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Introduction:

Every time a phone-call is made, a stream of electrons (current) is launched across a wire or an antenna. Our voice is encoded in the flow of small charged particles. Similarly, electronic signals for computing, sensing or any modern application, are carried by electronic currents or radiated electromagnetic fields.

In recent years, the ability to manipulate and guide light via optical devices such as lasers, leds, optoelectronic modulators, optical fibers and waveguides, allowed to encode our voice, computer signals and other sources of information in streams of photons. Using photons instead of electrons offers huge advantages in terms of speed of telecommunications, precision in sensing and overall power consumption for general signal processing applications.

Key to all of these technologies, electronic or optical, is miniaturization. This is the process that made our smartphones fit our pockets, yet containing billions of transistors. To set a scale, the first transistor built right after world war two was few cm in size. An Iphone containing 10⁹ of those would require a pocket few millions of km deep, out of the reach even for a good-sized dinosaur.

This text is not meant as a scientific manuscript and therefore lacks proper referencing. More information, including well referenced articles and review articles, can be found on lumes.epfl.ch. If necessary I would also be happy to guide any reader to more appropriate scientific literature. Furthermore, in the discussion many concepts are introduced in a non-rigorous way, and should thus be used to replace a proper scientific discussion. Most importantly this text inherently fails to acknowledge the crucial contribution of a large number of excellent scientist working in the field. This in no way reflects any lack of appreciation for the great work many people have done.



Fig.1 Left: pictorial representation of a Surface Plasmon Polariton (SPP). The blue lines indicate the electromagnetic field sustained by the charge distribution accumulated at the surface. In the small graph, the spatial confinement of such an SPP field is highlighted. **Right:** when traveling SPPs reflect from the edges of a nanostructure, such as a nanowire, a standing-wave can be created; this is referred to as a plasma resonance

Despite this success, modern technology is reaching a limit in miniaturization, not necessarily due to limitations in the fabrication processes, but rather in the physics governing the transport of electrons or photons at such confined scale.

In particular, despite its above-mentioned advantageous properties for signal processing, light as we know it cannot be confined in spaces smaller then its wavelength (color), and travels only in straight trajectories. These are severe limitations for miniaturization as guiding signals in small spaces requires sharp corners and small waveguides.

For this reason, a form of light termed surface plasmon polariton (SPP) gained considerable attention in the last years. SPPs are an electromagnetic field bound to a charge distribution located at the interface between a metal and a dielectric, see Fig. 1 left panel. Because of this binding, SPPs can propagate on curved surfaces, follow sharp corners and they can be confined in spaces smaller then their wavelength. A great deal of research nowadays is dedicated to develop new plasmonic devices, analog to the conventional electronic or optical ones, for exploiting SPPs in ultra-miniaturized circuits. Engineering these EM fields and their properties requires nano-fabrication abilities, but also



Fig. 2: Schematics of an ultrafast TEM, see text.

techniques that can characterize their behavior once implemented. This proved extraordinarily hard to do because a combined nm (10^{-9} m) and fs (10^{-15} sec) space and time resolution are needed to capture their static as well as dynamical properties. The work presented here describes a novel methodology, enabled by transmission electron microscopy (TEM), which allows to capture fs-nm snapshots of SPPs confined in nano-sized spaces.

Surface plasmons polaritons and plasma resonances

When an SPP is trapped on the surface of a nanostructure, such as a nanowire in the example depicted in the right panel of Fig. 1, it can bounce back and forth from the edges of the structure itself creating a so called standing-wave. The distance between the crests of such a wave depends on how many integer spatial periodicities of the SPP can fit between the edges of the nanostructures, and the different resulting field intensity distributions (or modes) are called plasma resonances, see right panel of Fig. 1.

Taking static images of SPPs and plasma resonances has been demonstrated via different techniques. In a special optical microscope the tip of an optical fiber is scanned on the surface of a nanostructure to capture the radiative component of the SPP and map its spatial profile. In Transmission Electron Microscopy instead, a beam of electrons is directed to the nanostructure and their trajectories, modified by the interaction with the SPP field, are mapped onto a special detector able to perform both spectroscopy and imaging. The advantage of this approach is that the spatial resolution obtained using high energy electrons (200 KV) is much better then what can be done with optical methods, reaching the 1 nm regime. Other techniques are also available, although less frequently used; the discussion of these is beyond the scope of this article.

Photon-Induced near field electron microscopy (PINEM)

As we anticipated above, the aim of modern technology is to capture the dynamical evolution of SPPs with a combined spatial and temporal resolution of nm and fs. To achieve these performances, the abovementioned techniques need to be extended to real-time recorders. In recent years, the technique of time-resolved transmission electron microscopy has been developed. A scheme of the experimental apparatus is depicted in Fig. 2. A TEM is modified to allow access to optical beams generated by an ultrafast laser. One is used to generate a train of photoelectron pulses from a cathode (violet beam), and a second one is used to photo-excite a specimen (blue beam). The delay between the pulses in these beams is controlled via a motorized delay stage and image or spectra of a sample under test can be obtained for different values of such a delay in a stroboscopic fashion.

When light excitation is directed to a nanostructure, for example a nanowire, Fig. 3a, SPPs can be induced on the surface of the wire. Their multiple reflections from the edges of the wire lead to the formation of a plasma resonance on the wire that can interact with the pulsed electron beam.

In an ultrafast TEM, it is possible to image the electrons that interacted with plasma resonances, even when these are buried at an interface between different materials. As depicted in Fig. 3 B and C, one can thus obtain snapshots of plasma resonances bound to a single nanowire (Fig. 3

B) or radiating from nano-cavities in a film and forming spatial interference patterns (Fig. 3 C).



Fig. 3 A: Schematics of the photoexcitation of SPPs on a nanostructure. B: ultrafast TEM image of a plasma resonance on a single nanowire. C: ultrafast TEM image of the plasmon-plasmon interference generated by an array of nano-antennas. D: single frame of a movie capturing the propagation of SPPs in a thin film once they are radiated by the antenna array at the top of the image. E: Temporal evolution of the SPP propagation

By varying the delay between the photoexciting pulses and the imaging electron pulses, different time-points of the temporal evolution of the SPPs can be recorded. For example, in a recent experiment, we fabricated an array of nano-antennas at the interface between a silver thin film and a SiN_4 membrane. The antennas were lit up by photoexciting SPPs via light pulses (Fig. 3 D) and their propagation and interference was filmed by

recording their spatial distribution at different time-delays, see Fig. 3 D and E.

Outlook

These waves known as "surface plasmons" will be useful in telecommunications and future computers, where data will be transported between processors using light instead of electrons. Aside from being more energy-efficient, these processors could reach the nanoscale size and enable to build high-resolution sensors and ultrasmall signal processing systems. To achieve these performances however, these new systems need to be built from stacking different layers of advanced materials and, so far, no technique can map the guided light as it moves across interfaces. Trying to see SPPs in these interfaces buried between layers is like trying to capture events happening in a house from the outside. A regular camera won't work; but a microwave or a similar energy-tracking imaging device does the trick and sees through walls. Similarly, our experiments enable the fs-nm resolved characterization of SPPs in nanostructures and at buried interfaces, providing a unique tool for observing and manipulating light at the nanoscale.