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Ion transmission through a dielectric hollow tip for scanning probe microscopy

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Keywords: Point-projection microscopy Hollow tip Photoions Scanning probe microscopy Femtosecond laser radiation	We present a new concept for scanning probe microscopy characterization of molecular microstructures. It is based on a thin capillary using as a sharp tip to probe and map the morphology of a surface. In our experiment a collimated ion beam is formed by tungsten ions passing through a quartz tapered capillary with a 100-nm aperture and enters a 2D position-sensitive detector. We demonstrate that such ions are capable of producing the image of a dielectric nanoaperture in the case of low-dose ion beam. Ion transmission through a nanoscale capillary opens the door to observing photodesorption of large organic molecular ions with high spatially-ele- ment resolution using the combination of a hollow-tip vacuum scanner with time-of-flight mass spectrometer.

1. Introduction

Electron transmission through a dielectric hollow tip opened new perspectives for different applications in science and technology (Stolterfoht and Yamazaki, 2016; Aseyev et al., 2006, 2017; Stolterfoht and Tanis, 2018; Maljković et al., 2018). A thin capillary can be used in scanning probe microscopy (SPM) as a topologically-complex tip under vacuum conditions (Aseyev et al., 2017). Photoelectrons emanating from a substrate can be transmitted through such a capillary as a function of the tip position on the substrate. This approach was demonstrated on a model sample consisting of a gold reflecting layer on a compact disc which has been irradiated by second harmonic of femtosecond Ti:sapphire laser. The period of gold microstructure, shown to be 1.6 μ m, was measured by a quartz tapered capillary with 2- μ m outlet diameter operating in shear force mode. In this regime, the dielectric thin capillary has been used as a SPM tip, which provided images reflecting the surface topology. In a photoelectron regime photoelectrons passed through this capillary and entered a detector. The spatial distribution of the detected photoelectrons consisted of periodic strips, resembling the surface profile of the model substrate. In (Aseyev et al., 2017) submicron lateral resolution has been achieved and the surface topography of the sample was reproduced, although the period of substrate microstructure was less than outlet diameter of the hollow-tip. We anticipated that the resolving possibilities of the method could be improved by using a nanometric hollow probe.

A photoion regime, using a very thin capillary in transmittance geometry, paves the way to new possibilities of vacuum scanning capillary microscopy. In this connection it is of interest to investigate ion transmission through such a hollow tip with reference to a possible visualization of different nanostructures. In proof-of-principle experiment we study the ions escaping a nanometer quartz capillary. For these measurements we use point-projection microscopy technique (Muller, 1965; Letokhov, 2007; Quinonez et al., 2013) which forms a highly enlarged real image of a 100-nm aperture of the quartz tapered nanocapillary on a distant position-sensitive detector. The obtained results make it possible to develop a new concept for scanning probe microscopy characterization of organic molecular nanostructures.

2. Experimental

In our apparatus, shown schematically in Fig. 1, the point-projection microscope is equipped with a 2D position-sensitive detector comprising a pair of micro-channel plates with active zone diameter of 24 mm and a phosphor screen. The image formed on the screen is monitored by a CCD camera. The face of the detector is at ground potential. The photoions pass through a quartz conical capillary with an aperture of ~100 nm (μ TIP^{**}, WPI) and travel a distance of 100 mm before being detected.

A tungsten sharp electrode with a radius of curvature of $1 \mu m$ is inserted in the capillary from the rear part in such a way, that the tungsten apex is separated by ~4 mm from the quartz apex. An assembly including the tungsten sharp electrode and the nanoscale capillary is mounted on a movable feedthrough and is oriented so that it is facing the detector. The tungsten electrode is illuminated by the second

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Fig. 1. Schematic of the experimental setup. A 2D position-sensitive detector measures the distribution of an ion beam passing through a quartz nanometric capillary.

harmonic ($\lambda_{\text{laser}} \approx 400 \text{ nm}$) of a Ti:sapphire laser through the transparent quartz walls. The assembled ion source is driven by focused microjoule-energy 1-kHz laser pulses of about 100 fs in duration at different power densities of up to $\sim 5 \times 10^{12} \text{W/cm}^2$. A copper grid with a mesh size of about 1 mm, placed downstream at a distance of about 7 mm from the capillary apex, makes it possible to observe nanometer collimation of slow ions with kinetic energy down to a few eV, determined by the electric potentials of U_W electrode and U_{Cu} grid, applying across the tungsten electrode and the grid, respectively.

The experimental setup is housed in a vacuum chamber pumped by a turbo-pump to an oil-free vacuum of $\sim 10^{-7}$ mm of Hg. Our apparatus is also equipped with a secondary electron multiplier (not shown in the Fig. 1), which is a part of homemade mass spectrometer with a time-of-flight base of about 150 mm. The photoion signal from different laser-irradiated targets (tungsten, copper and aluminum) makes it possible to calibrate the mass spectrometer response.

3. Results

In order to elucidate the physics of ion generation under an intense optical field, the photoions emitted from a laser-illuminated tungsten surface are detected as a function of laser power density (Fig. 2) in a time-of-flight mode. Based on the data collected, the following mechanism was formulated to explain the formation of tungsten ions.

Intense laser radiation heats the metal and, as a result, neutral tungsten atoms can be formed near the surface. Then, free tungsten atoms are ionized by femtosecond laser field. The heating effect is



Fig. 2. Number of detected W^+ photoions as a function of the intensity of focused 400-nm, 1-kHz femtosecond laser pulses illuminating the tungsten surface. Circles and solid line indicate the experimental data and the power-law approximation, respectively.

verified by decreasing the repetition rate of the laser pulses down to 50 Hz, which is accompanied by the actual disappearance of a photoion signal under these conditions. For 1 kHz pulsed laser irradiation the experimentally-observed yield of the photoions (see Fig. 2) is satisfactory well described by power-law dependence as a function of laser intensity:

$N_{W^+} \propto I_{laser}^n$,

where $n \approx 2.51$. In this connection, note that the 7.9 eV ionization potential of W is approximately 2.55 times higher than 3.1 eV photon energy for the second harmonic of the output of the Ti:Sapphire amplifier system. In (Keldysh, 1965) Keldysh derived the tunneling criterion for the possibility of ejecting an electron that has bounded potential:

$$\gamma = \frac{\omega_{laser} \sqrt{2m_e I_{pot}}}{eE_{laser}} \le 1,$$

where γ is the Keldysh `s adiabaticity parameter, ω_{laser} is the laser frequency, $m_{\rm e}$ and e is the mass and the charge of an electron, I_{pot} is the atom ionizing potential and E_{laser} is the amplitude of a laser electric field. It was found that the multiphoton ionization (MPI) and the tunnelling ionisation are two limiting regimes of nonlinear ionisation, while MPI occurs if $\gamma > 1$ (Keldysh, 1965). Under the experimental conditions, the MPI criterion is satisfied as $\gamma \geq 2$.

Fig. 3(a) presents the observed ion density distribution when U_W $_{electrode} = 1500$ V and $U_{Cu grid} = 1300$ V, while Fig. 3(b) corresponds to the case when U_W $_{electrode} = 1500$ V and $U_{Cu grid} = 1494$ V. The time of acquisition is about 2 and 6 s for the data presented in Fig. 3(a) and (b), respectively. Such a low acquisition time, corresponding only to a few thousands femtosecond laser pulses, allows us to prevent noticeable degradation of the tungsten electrode due to its ablation under an intense optical field. The two images resemble each other, however some blurring is present in Fig. 3(b). We attribute this to spreading of a few eV ion beam due to Coulomb repulsion of the ions during transit from tungsten electrode to copper grid. From the experimental results it is clearly seen, that the detected ion density distribution has an arc shape, which can be explained by the following model.

Actually the ions emitted within a broad solid angle inevitably collide with the capillary walls before reaching the outlet of the tapered capillary. But these ions after their collisions with the capillary walls develop a circular distribution in the plane of the 2D position-sensitive detector, while only a rather small portion of the ion beam, depending upon the capillary opening, is ballistic. To our opinion, such an argument holds true in the case of low-dose ion beam transmission, when it is possible to neglect the charging effects. This explanation is illustrated in Fig. 4. We recall that such a nanocollimation was discovered previously for pulsed photoelectron beam, transmitting through the tapered quartz capillary with ~ 100 nm outlet diameter (Aseyev et al., 2006).

From the above-mentioned model, we can conclude that the

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