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Scanning photoelectron microscopy using a pointed capillary probe

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Abstract. **The possibilities of a new type of scanning probe microscopy (SPM) for two different samples are experimentally demonstrated. The method is based on the use of a pointed capillary, which can simultaneously act as a 'classical' SPM probe and also as a controlled thin channel for transporting charged particles emitted by the surface to the detector. In the experiment, photoelectrons pass through a dielectric hollow cone probe with an aperture radius of 1 μm and detected by microchannel plates at different points of the investigated conducting surface irradiated by the second harmonic of a femtosecond Ti:sapphire laser. As a result, the sample's surface profile is visualised with a subwavelength spatial resolution. This method makes it possible to control spatially localised beams of electrons, ions, neutral atoms (molecules) and soft X-ray radiation, as well as opens a possibility for research in the field of nanoscale photodesorption of molecular ions.**

Keywords: *femtosecond laser radiation, photoelectron microscopy, capillary probe.*

1. Introduction

The use of a hollow needle with a channel along its axis offers ample opportunities for scanning probe microscopy (SPM), which cannot be implemented when the sample is analysed by means of a 'classical' tip. Under these conditions, experiments can be fulfilled using various beams passed through a hollow needle: electrons, ions, neutral atoms and molecules, as well as short-wavelength electromagnetic radiation, for example, soft X-rays. Actually, thin channel acts here as a versatile collimator. At the same time, within the framework of this approach, it is possible both to modify the sample surface and to study its relief by recording the transmitted beam at various points of the sample under study. Naturally, thin channels can be formed in solid structures which differ in their geometric dimensions and chemical composition.

Thus, earlier experiments in the field of transmission of multiply charged ions [1] and electrons [2] through pinholes in dielectric materials (track channels in polymer films [1]) have been conducted in the framework of the formation of promising elements for controlling charged particle beams that do not require the use of electric and magnetic fields [3].

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In these works, it has been shown that charging the inner surface of a dielectric plays a rather important role in the propagation of multiply charged ions inside such a channel, when it is necessary to avoid the beam collisions with the walls. Conducting materials are of certain interest for transporting beams of electromagnetic radiation. A thin conducting channel has successfully showed itself as the basis for a promising point source of shortwavelength electromagnetic radiation of ultrashort duration, formed with the use of femtosecond light pulses generated from a high-repetition-rate laser generator [4, 5].

A pointed capillary made of a dielectric material may act as a single channel. The developed procedure for 'pulling out' the conical point makes it possible to form hollow needles with various geometric dimensions: in the laboratory version, the through-hole diameter at the tip may constitute up to 10 nm [6]. There are commercially available quartz capillaries with a diameter of 0.1 μm. The collimation of beams in such a capillary made it possible to form spatially localised electron bunches [7, 8], ions [9] and soft X-rays [10]. Using such a capillary, it became possible to implement for the first time laser photoelectron projection microscopy of an organic sample [11] and to establish the existence of redox heterogeneity of an organic conducting polymer, stipulated by the contact of polyaniline regions having different oxidation degrees [12]. In this work, a quartz capillary with a layer of a conductive organic polyaniline-based polymer applied to its surface was used as a projection microscope needle. At the next stage, it turned out advisable to investigate the possibilities of the SPM method when using a hollow pointed capillary probe.

The use of a thin capillary as a hollow probe is not the only possible solution: for example, a probe in scanning optical and microwave microscopy may represent a planar microstructure with a very small aperture [13]. However, this scheme has its limitations. It is virtually impossible to introduce a planar diaphragm into the area of contact with a surface having a rough relief. In addition, difficulties may arise with the controlled supply of a focused laser beam to a part of the sample under investigation, which is conjugated with the 'capture' region of the photoelectron signal.

In this paper we present the results of a study of two conducting samples by means of scanning vacuum photoelectron microscopy, when electrons emitted under the action of laser radiation are directed by a hollow conical tip to the detector and recorded at various points of the surface under investigation. Earlier, this method has been employed to demonstrate the possibility of achieving subwavelength spatial resolution using a gadolinium sample [14]. The purpose of this paper is a more detailed description of our experiment, including application of the new method to studying a spatially periodic gold microstructure.

2. Scheme of the experiment

The scheme of experiment is shown in Fig. 1. A vacuum chamber is pumped by a turbo-molecular pump to a pressure of $\sim 10^{-7}$ Torr. A scanning microscope based on piezoelectric modules and equipped with a quartz capillary with an aperture diameter of 2 μm at the tip is placed inside the chamber. The microscope is designed and assembled at the Institute for Spectroscopy of the Russian Academy of Sciences. To eliminate the impact of the oscillations associated with the operation of vacuum pumps on the measurement results, a spring-actuated vibration uncoupler is provided in the device. To solve the problem of nondestructive contact between the micron or submicron aperture of a rigid capillary with the capillary probe surface during the scanning process and numerous measurement cycles using the same probe, a special design of the contact sensors based on quartz resonators is applied [14, 15]. This allowed us to achieve the same high sensitivity and operation speed as in atomic force microscopy. The instrument scanning area may reach 20×20 μm. The probe positioning accuracy during the vacuum equipment operation amounts to several tens of nanometres.

The experimental setup is equipped with a position-sensitive detector of charged particles, based on a pair of microchannel plates (MCPs) and a luminophore. In the experiment, a highly sensitive recording scheme coupled with a detector is used, which allows integrating the flux of photoelectrons transmitted through the hollow probe. A negative (pushing) potential is applied to the metal sample. To cut off the electrons that have not passed through the tip, a special screen consisting of several metal plates is installed to prevent the electrons from hitting the detector. As a result, a pulse signal reflecting the local emissivity of the surface under study is recorded. The scan step is a few tens of nanometres.

A metal sample was irradiated with the second harmonic of a femtosecond Ti:sapphire laser (λ = 400 nm) operating at a repetition rate of 1 kHz. In the experiment, a linearly polarised laser beam was used. The duration of the laser pulses was about 70 fs.

To demonstrate the photoelectron scanning microscope operation, a gadolinium sample having a tip with submicron roughness was selected. A small rod with a 'triangular' top was pasted in vertical position into a drop of epoxy glue on a substrate, and a spring-loaded electrical contact was applied to the rod to supply the potential to accelerate electrons ($U = -130$ V).

Laser radiation was focused by means of a lens with a focal length of about 0.3 m into a spot about 40 μm in diameter, which covered the pointed region of the gadoline sample. The light beam was directed to the microscope horizontally, and, by means of precise adjustment of the mirrors, was directed into the region of the capillary tip placed above the Gd sample. Scanning with a capillary probe was carried out in the region of the pointed part of the sample, with the probe not going beyond the focal spot.

To search for a highest point of the vertically-installed metal rod, a cycle of measurements were performed with the laser beam blocked. After each measurement, the capillary was moved by means of a programme to a higher region of the sample, predicted on the basis of the obtained data. Then the next sample scanning in the plane was performed.

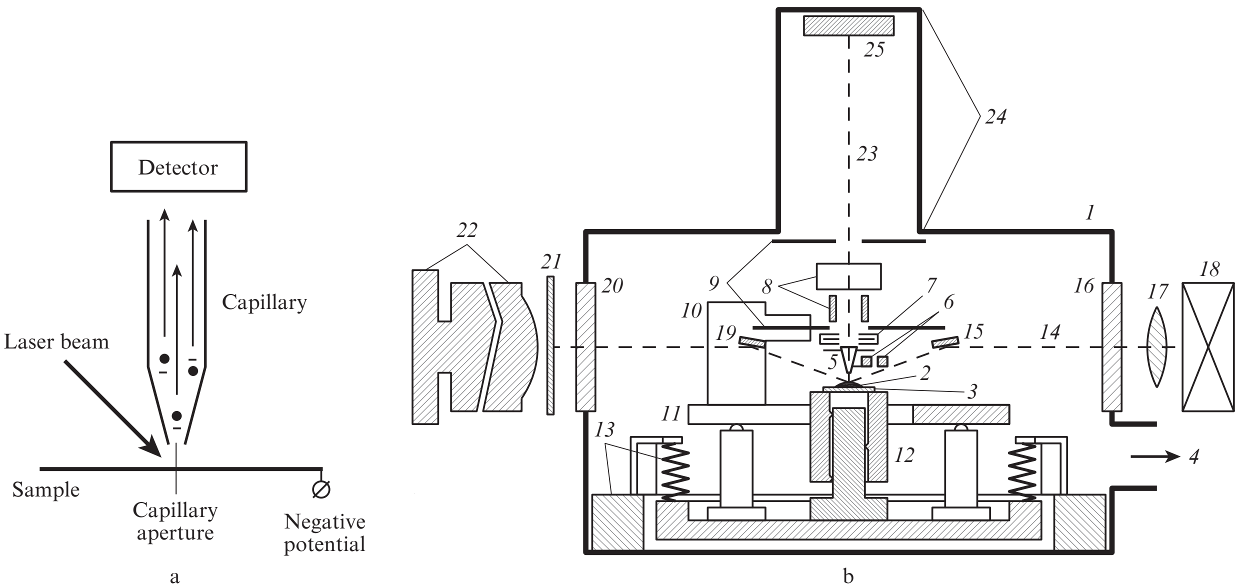

Figure 1. (a) Schematic of the experiment and (b) design of the scanning photoemission microscope:
(*1*) vacuum chamber body; (*2*) sample; (*3*) substrate with a negative electrical potential; (*4*) vacuum pumping; (*5*) quartz conical capillary glued into the plate; (*6*) horns of the contact sensor's tuning fork; (*7*) electrostatic lens for focusing onto the microchannel plates (MCPs); (*8*) electrostatic *XY*-deflecting plates; (*9*) screens for spurious signal blocking; (*10*) rotary-translational cantilever mechanism connecting the assembly unit, which includes elements (*5*, *6*, *7*, *8*, *15*, *19*), with the *XY*-scanner table; (*11*) *XY*-scanner table; (*12*) *Z*-scanner carriage with the sample; (*13*) spring-actuated suspension of the scanner; (*14*) laser beam; (*15*, *19*) aluminium mirrors; (*16*) quartz window; (*17*) focusing electrostatic lens; (*18*) laser beam positioning system; (*20*) output window; (*21*) light filter for laser beam attenuation; (*22*) telescope with a TV camera; (*23*) trajectory of photoelectrons travelling along the axis; (*24*) time-of-flight tube; (*25*) MCP.

This process was continued until the rod's highest point turned located approximately at the centre of the microscope scanning area. Conducting the experiment at the upper point made it possible to control the process of pointing a tightly focused laser beam at the capillary tip using an auxiliary television camera and a telescope. In addition, at the upper point of the sample, the intensity of the electrostatic field accelerating electrons must be directed along the capillary axis, which facilitates the ballistic flight of photoelectrons through such a probe.

When tuning the instrument, an electrostatic lens was used (Fig. 1b), which allowed us to focus the photoelectron beam into the region of the position-sensitive detector. As a result, the photoemission signal was a bright spot. As an additional control, we used deflecting metal plates to vary the position of this spot on the detector in the XY plane.

In the process of scanning the gadolinium sample irradiated with a laser beam, a map of local photoemissivity of the metal surface with a rough relief was recorded. These data were then compared with the results obtained without laser radiation in the 'classical' scanning microscope regime (shear force regime), when the thin capillary functioned as a tip.

In the second stage, a gold reflective layer of a compact disc (CD) from which the plastic cover was beforehand removed was used as a sample. The investigated surface was similar to a diffraction grating having rectangular shape of fringes with a period of 1.6 μm, covered with a thin layer of gold. We should note that the probe aperture diameter exceeded the period of the metal structure. The sample was irradiated by a wide laser beam with a diameter of 4 mm. The beam incidence angle was about 70°. The accelerating potential with respect to the camera body was $U = -300$ V. To increase the operation speed and reliability of the measurements, the sample surface relief in the shear force regime and the map of its local photoemissivity were recorded simultaneously.

3. Results and their discussion

The results of scanning the tip of a pointed gadolinium rod in the shear force regime are shown in Fig. 2a. The scanning time was about 6 min, and the scanning step was 25 nm. The characteristic 'jackboot' shape reflects the test sample relief.

The image obtained in the photoelectron regime (Fig. 2b) has its basic features similar to those shown in Fig. 2a. In this case, the scanning time was about 13 min, and the scanning step was 80 nm. Let us take note of the presence of bright spots with a diameter of about 400 nm, which is much smaller than the aperture size of the hollow probe. These features are present in both image frames. Such a correlation may be indicative of the presence of a protrusion or a submicron cavity at the sample tip.

For a more vivid illustration of the method, let us consider the results we have obtained using a spatially regular gold microstructure (Figs 3, 4). The image of local emissivity (Fig. 4a) is in good agreement with the image of the relief (Fig. 3a), which confirms the photoelectron image objectivity. As it turned out, the photoelectron image of fringes has the fronts of 200 nm (Fig. 4b), which is an order of magnitude smaller than the aperture diameter and twice less than the wavelength of the laser radiation irradiating the microstructure. It also follows from the results obtained that the inner surface of a dielectric capillary, when the sample is irradiated by the laser, is charged by electrons, which may result in the signal decrease. This is clearly shown in the left part of Fig. 4a, where a dark band is present, reflecting a decrease in the

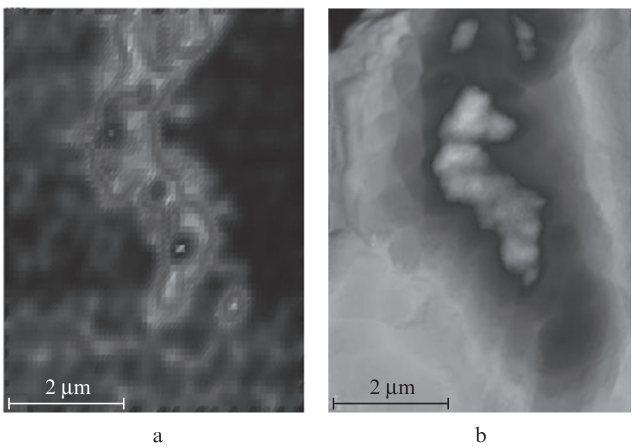

Figure 2. (a) Topography of the gadolinium sample surface in the 'classical' probe regime and (b) the image of this surface in the photoelectron regime when charged particles pass through a two-micron quartz capillary.

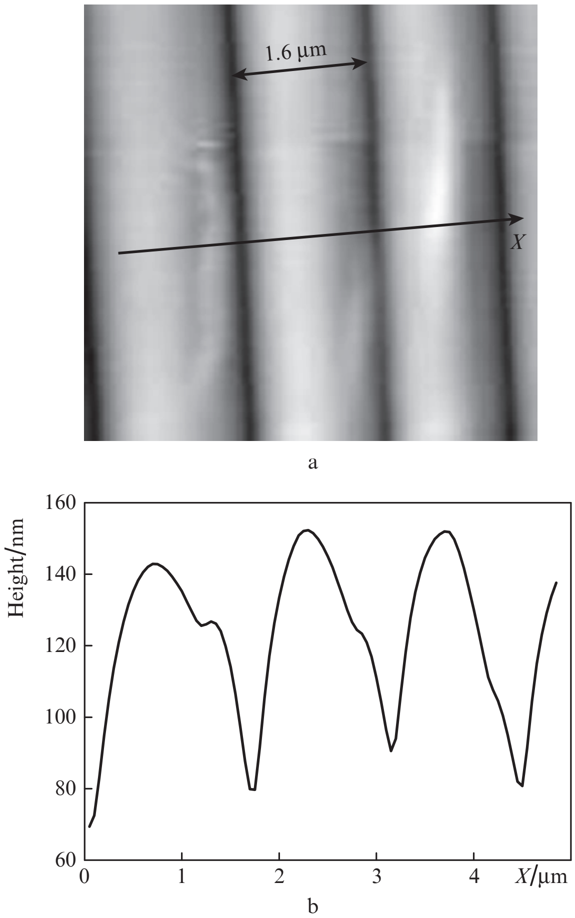

Figure 3. (a) Image of a spatially periodic gold microstructure obtained by the shear force method and (b) the image cross section along the X axis, averaged over all scanning lines.

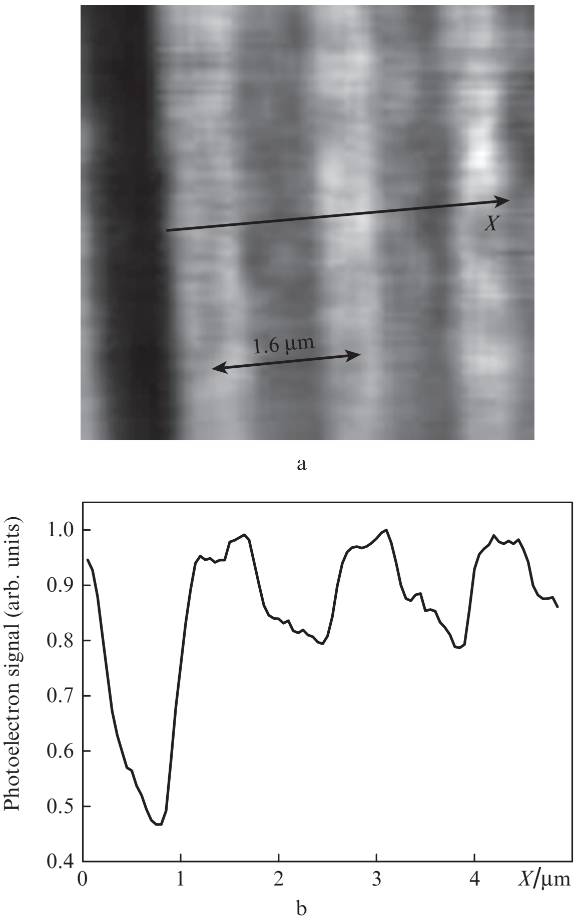

Figure 4. (a) Map of local photoemission activity of a spatially periodic gold microstructure obtained in detecting a photoelectron signal through a hollow probe and (b) the cross section of the photoelectron image along the X axis, averaged over all scanning lines.

number of recorded electrons. The point is that the return of the probe each time after scanning the subsequent fringe is performed in a relatively fast noncontact way, and therefore is accompanied by the inevitable charging of the capillary tip at the expense of emitted electrons. In the scanning process, when the tip contacts the conducting surface, a discharge with restoration of the photoelectron signal level occurs.

At first glance, the spatial resolution of the given type of SPM should be determined by the aperture size at the capillary probe tip. However, it can be substantially higher, as demonstrated experimentally. For a possible explanation, we should take into account that in the ‘classical’ shear force regime, when the probe tip has a clearly asymmetric shape, the microscope capabilities are determined by the characteristic dimensions of the protrusion at the capillary tip.

As demonstrated experimentally, the method provides a high degree of detail in the research. We should note that the method’s spatial resolution can be improved by using a capillary probe with a diameter of 0.1 μm or by depositing a thin metal layer onto the tip of the dielectric needle. In terms of the method development, it is of particular interest to investigate the possibilities of the capillaries made of weakly conducting materials. To increase the microscope operation speed, it may be advisable to use femtosecond laser radiation with a megahertz pulse repetition rate, generated, for example, by a Mai Tai laser.

4. Conclusions

Thus, a method based on transmission of photoelectrons through a thin dielectric capillary and recording the photoelectron signal at various points of the sample irradiated by femtosecond laser radiation makes it possible to obtain a surface image qualitatively identical to the results obtained by means of a ‘classical’ scanning probe microscope. The possibilities of this approach are demonstrated for two conductive samples with different reliefs and chemical compositions. The spatial resolution achieved lies in the region of ~200 nm, which is less than the wavelength of the laser irradiating the sample and substantially less than the aperture diameter at the probe tip. In the future, it seems expedient to proceed to working with the capillaries having an aperture of ~100 nm, which will allow experimental implementation of pulsed nano-local photodesorption of molecular ions and combine it with time-of-flight mass spectrometry. The corresponding concept of a ‘nanojack plane’ formulated by V.S. Letokhov in the 2000s is aimed at developing high-sensitive microscopy of organic molecular structures on the surface, when it is necessary to determine the spatial–chemical composition of the sample under study with a high degree of detail. It was assumed to use ultrashort laser pulses for photodesorption of molecular ions, to implement multiphoton processes without significant thermal heating of the sample, and to lower the photodetachment threshold of molecular ions from the surface [16, 17]. The transmission of a pulsed beam of photoions through a controlled capillary probe opens a possibility for achieving a high spatial resolution.

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