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Frederik Walla, Matthias M. Wiecha, Nicolas Mecklenbeck, Sabri Beldi, Fritz Keilmann, Mark D. Thomson, and Hartmut G. Roskos*

Anisotropic excitation of surface plasmon polaritons on gold by a scattering-type scanning near-field microscope

Supplemental information

Supplemental A: Probe tips

Figure 3(a) in the main text displays three images of a cantilever/probe tip used in our experiments (Mikromasch HQ:NSC14/Pt). Fig. 1 of this supplemental note displays additional scanning-electron-beam (SEM) micrographs of one of our HQ:NSC14/Pt probe tips which has been in use as evidenced by the damage or contamination at the apex. When approximating the shape of the tip's shaft as conical, one finds a full cone angle of roughly 40°. The actual shape of the shaft, as revealed in Fig. 1, is that of an asymmetric quadrilateral pyramid at the shaft's base, which changes towards the apex into a triangular pyramidal form (see also website of manufacturer at http://www.spmtips.com/afm-probes.afm). The ridge facing towards the front of the cantilever disappears, leaving a slightly curved, outward-bulging surface in the vicinity of the apex. This is an important observation for our experiments, because it makes it understandable why the

Frederik Walla, Matthias M. Wiecha, Nicolas Mecklenbeck, Sabri Beldi, Mark D. Thomson, Hartmut G. Roskos, Physikalisches Institut, Johann Wolfgang Goethe-Universität, Max-von-Laue-Strasse 1, 60438 Frankfurt am Main, Germany
Fritz Keilmann, Soft Condensed Matter Group and Center for NanoScience, Ludwig-Maximilians-Universität, Geschwister-Scholl-Platz 1, D-80539 München, Germany
Frederik Walla, Present address: Department of Electronics Technology, Universidad Carlos III de Madrid, Leganes, Madrid 28911, Spain
Sabri Beldi, Permanent address: Centre de Recherche en Microélectronique et Nanotechnologie, Technopôle de Sousse, Sahloul, Tunesia
Frederik Walla, Matthias M. Wiecha, These authors contributed equally
***Corresponding author: Hartmut G. Roskos,** roskos@physik.uni-frankfurt.de

laser radiation is so pronouncedly scattered/reflected in backward direction in our measurements.

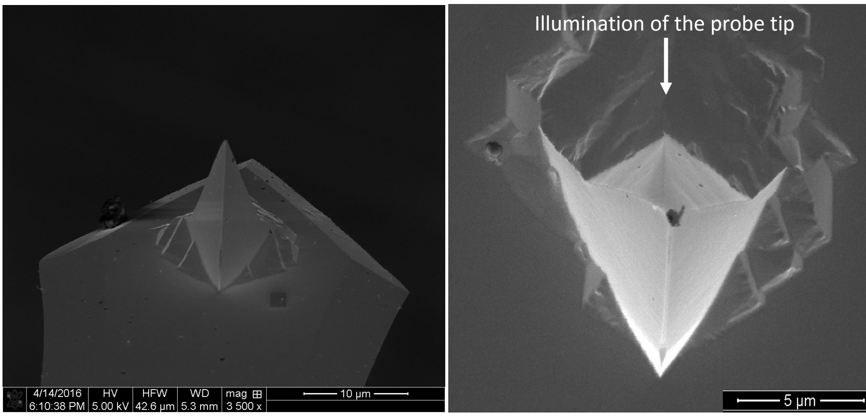

Fig. 1: SEM images of a probe tip. Left side: side view, right side: top view. In both images, the front of the shaft, onto which the laser radiation impinges, faces upwards. The direction of the laser illumination is indicated by a white arrow in the right image.

Supplemental B: Reproducibility of the measurements

The s-SNOM image shown in Fig. 1(b) of the main text has been reproduced with other similar samples (again on ITO float glass substrate). In Fig. 2 of this supplemental note we present such measurements registered with demodulation at the second harmonic (a) and the third harmonic (b) of the tip's oscillation frequency Ω. The sample differs from that described in the main text in that the window contains only an indium tin oxide (ITO) layer instead of an ITO layer with an additional nano-ring array on top. The features discussed and analyzed in the main text (in particular, the complex structure in the lower right region) are also observed here. The results were again robust upon repetition of the measurements after realignment which shows that they are not background-related artifacts in the sense as described in [1].

We observed similar signatures also with probe tips of the *Arrow* variant from Nanoworld AG. These tips are trapezoidal in shape, with an edge pointing forward and with flat areas sideways. We observed reflective plasmon signatures on our sample when the tip was illuminated from the side.

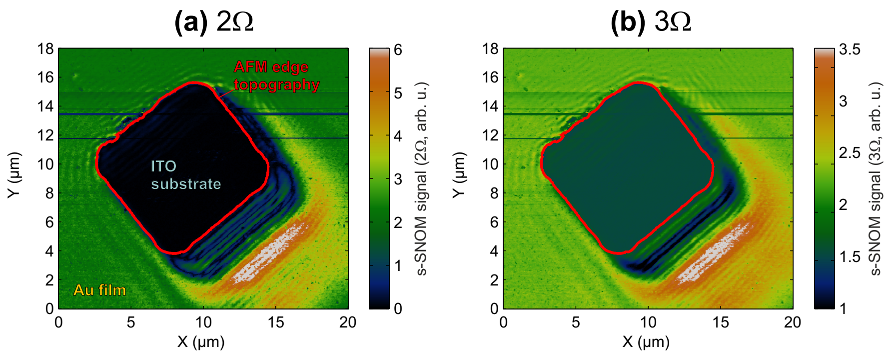

Fig. 2: Additional s-SNOM measurements on a gold frame. Data demodulated at the 2nd harmonic (a), respectively the 3rd harmonic (b) of the tip's oscillation frequency Ω. The measurement geometry is identical to that of Fig. 1(b) of the main text, with the laser light impinging from the upper left.

Supplemental C: Orientational dependence of the tip-induced SPPs

In the main text, we identify the complex fringe pattern close to the metal edge in region 3 of Fig. 1(b) as evidence for two types of tip-induced SPPs. The explanation is based on the way how light is scattered/reflected off the probe tip and onto the sample's surface, as illustrated in Fig. 3(b) of the main text. If the laser beam impinges along the symmetry axis of the cantilever, the intensity of the scattered/reflected light is highest on-axis, see Fig. 3(b). Accordingly, as the scattered/reflected light excites SPP waves, one expects that the s-SNOM signal due to the tip-induced SPPs should be strongest if the laser beam impinges perpendicular to the metal edge, and should become less significant away from this orientation.

This is experimentally confirmed by measurements taken for different values of ϕ, the azimuthal angle-of-incidence of the laser beam relative to the metal edge, as shown in Fig. 3. As a sample, we chose one with a long Au edge (i.e. much larger than the incoming beam diameter) on a soda-lime glass substrate, in order to eliminate any possible contributions from corners. The data exhibit an oscillatory signal both on the glass and the Au film, of which only the latter is of relevance here. As the value of ϕ changes away from 90° (perpendicular incidence), the strongly modulated high-spatial-frequency signal on the Au film and parallel to its edge becomes less pronounced, as expected from the theoretical considerations presented in the main text.

A weak high-frequency modulation remains (indicated by the dashed blue line) which is always perpendicular to the beam and which – in contrast to the tip-induced SPP wave with similar periodicity – does not show a decay of the amplitude with increasing distance from the edge. This is an omnipresent measurement artifact which may be associated with a residual nonlinear (non-sinusoidal) component of the oscillation of the probe tip, allowing interfering signals from the tip and points on the sample to pass the lock-in amplifier and produce the modulation.

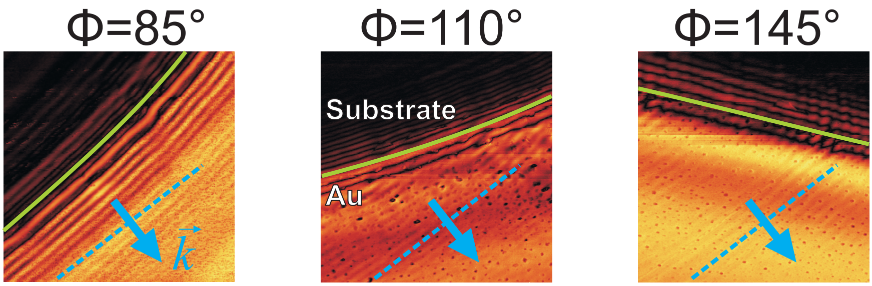

Fig. 3: s-SNOM measurements (signal at 2Ω) for long Au edge vs. azimuthal beam angle ϕ. The blue arrow marks the in-plane wavevector $\vec{k}$ of the incoming laser beam. Dashed blue line: orientation of the spurious interference signal. Green line: position of the metal edge. Note that the edge is straight in reality. The curvature of the edge and of the interference fringes in the figures results from an uncorrected nonlinear response of the piezoelectric translation stage to the applied voltage (the corresponding data in Fig. 1(b) of the main document have been re-calibrated to correct for this nonlinearity.)

Supplemental D: Role of light scattered at the edges

In the main text, when discussing Figs. 1(b) and (c) in Sec. 3, we consider the s-SNOM signal modulations, which manifest as peaks and valleys parallel to the edges of the window in the gold film. We argue that they arise from SPPs generated at or reflected from the edges. We note here that the question arose whether light which does not convert to SPPs at an edge, but scatters directly through free space to the photodiode and interferes there with light coming from the probe tip, could contribute to the formation of the observed modulations parallel to the edges. We expect a negligible modulation contribution. The reason is that the path length traveled by the light waves from the edge to the photodiode continuously changes with the scattering point on the edge, if, as is the case in our experiments, the edge is not parallel to the phase front of the incoming light. These light waves should hence interfere mostly destructively with each other at the photodiode, leaving at most a weak signal for superposition with the light waves coming from the probe tip.

References

[1] Hillenbrand R, Knoll B, Keilmann F. Pure optical contrast in scattering-type scanning near-field microscopy. J Microsc 2001; 202: 77-83.