Upgrade of the Surface Spectrometer at NEPOMUC for PAES, XPS and STM Investigations

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Abstract. The characterization of the elemental composition of surfaces is of great importance for understanding of many surface processes, such as surface segregation or oxidation. Positron-annihilation-induced Auger Electron Spectroscopy (PAES) is a powerful technique for gathering information about the elemental composition of only the topmost atomic layer of a sample. The upgraded surface spectrometer at NEPOMUC (NEtron induced POsitron source MUniCh) enables a comprehensive surface analysis with the complementary techniques STM, XPS and PAES. A new X-ray source for X-ray induced photoelectron spectroscopy (XPS) was installed to gather additional information on oxidation states. A new scanning tunneling microscope (STM) is used as a complementary method to investigate with atomic resolution the surface electron density. The combination of PAES, XPS and STM allows the characterization of both the elemental composition, and the surface topology.

1. Introduction
The surface of a solid is involved in many processes of particular interest such as oxidation, surface segregation, or catalysis. These processes significantly affect many macroscopic properties of thin films or nanoparticles. Detailed knowledge of the surface structure and elemental composition is necessary to understand these processes.

PAES is a non-destructive technique to determine the elemental composition of only the topmost atomic layer of a solid. Positrons are implanted into the sample with an energy of only 20 eV. In the solid, the positrons thermalize within a few picoseconds, and the vast majority diffuse back to the surface [1]. There they become trapped in the surface potential, and initiate the Auger process [2], which is a radiationless transition of an excited atom: an electron from a higher level fills the hole at a lower level, and the released energy is transferred to another electron, which leaves the atom with an element-specific energy. In the case of PAES, the hole in the inner shell is created by electron-positron annihilation. Typically, the core annihilation probability for positrons is in the range of 1 to 10% [3]. PEAS exhibits two essential advantages: due to the low energy of the incident positrons, the spectrum is — in contrast to XAES and EAES — free of secondary electron background in the energy range of the Auger transitions, leading to an excellent signal-to-noise ratio. The surface trapping leads to the outstanding surface sensitivity of PAES.
To overcome the main drawback of PAES measurements — the long measurement time when using $\beta^+$ lab sources — the surface spectrometer is attached to the positron beam facility NEPOMUC at the research reactor Heinz Maier-Leibnitz. This reactor-based source delivers a 1 keV positron beam with an intensity of $10^9$ positrons per second. For brightness enhancement, this primary beam is re-moderated with a tungsten single-crystal with an efficiency of 5% [4]. This high-intensity positron beam yields measurement times of only a few minutes per PAES spectrum, and enables time-dependent observation of e.g. surface segregation processes [5].

A new X-ray source for XPS measurements was recently installed at the spectrometer. XPS, as an established method for elemental characterization with its high chemical specificity, is a particularly suited completion of PAES for e.g. the determination of oxidation states. Due to the high penetration depth of the X-rays, XPS, in contrary to PAES, shows the average of the elemental composition of 3-10 atomic layers, depending on the energy of the photo electrons.

As a complementary method to determine the surface structure, a new scanning tunneling microscope is used. STM images the electron density distribution with atomic resolution. Thus, changes in the crystal structure, such as dislocations or grain boundaries leading to a variation in the surface potential, become visible. These deviations are attractive for positrons, and therefore influence the probability density of positrons. Hence, STM data is of special interest for the interpretation of PAES spectra, in order to determine the annihilation site of the positron. Furthermore, in a first approximation, the mapping of the electron density distribution can be seen as the topology of the surface.

2. Experimental setup of SuSpect

The upgraded Surface Spectrometer (SuSpect) combines sample preparation, a sample storage, and analysis with STM, XPS and PAES under UHV conditions in one setup (see figure 1). For sample cleaning, an Ar-ion sputter gun and electron impact heating can be used. Three electron beam evaporators are installed, to prepare coated samples. The thickness of the coating is controlled by a piezo-oscillator serving as a thickness monitor. Thicknesses from a fraction of an atomic layer (ML) up to several hundred nm are possible.

The samples are transferred by magnetically linked linear feedthroughs to the analysis chambers. In the first analysis chamber the STM Aahrus 150 HT, with a temperature range between 90-1300 K, a sample storage for up to 5 samples, and an electron impact heater for sample preparation are installed.

The next analysis chamber houses the setup for Auger spectroscopy. For conventional Auger spectroscopy a focusable electron gun with a tunable emission energy up to 5 keV and an X-ray source with Mg/Al twin anode are used. For PAES measurements the re-moderated positrons are guided adiabatically to the analysis chamber where they pass a magnetic field termination and are focused electrostatically onto the sample. To avoid the influence of external magnetic fields, the analysis chamber is $\mu$-metal shielded. A NaI(Tl) scintillator is used for detection of $\gamma$-radiation from the positron annihilation. For beam adjustment a moveable MCP with a phosphor screen and CCD readout was recently installed. The electron detection is done with a Phoibos 150 hemispherical energy analyzer with a large acceptance angle of up to $\pm 13^\circ$. The energy analyzer is bipolar, with the result that reemitted or backscattered positrons are also detectable. Readout is done with a cooled CCD camera. In both analysis chambers, an ion getter pump and a titanium sublimation pump are used, in addition to turbo molecular pumps, to obtain a vacuum in the range of $10^{-9}$ mbar and below. For residual gas analysis a mass spectrometer is used.

3. First measurements

Several measurements were done at the upgraded spectrometer to demonstrate the performance of all components. For the STM a Si(111) wafer was used as reference. Figure 2 shows the
occupied and empty states of the Si(111) \( 7 \times 7 \) surface reconstruction with atomic resolution, recorded with the new STM. The sample was cleaned with acetone and deionized water, followed by annealing for 30 minutes and flashing using electron impact heating. Afterwards, a Cu(100) crystal was investigated. The STM image in figure 3 shows the \( \sqrt{17} \times \sqrt{17} \) R14-reconstruction of S atoms on the Cu(100) surface in atomic resolution.

XPS measurements with the new X-ray source were performed on Cu and Fe-coated Cu foils. For this purpose Cu foils were cleaned with Ar-ion sputtering at 1 keV for 1 hour and annealed with electron impact heating. Subsequently, the foils were coated with 3 ML and 6 ML Fe respectively, evaporated from a Fe rod with a purity of 99.99% using electron beam evaporation. The thickness was controlled with a piezo-oscillator thickness monitor. The XPS spectra of the three samples are shown in figure 4 with different offsets for reasons of clarity.

Recently, a first PAES measurement on Cu was performed, after the modification of the spectrometer and the upgrade of the NEPOMUC source [4]. The re-moderated positron beam with an energy of 20 eV was used. From the count rate of the NaI(Tl) scintillator the positron intensity on the sample was calculated to be \( 7.5 \times 10^6 \) e\(^+\)/s. Figure 5 shows the PAES spectrum with the Cu MVV Auger transition at 60 eV.

4. Conclusion and outlook
In this work the upgraded surface spectrometer at NEPOMUC was presented. The spectrometer enables a comprehensive surface analysis using PAES as a non destructive, element selective analysis method with outstanding surface sensitivity, XPS with its chemical specificity, and STM for topological imaging of the surface with atomic resolution. In order to reduce the measurement time for PAES spectra and enable time-dependent PAES measurements, a moveable MCP was recently installed and will be used for focusing a precise beam onto the sample. Future studies
Figure 2: STM image of occupied (left) and empty states (right) of the Si(111) 7 × 7-reconstruction with atomic resolution.

Figure 3: STM image of $\sqrt{17} \times \sqrt{17}$ R14-reconstruction of S atoms on the Cu(100) surface with atomic resolution.

Figure 4: XPS spectra of a Cu reference and Fe coated Cu foils with different offsets.

Figure 5: PAES spectrum of Cu with Cu MVV Auger-transition.

will be focused on the detection of surface segregation processes in order to benefit from the surface sensitivity and elemental selectivity of PAES in combination with complementary STM imaging and XPS.

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