission to measure the Ag(1 1 1) surface state, which is quenched by minute concentrations of typical residual-gas adsorbates [12]. The ultimate check for cleanliness was then performed by imaging the sample surface using STM.

The Ag(1 1 1) sample was mounted on a manipulator and cooled to below $T = 50$ K by means of a liquid-helium flow cryostat. This manipu-

lator allows cold sample transfer between the two chambers of the surface-science facility [16]. For the measurements presented in this paper 0.5 (± 0.05) monolayers (ML) of Xe were dosed onto the sample at a temperature of 40–50 K by controlling the partial pressure with a leak valve. The Xe coverage was cross-checked with UPS monitoring the Xe 5p photoemission signal during the adsorption process [15]. The coverage could be re-checked with UPS after the STM experiments had been performed at a temperature of $T = 5$ K.

The STS data, either dI/dV spectra measured with varying V at a single position or dI/dV maps measured for fixed V , were taken under open loop conditions by lock-in detection. The setpoint before switching off the feedback loop was $V = +0.2$ V and a tunneling current of $I = 0.12$ nA. For the single-point spectroscopy data we used a modulation voltage amplitude of 1.7 mV_{rms} and a frequency of 700 Hz. The lock-in time constant was set to 10 ms and the acquisition time was 82 ms per data point to avoid time-constant effects. We can give an absolute scaling for the dI/dV data, because we use an active compensation of the cross-talk signal between the modulation voltage and the tunneling current to cancel the corresponding background signal before the tip is approached to the sample. We checked the scaling by a comparison with the numerical derivation of simultaneously measured $I(V)$ curves, which give the same result with a much lower signal-to-noise ratio. dI/dV maps were recorded using a 150×150 pixel grid. At every grid point a dI/dV curve of 15 data points was taken while simultaneously scanning the surface topography in a constant current image. The recording of this entire data set took about 20 h, thus demanding an excellent mechanical and thermal stability of the STM.

3. Results and discussion

In Fig. 1 we present an image of the Xe/Ag(1 1 1) surface as measured with STM at $T = 5$ K. It shows silver terraces divided by steps of monoatomic height. In the left part of the image one terrace is partly covered with Xe which grows in the form of large islands attached to the lower step

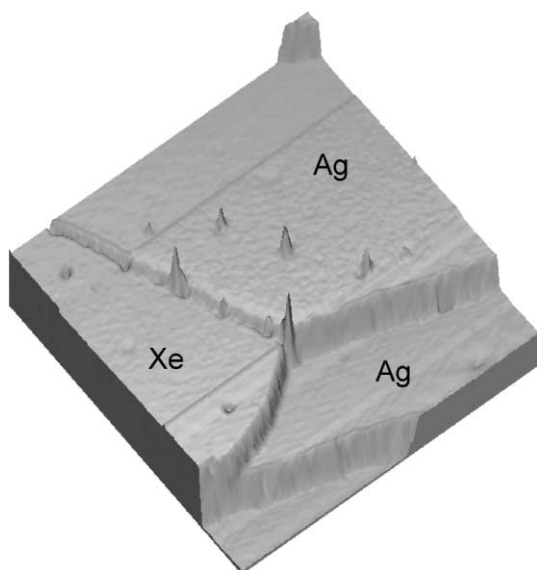


Fig. 1. Pseudo 3D image of the Xe/Ag(111) as measured with STM at $T = 5$ K; image size: 100×100 nm². Silver terraces are divided by steps of monoatomic height. One terrace is partly covered with a large Xe island attached to a lower step edge. The small tip artifacts parallel to the scanning direction from bottom left to top right with a corrugation much smaller than the steps should not be mistaken as Xe boundaries.

edge. A similar island structure was found for 0.5 ML of Xe on Cu(111) annealed to $T > 30$ K [17]. Measurements with atomic resolution (not shown) prove that the Xe atom rows are aligned to the Ag atom rows of the substrate as described in [18]. The topographic measurement without atomic resolution in Fig. 1 alone does not allow a final distinction between clean and Xe covered Ag areas, but as we will show below this is easily accomplished with STS.

For a better illustration we present photoemission spectra of the Ag(111) surface with varying Xe coverage which we measured close to the Fermi energy ($E_F = 0$) with a photon energy of 21.2 eV (cf. Fig. 2). They show a decrease of the surface-state intensity proportional to the Xe coverage with no shift of the peak maximum. In Ref. [15] similar data were measured and explained by a shift of the surface state into the unoccupied region analog to the Xe/Pt(111) system, where the surface state is shifted by $\Delta E \approx 150$ meV after adsorption of 1 ML Xe, which could be measured

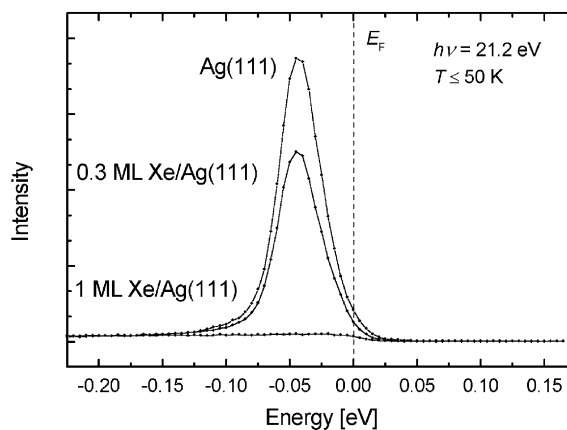


Fig. 2. Photoemission spectra of the Ag(111) surface with varying Xe coverage. The position of the peak maximum is slightly above $E_0 = -67$ meV, resulting from a small deviation from normal electron emission in our experiment because one angular direction is fixed and not adjustable. We mention that similar data were measured in Ref. [15].

with UPS [19] since the surface state stays in the occupied region in this case. Similarly a shift of $\Delta E = 100 \pm 20$ meV has been measured for the system Xe/Cu(111) with two-photon photoemission [6]. With this, one expects a shift of about 100 meV also for the Xe/Ag(111) system but a quantitative value could not yet be determined.

For Ag(111) the influence of Xe adsorption on image states close to the vacuum energy was investigated with two photon photoemission [20], however, in these experiments the photon energies for the excitation were too small to reveal the shifted surface state for Xe/Ag(111). In the past one main method for the investigation of the energy region between the Fermi and vacuum energies was inverse photoemission [21,22], but the limited resolution of this method in combination with the influence of the Fermi distribution function does not allow to identify an energy shift of the order of 100 meV close to the Fermi level.

In contrast, STS is very well suited for this system, because it provides information on both, occupied and unoccupied states. For measurements close to the Fermi energy, as presented here, there is a good correspondence between $dI(x, y)/dV$ and the local density of states (LDOS) $\rho(E = eV, x, y)$, see Refs. [23–25]. The STS data in Fig. 3

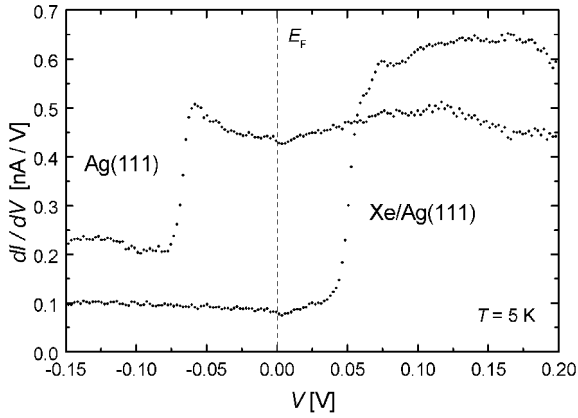


Fig. 3. STS data measured on a clean Ag(111) area and on an area covered with 1 ML Xe; setpoint values: $V = +0.2$ V, $I = 0.12$ nA; modulation voltage: 1.7 mV_{rms}.

show dI/dV curves measured on a clean Ag(111) area and on an area covered with 1 ML Xe. The spectra are step shaped as expected for the LDOS of two-dimensional surface states. The position of the step is $E_0 = -67 \pm 2$ meV for the clean Ag(111) surface, which is in agreement with the data given in [9–11]. For the Xe covered area the step is shifted to $E_0 = +52 \pm 2$ meV. This gives a quantitative measurement of the shift with $\Delta E = +119 \pm 3$ meV. We note that these characteristic curves also allow to distinguish clearly the clean and the Xe covered areas.

For a determination of the effective mass of the surface state we measured dI/dV maps. The surface area in Fig. 4 shows bottom/right a clean Ag(111) surface, which is separated by a Ag(111) surface step of monoatomic height from the area top/left, which is covered with 1 ML Xe attached to the surface step. We present four dI/dV maps for the voltages $V = -20, +60, +80$ and $+140$ mV out of a set of 15 which were measured for voltages in the range of -80 mV to $+200$ mV. The map at $V = -20$ mV shows a standing wave pattern on the clean Ag(111) area while the area covered with Xe appears dark because of the smaller LDOS below the onset of the surface state (cf. Fig. 3). Above $V = +60$ mV both areas show a standing wave pattern. The wavelength of the patterns decreases with increasing V as expected due to the dispersion of the surface state. The wavelength is

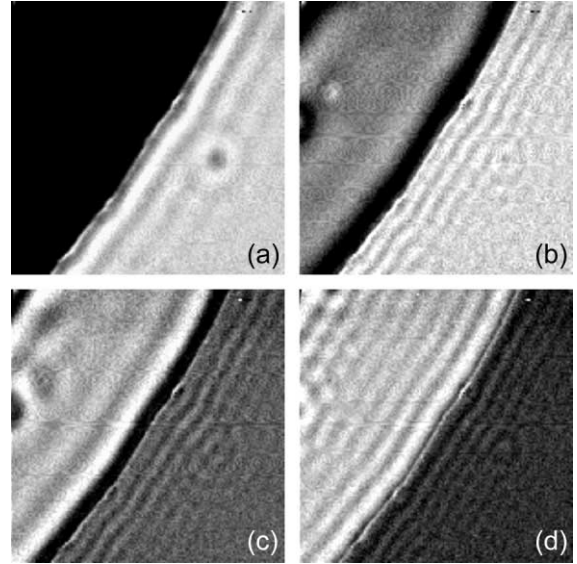


Fig. 4. Maps of $dI(x,y)/dV$ of a Ag(111) surface area crossed by a surface step; image size 50×50 nm². Bottom/right of the step there is a clean Ag(111) surface, top/left the surface is covered with 1 ML Xe attached to the step. (a) $V = -20$ meV, (b) $V = +60$ meV, (c) $V = +80$ meV, (d) $V = +140$ meV. Setpoint values: $V = +0.2$ V, $I = 0.12$ nA; modulation voltage: 7 mV_{rms}.

always smaller on the clean Ag(111) area. For the determination of the dispersion relation $E(k_{\parallel})$ from these maps, we use the fact that the LDOS which results from the reflection of the surface state at a step edge is given by $\rho(E,x) \propto 1 - rJ_0(2k_{\parallel}x)$, with J_0 the Besselfunction of order zero, r the amplitude of the reflection, and x the distance to the step [11,23]. For large x this can be approximated by $\rho(E,x) \propto 1 - r(1/k_{\parallel}x)^{1/2} \cos \times (2k_{\parallel}x - \pi/4)$. Therefore we used as a simple evaluation method the measurement of Δx as the distance between the maxima in the dI/dV maps averaging over several maxima and leaving out the region for small x , i.e. the first maximum behind the step. We get the data for the dispersion relation by this evaluation using $E = eV$ and $k_{\parallel} = \pi/\Delta x$ for the different tunneling voltages. Additionally, we can use the data points $E(k_{\parallel} = 0) = E_0$ with E_0 given by the dI/dV curves of Fig. 3. We show the data points together with fitted parabolic dispersion curves in Fig. 5. We do not give absolute

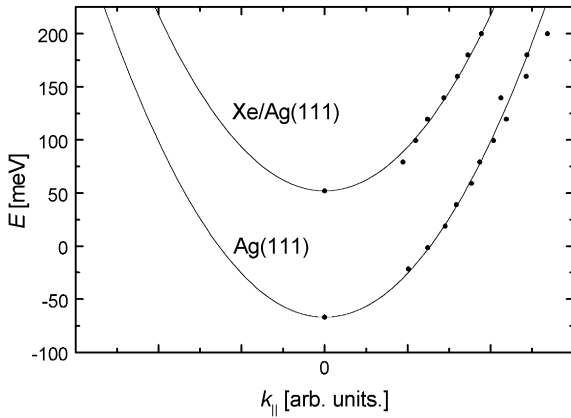


Fig. 5. Dispersion curves $E(k_{\parallel})$ for the clean Ag(111) surface and the Ag(111) surface covered with 1 ML Xe. Data points for $k_{\parallel} \neq 0$ evaluated from Fig. 4 (see text for a detailed discussion), data points for $k_{\parallel} = 0$ taken from Fig. 3. Lines are parabolic curves fitted to the data points.

values of k_{\parallel} in Fig. 5, because our uncertainty of k_{\parallel} due to the errors of the piezo calibration is of the order of 5%, which is a typical value caused by hysteresis, creep etc. because no sample with reference structures, e.g., an atomic lattice was available for this image size. This leads to an error of 10% for the determination of m^* , and additionally the simplified evaluation of k_{\parallel} possibly will cause some systematic error [24,25]. Therefore an absolute determination of m^* out of our data might be misleading. Nevertheless we can conclude from our data that the effective masses m_{Ag}^* and $m_{\text{Xe/Ag}}^*$ are equal within the statistical errors, resulting in $m_{\text{Xe/Ag}}^*/m_{\text{Ag}}^* = 1.00 \pm 0.15$, since the simultaneous measurement of the clean and the Xe covered surface, as shown in Fig. 4, eliminates the systematic errors for the comparison of both.

The standing wave pattern on the Xe covered area in Fig. 4 is brighter and more pronounced as compared to the clean Ag(111) surface. This can be verified by the dI/dV spectra in Fig. 3 looking at the background signal ρ_{bulk} for energies below the surface state onset, which corresponds to tunneling into bulk states, and the height of the step ρ_{2D} , which can be identified with tunneling into the 2D surface state. For the clean Ag(111) surface we get $\rho_{\text{bulk}}/\rho_{2D}$ in the range of 1/1 to 1/2 probably depending somewhat on the tip condi-

tion and similar to the values are given in Refs. [11,26]. On the Xe covered surface we find in contrast a $\rho_{\text{bulk}}/\rho_{2D} \approx 1/6$ (see Fig. 3) which shows that the relative contribution of the surface state electrons detected by STS is much larger for the Xe covered surface than for the clean Ag(111). This indicates that the Xe layer forms a stronger barrier for the tunneling into the bulk states than for the tunneling into the Shockley-type surface state. In both cases dI/dV will correspond closely to the LDOS, and an additional energy dependent background signal [24,25] will be small, because of the almost symmetric positions close to the Fermi energy.

Finally we can use the dI/dV curves in Fig. 3 for an evaluation of the lifetime of the surface state in a similar way as it was already done for the clean Ag(111) surface [9,10]. We determine the parameter Δ given by the intersections of a straight line extrapolating the slope at the midpoint of the rise with the extrapolations of the levels below and above the onset like in Refs. [9,10], which gives $\Delta_{\text{Ag}} = 12$ meV for the clean Ag(111) surface and $\Delta_{\text{Xe/Ag}} = 18$ meV for the Xe covered surface. This indicates a small decrease of the lifetime if the Ag(111) surface is covered with Xe.

For the discussion of the energy shift of the surface state we first look at the influence of the workfunction change induced by the Xe adsorption. The valence states of Xe atoms keep their position relative to the vacuum energy if the workfunction of the surface is changed by previous adsorption of other substances [27], which indicates a weak coupling of the Xe atoms to the surface. But also the Xe adsorption itself induces a decrease of the workfunction ($\Delta\phi = -470$ meV see Ref. [28]). We checked this workfunction decrease in our experiments by measuring the low-energy cutoff of the photoemission spectra. For the adsorption of a low coverage $\ll 1$ ML of alkali metals on Cu(111) a workfunction decrease is accompanied by a smaller decrease of the surface state energy [29,30]. This could be explained in the frame of the phase accumulation model, a model which was successfully used in the description of image states [31]. In this model the electron states confined to the surface are described by a one dimensional potential well formed on the one side by

the band gap of the solid crystal and on the other side by the vacuum barrier. With the energy dependent reflection coefficients $r_C \exp(i\varphi_C)$ to the crystal side and $r_B \exp(i\varphi_B)$ to the vacuum barrier side bound surface states have to fulfill the Bohr-like quantization condition $\varphi_C + \varphi_B = 2\pi n$ where n is an integer. Applied to the case of surface states close to the bottom of the crystal band gap, as it is the case for Cu(111) but also for Ag(111), this theory gives a small influence of workfunction changes on the barrier phase φ_B . Hence a shift of the surface state to lower energies is predicted if the work function is lowered, but by an reduced amount [31]. This is in agreement with the measurements for the alkali adsorption experiments [29,30], but it cannot explain the experiments here – mainly because we observe a upward shift of the surface state to higher energies for the Xe adsorption, i.e., in the opposite direction of the workfunction change.

Two photon photoemission was used in Ref. [20] for a study of Xe/Ag(111). The focus in this experiments was on the image states, close to the vacuum energy, and the photon energies used were too low to detect the unoccupied surface state on the Xe covered surface. A detailed analysis was given in [13]. The energies for the image states on Ag(111) with different Xe coverage up to 9 ML were calculated with two different models. One describes the Xe layers as a dielectric continuum, the other considers quantum well states assuming properties of bulk Xe inside the layer. Both models describe the experimental observation that the image state with $n = 1$ differs from the $n = 2, 3$ states. The reason for this lies in the different spatial distribution of the wavefunctions, and therefore an application of these models also for the Shockley-type surface state (with $n = 0$) measured in our experiments is doubtful, because its wavefunction is positioned very close to the surface, where the details of the potential and the changes by the Xe coverage are not well described by simple model potentials.

The fact that the effective mass of the surface state dispersion stays unchanged by the Xe adsorption can be explained by its energy close to the bottom of the Ag(111) band gap [32]. In this case its dispersion mainly follows the lower edge of the

projected band gap, and this will change only negligibly by the small upward energy shift [33].

The lifetimes of the image states for Xe/Ag(111) were studied with femtosecond two-photon spectroscopy [7]. A lifetime increase by more than a factor of 10 was observed after Xe adsorption. This can be explained by less overlap of the image state wavefunctions with the metal in the case of the Xe covered surface, which results in a weaker coupling to substrate excitations. This behavior can be studied, e.g., by treating the Xe layer as a dielectric continuum [14]. In contrast we observe a slightly increased width of the step in the dI/dV spectrum which indicates a decreased lifetime and supports the assumption that the wavefunction of the Shockley-type surface state stays close to the metal surface also after Xe adsorption.

In summary, we believe that detailed calculations, considering explicitly the potentials at the surface on an atomic scale, will be needed for a quantitative explanation of the shift and the lifetime changes of the Shockley-type surface state for the system Xe/Ag(111). The change of the workfunction caused by the Xe adsorption has probably only a small influence on the observed shift, which is obviously overcompensated by other effects.

4. Conclusion

We have presented results for the Shockley-type surface state of the system Xe/Ag(111). We have shown it to be a special case since the surface state is occupied close to the energy minimum at -67 meV with respect to the Fermi energy for the clean Ag(111) surface and becomes completely unoccupied with a minimum energy of $+52$ meV after Xe adsorption. Due to this two almost symmetric positions close to the Fermi energy a precise measurement of the energy shift with photoemission and inverse photoemission is impracticable. The exact energetic positions are masked by the influence of the Fermi distribution function in combination with a limited energy resolution especially in inverse photoemission. Using STS we were able to determine the energy shift very exactly to $+119 \pm 3$ meV. Hence the upward energetic

shift of the surface state after Xe adsorption is in the opposite direction as compared to the simultaneous decrease of the workfunction. Additionally we used locally resolved dI/dV maps to measure the dispersion $E(k_{\parallel})$ for the clean and the Xe covered surface. The resulting effective masses are equal within the statistical errors: $m_{\text{Xe/Ag}}^*/m_{\text{Ag}}^* = 1.00 \pm 0.15$. A line width analysis of the surface state onset in the dI/dV spectra showed a slight increase of the energy width ($\Delta_{\text{Ag}} = 12$ meV, $\Delta_{\text{Xe/Ag}} = 18$ meV) which indicates a smaller lifetime of the Shockley-type surface state on the Xe covered surface, which is in sharp contrast to the large lifetime increase reported for the image states. The possibility to perform the measurements in a locally resolved manner opens the perspective to extend such studies on further confinement conditions for electronic states by lateral nanostructures on the surface.

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