SURFACE PLASMON PROPAGATION IMAGING BY A PHOTON SCANNING TUNNELING MICROSCOPE

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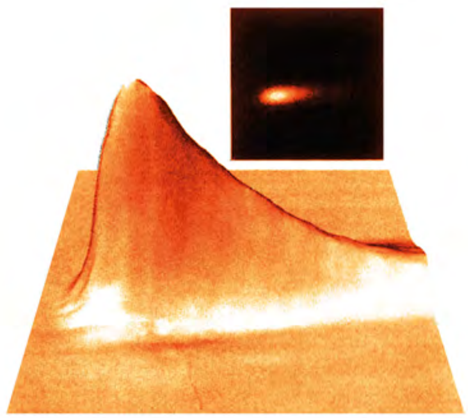

This paper describes the first direct observation of surface plasmon propagation on a metallic thin film deposited on a glass slide, thus facilitating the first direct measurement of the propagation length. This was achieved using photon scanning tunneling microscopy (PSTM). Subsequently, near-field observations of the surface plasmon buried at the metal-glass interface, generated from a discontinuity in the metallic thin film, were reported.

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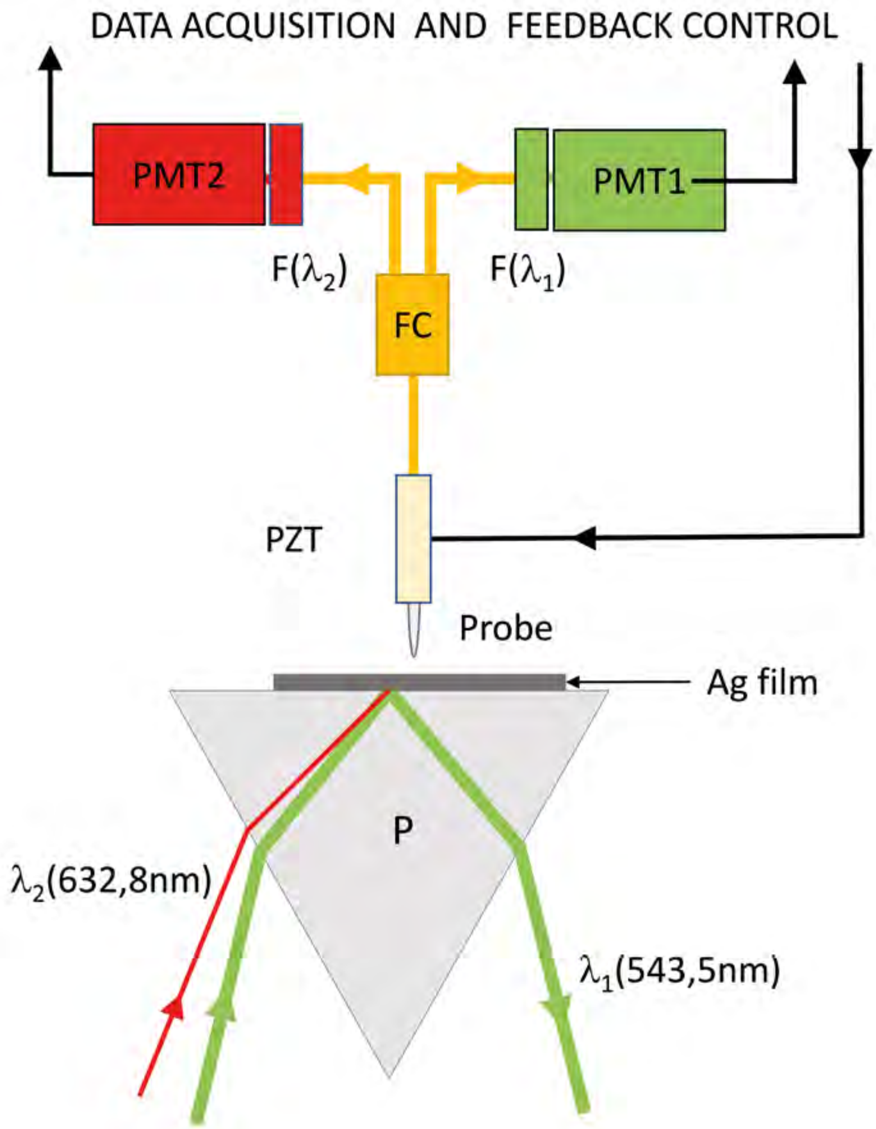
In the 1990's, near-field optical microscopies took off promising to become a unique tool for exploring optical phenomena at the nanoscale. At that time, the photon scanning tunneling microscope (PSTM) [1,2] which was the analog for photons of the electronic scanning tunneling microscope (STM) demonstrated these capabilities. Using this technique, it became possible to measure the optical near field in the vicinity of subwavelength structures illuminated under various conditions. Among them, the PSTM has proven to be a powerful tool for the study and the analysis of surface plasmons which are resonant electromagnetic modes associated with the collective oscillation of an electron plasma at the interface between a metal and a dielectric [3]. Indeed, for a thin film of silver deposited on glass, the metal-air surface plasmon excited in Kretschmann configuration [4] has an extension in air of a few hundred nanometers in the visible range, making it an ideal candidate for optical near-field measurement with a PSTM. In the seminal paper of Dawson et al [5], the propagation of a surface plasmon was shown for the first time and its damping was directly quantified by viewing its propagation length. These results were further completed by the demonstration of the contribution of a buried metal-glass plasmon in the near- field pictures thanks to successive improvement in near-field measurement techniques.

DIRECT OBSERVATION OF THE METAL-AIR SURFACE PLASMON

Figure 1 shows the schematic of the seminal experiment performed at the University of Bourgogne. A 50 nm thin silver film was illuminated by two Helium:Neon lasers in oblique incidence. A first, green one at λ_1=543,5nm was unfocussed to yield a spot size on mm^2 scale on the silver film while the second one at λ_2 = 632.8 nm was tightly focused into a ~10 µm diameter spot at the silver film. The angle of incidence of the red beam was chosen so that it excited the silver-air surface plasmon by phase matching the parallel (to the sample interface) component of wave-vector of the incident light to that of the surface plasmon. In plasmonics this phase matching technique is well known as the Kretschmann configuration [4]. The angle of incidence of the green laser was likewise chosen to excite ● ● ●

Figure 1. Schematic of the experiment: a silver film evaporated on a glass prism was illuminated by two lasers. The green one acts as a pilot for scanning a tapered optical fiber in the near-field of the silver film while the red one excites the silver-air surface plasmon in Kretschmann configuration.



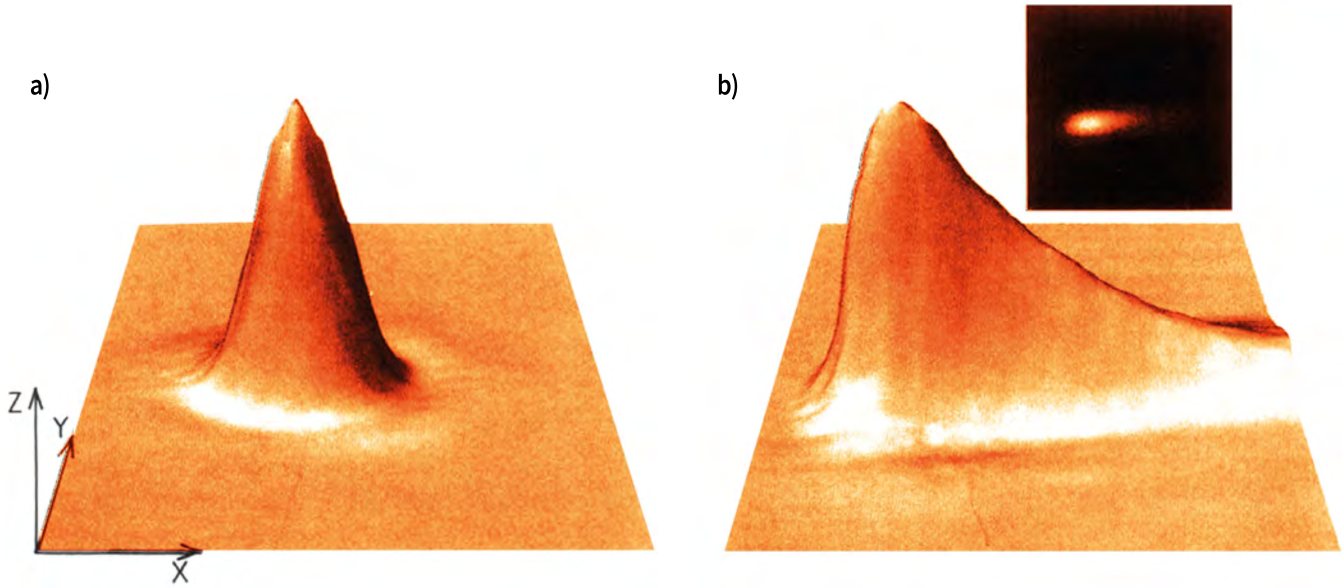


Figure 2. a) PSTM image of the optical near field intensity of the incident beam, λ_1 = 543.5 nm, on a simple prism illuminated under total internal reflection. b) PSTM image recorded at λ_2 = 632.8 nm at the surface plasmon excitation angle for red light on a section of the same prism covered by a 50nm thin silver film. The right side of the image displays a clear exponentially decaying tail explicitly demonstrating the propagation and damping of the surface plasmon. All images are 40 µm x 40 µm.

surface plasmons at the silver-air interface at that wavelength. An optical probe, obtained by tapering an optical fiber, was brought into the optical near field of the metal film in order to 'collect the surface waves' (i.e. by scattering the surface-bound optical field of the surface plasmons to photons travelling up the tapered optical fiber) while scanning the metal film with help of a home-made stack of piezoelectric tubes.

In this configuration, the near-field probe simultaneously detects signals coming from the two lasers at 632.8 nm and 543.5 nm. A fiber optic coupler then separates the signal in two arms. On one arm, a spectral filter selected the green laser which acts as a pilot for driving a feed-back loop allowing the probe to scan the surface without touching it. For this reason, the green laser spot is large (mm scale) in order to generate a surface intensity distribution on the topside of the silver film that is (near-) constant on the ~10µm scale of the surface plasmon propagation length. The green signal is used as the input signal for the feedback loop of the piezoelectric scanners to allow for scanning the silver film at a constant height. On the second arm, the red laser exciting the surface plasmon is filtered out and then sent to a photomultiplier while the

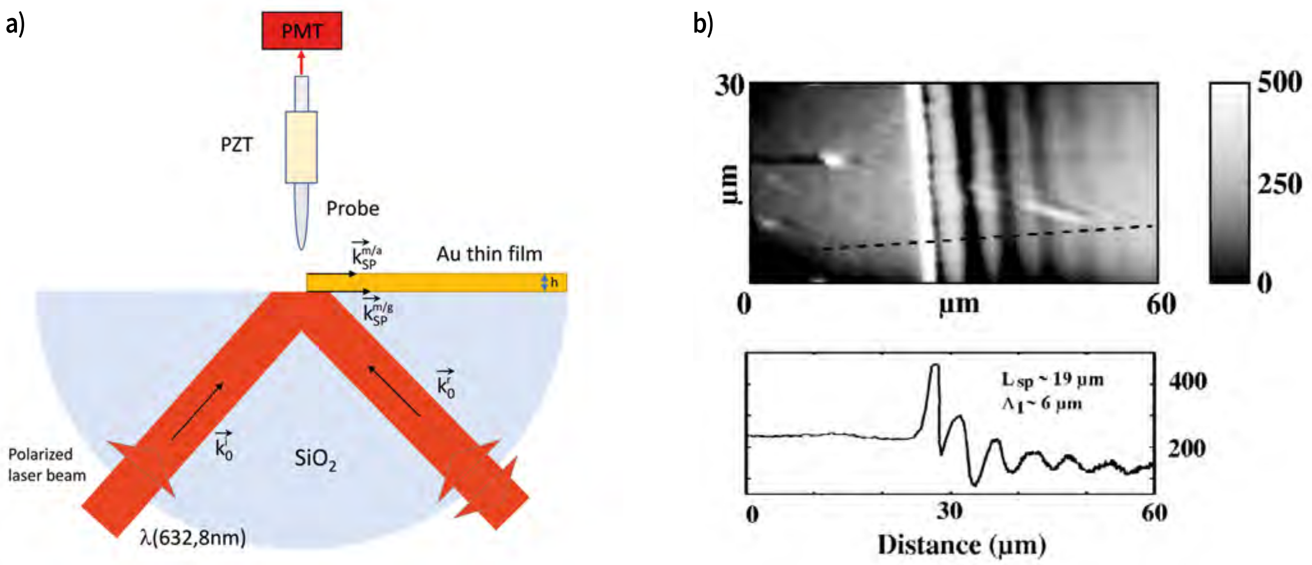


Figure 3. a) Scheme of the experiment. A semi-infinite thin gold film of thickness h=55 nm is vacuum deposited onto a glass prism. The illumination is carried out at various incident angles θ, by a collimated laser beam preliminary linearly polarized. The optical signal is collected via the probe and a photomultiplier tube (PMT). b) PSTM image operating in a constant intensity mode for an angle of incidence of 52°

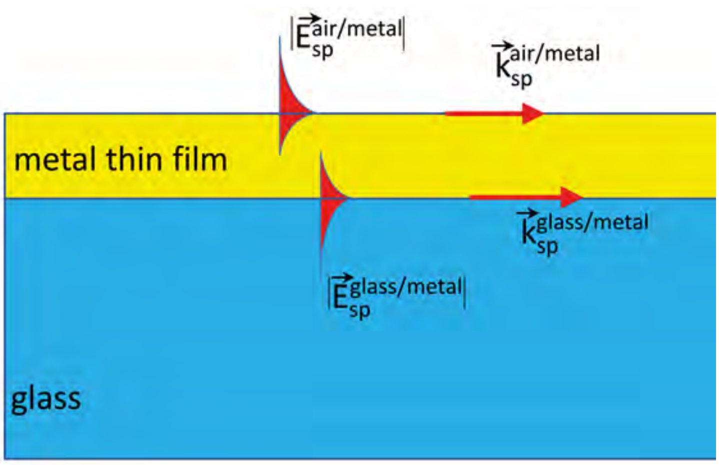
This was the first direct observation of the propagation of a surface plasmon on top of a metal film by near- field imaging paving the way to many other achievements and realizations in this field and development of commercial near-field microscopes for studying the optical near-field at nanoscale.

probe is scanning the surface. At 632.8 nm, the typical images reported in [5] are displayed in Fig. 2. It shows the original near-field pictures obtained in the same conditions on a section of the glass slide without the silver film (Fig.2a) and with the silver film (Fig.2b). Without the silver film, the measured laser spot is approximately circular with a ~ 10 µm diameter, while with the silver film the right side of the imaged spot appears elongated with an exponential decay directly mapping the surface plasmon excitation, propagation and attenuation.

From the image of Fig. 2b) the propagation length was measured to be 13.6µm, which is less than that predicted for a thin silver film of this thickness. The difference was attributed to re-radiation of the surface plasmon back into the prism as it propagates, contamination of the silver surface with a thin, optically •••

SURFACE PLASMON POLARITON

A surface plasmon polariton (SPP) is an electromagnetic excitation that propagates in a wave like fashion along the planar interface between a metal and a dielectric medium. Thus, a SPP is a surface electromagnetic wave, whose electromagnetic field is confined to the near vicinity and whose amplitude decays exponentially with increasing distance into each medium from the interface of the dielectric–metal interface. For a film deposited on glass, plasmons at each interface can exist.



dissipative silver sulphide layer and a possible angular shift between the theoretical and actual angle of incidence. This was the first direct observation of the propagation of a surface plasmon on top of a metal film by near- field imaging paving the way to many other achievements and realizations in this field and the development of commercial near-field microscopes for studying the optical near-field at nanoscale.

However a metallic thin film has two interfaces and further studies were then conducted to explore the surface plasmon buried at the metal-glass interface, which is not excited in Kretschmann configuration.

BURIED SURFACE PLASMON OBSERVATION

In this second experiment, a semi-infinite metallic film was deposited on glass prism as shown in Fig. 3. It is noteworthy that here, gold was preferred to silver to avoid sulfurization. Under collimated illumination, the edge of the metal film scatters the incident laser and generates both the metal-air and the metal-glass surface plasmons.

First measurements in this configuration were reported in [6] which was employing the same laser for exciting surface plasmons as well as for driving the piezoelectronic scanners through a feedback loop. In this configuration, the height of the probe during the scan was modulated by the detected light intensity. Recorded near-field images showed both an exponential decay related to the propagation and damping of the gold:air surface plasmon and but also the presence of oscillations whose period corresponds to an interference between the incident light and the gold:air plasmon. These results were in very good agreement with numerical simulations. However, these latter also predicted that interferences between the glass-to-metal plasmon and the incident beam should also be visible in the measurements but was not observed experimentally. This was finally achieved by Brissinger *et al.* [7]

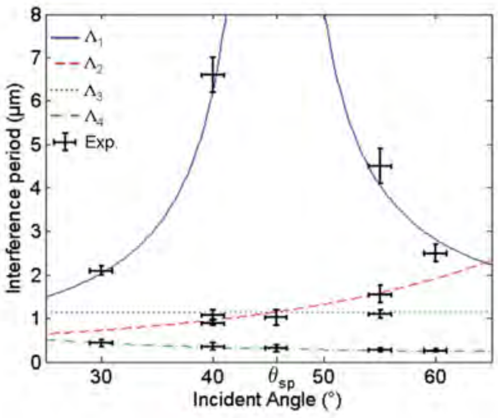


Figure 4. IInterference periods Λ_i in near-field images obtained in the experiment described in Fig.3a by varying the angle of incidence. Λ_1 and Λ_2 are associated with the interferences of the incident wave and the plasmonic waves, respectively at the m/a and the m/g interfaces. Λ_3 is associated with the interferences between both surface plasmon waves and Λ_4 between the incident wave and the back reflection at the prism/air interface. The solid and dotted lines are the values from the numerical simulations and the crosses are recovered from the near-field optical images.

If the photon scanning tunneling microscope in constant optical intensity mode is no longer used, it has nevertheless allowed the direct and first imaging of the surface plasmon excited at oblique incidence in Kretschmann configuration.

many years after, benefiting from a shear-force feedback for driving the near-field probe with an improved reliability.

At that point, the angle of incidence was tuned from 30° to 60° and both surface plasmons were finally visible in the near-field images. Interferences were in very good agreement with numerical predictions. It should be noted that the wavector of the buried surface plasmon, that of the metal: glass interface, was recovered experimentally (1.63±0.08) ω/c quantitatively compared to its theoretical value of 1.59ω/c.

CONCLUSION

If the photon scanning tunneling microscope in constant optical intensity mode is no longer used, it has nevertheless allowed the direct and first imaging of the surface plasmon excited at oblique incidence in Kretschmann configuration As such these experiments have paved the way to the emergence of plasmonics as an entire field of research which has enabled applications in optoelectronics, medicine, chemistry, or sensing to cite just a few. In addition, current improvements in the stability of feedback loops, piezoelectric scanners, nanotechnologies used for shaping near-field probes, as well as recent developments in laser technologies have allowed the near-field microscope to move beyond the walls of the laboratory and to become a standard imaging tool for materials science.

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COMPLETE SOLUTIONS FOR QUANTUM APPLICATIONS

Menlo Systems' optical frequency combs and ultra-stable lasers enable the second quantum revolution

Quantum technology lets us exploit the laws of quantum mechanics for tasks like communication, computation, simulation, or sensing and metrology. As the second quantum revolution is ongoing, we expect to see the first novel quantum devices replace classical devices due to their superior performance.

There is a strong impetus to transform quantum technologies from fundamental research into a broadly accessible standard. Quantum communication promises a future with absolute security through quantum key distribution; quantum simulators and computers can perform calculations in seconds where the world's most powerful supercomputers would require decades; quantum technologies enable advanced medical imaging techniques. Further applications will likely arise that we cannot anticipate yet. The global market has realized the huge potential of quantum technologies. Menlo Systems, a pioneer in the field, provides commercial solutions for these novel challenges.

The link between photonics and quantum physics is obvious. Quantum simulation and computation use cold atoms and ions as qubits, labs worldwide use optical frequency combs and ultra-stable lasers in these types of experiments. Quantum communication often relies on single photons, which are generated with precisely synchronized femtosecond laser pulses in the near-infrared (-IR) spectral range. Quantum sensing and metrology require the highest stability and accuracy in frequency comb and laser technology. And – an application worth highlighting – optical atomic clocks are under way to replace the current definition of the second in the International System of Units (SI).

The transition frequency in optical clocks is on the order of hundreds of terahertz, corresponding to the visible or the ultraviolet region of the electromagnetic spectrum. Counting these optical frequencies is only possible using a frequency comb [1], a mode-locked laser with evenly spaced frequency modes within its optical spectrum. When referenced to a CW laser which is stabilized to a high-finesse optical cavity, the bandwidth of the comb lines narrows down to below 1 Hertz [2], corresponding to a stability of 10^{-15} or better. The newest generation of optical atomic clocks enabled by this technology reach an accuracy of 10^{-18} [3], two orders of magnitude higher than the best cesium atomic clock.

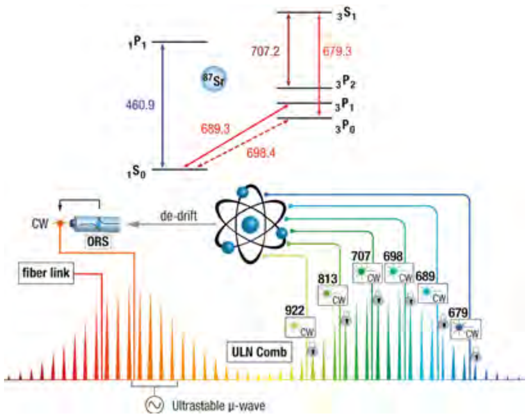

Figure: The hyperfine transitions in strontium (Sr) atoms with the ultra-narrow clock transition at 698.4 nm (upper part). A commercial FC1500-Quantum system for optical clock applications contains an ultra-stable laser transferring its spectral purity onto an optical frequency comb and all other lasers which are also locked to the comb (lower part).

Menlo Systems' FC1500-Quantum is a complete CW laser system with an ultra-stable frequency comb. It provides several CW lasers for atom cooling, repumping, and addressing the sub-Hertz linewidth clock transition in atoms or ions used in optical clocks (see figure). The low phase noise obtained on the comb-disciplined CW lasers is essential for coherent gate manipulation in many atom optical quantum computing schemes and for fast and high-fidelity gate operations. The laser light is delivered *via* optical fiber to the "physics package" consisting of vacuum chamber with all the necessary optics and electronics components and the atoms. Labs no longer need to undergo the time consuming process of designing and building their own ultra-stable lasers. Eventually, the comb itself has two purposes: It acts as reference for all the lasers to maintain their narrow linewidth, and it is the clockwork that transfers the optical frequency's spectral purity to the microwave region, or to a different optical frequency [4].

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