Real-space imaging of nanoplasmonic resonances

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Resonant nanoplasmonic structures have long been recognized for their unique applications in subwavelength control of light for enhanced transmission, focussing, field confinement, decay rate management, *etc*. Increasingly, they are also integrated in electro-optical analytical sensors, shrinking the active volume while at the same time improving sensitivity and specificity. The microscopic imaging of resonances in such structures and also their dynamic variations has seen dramatic advances in recent years. In this Minireview we outline the current status of this rapidly evolving field, discussing both optical and electron microscopy approaches, the limiting issues in spatial resolution and data interpretation, the quantities that can be recorded, as well as the growing importance of time-resolving methods.

1. Introduction

Plasmons are prominent members of the family of quasiparticle eigenstates found in coupled crystal-radiation quantum systems. Hopfield coined the generic term "polaritons"[1] for these quasiparticles. Surface plasmon polaritons (SPPs), in particular, are the quanta of coupled oscillations of photons and an incompressible electron gas (or Fermi liquid) that is confined to the interior volume of metallic structures surrounded by dielectric media or vacuum. SPPs have drawn increasing interest in recent years, because modern manufacturing technologies continue to enable many novel ways to put to service their unique features. Foremost, perhaps, SPPs are recognized for their intense electromagnetic fields that are bound to the metal-dielectric interface. Manufacturing structural confinement in the other two spatial dimensions enables further field enhancements, accompanied by "focussing" effects of the optical part of SPPs onto subwavelength scales.

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Being eigenmodes of the coupled electro-photonic system, their interaction with individual electrons or photons can exhibit strongly resonant character in any parameter dimension like energy, size, direction, or material composition. Beyond mere field enhancement, SPP resonances play an increasingly important role in sensor applications, as building blocks in metamaterials, in nanomechanical manipulations, as nanoscopic waveguides, as nano-optical antennas for the management of emission/reception directivity and efficiency, *etc*. Last not least, the quantum nature of SPP resonances represents a largely untapped reservoir of opportunities.

This Minireview highlights the recent advancements in microscopic techniques that facilitate imaging of resonant surface-plasmons in the spatial and increasingly also the temporal domain. As guiding principle for structuring the discussion we select the nature of the probing particles that interact with plasmonic structures. Fig. 1 provides a graphical summary of currently established microscopy techniques. Grouping them according to their use of photons/electrons for excitation/detection results in a natural division of the methodological landscape into four quadrants, which we discuss in sequence below.

2. Photon-in/photon-out methods

Farfield optical techniques

Photon-plasmon coupling was one of the first mechanisms used to study plasmon polaritons. The well-known wavevector selection rules for infinite planar interface plasmons have been a textbook subject for many years. For finite plasmonic resonators, however, conventional farfield optical microscopy by itself is often of limited use, because the relevant structure sizes are typically well below the diffraction limit. Consequently, confocal microscopy studies are applied mainly to extended structures like micron-sized plasmonic rings[2] or wires.[3] Notable exceptions in recent years are subwavelength structures that may act as so-called nano-optical antennas. Being designed for specific applications, their man-made physical dimensions are usually known independently, and one is chiefly interested in characterizing their spectral, polarization, or emission properties in real[4–6] or reciprocal space.[7] An intriguing inelastic approach to the mapping of plasmonic modes is the introduction of erbium atoms into plasmonic structures as local field strength indicators. Their upconversion luminescence has been used successfully in tapered plasmonic waveguides and subwavelength aperture arrays.[8,9]

Non-linear optical effects are frequently employed to enhance the spatial resolution of diffraction limited microscopy with an added advantage of considerable background suppression. Two-photon induced luminescence (TPL) is a prominent and well-established example, capable of exciting site-specific processes in, for example, composite gold nanoantennas.[10–12] This results in a wavelength-dependent real-space mapping that corresponds to the local $|\mathbf{E}|^4$ distribution for nanoantenna modes – convoluted with the diffraction-limited optical farfield point spread function (PSF) of the microscope.

Nearfield optical techniques

To overcome the farfield resolution limits, several variants of nearfield optical microscopy have been developed since the 1980s. They all require penetration of the sample's nearfield zone by a physical optical probe, which is typically held a few nanometres above the surface by atomic force or electronic tunnelling feedback. Images are acquired in a sequential, scanning manner. In nanoplasmonic microscopy, the probe facilitates the coupling (*i.e.*, scattering) of interface-bound nearfield optical modes with a farfield mode spectrum that can propagate to a conventional optical detector, either *via* wave guide or free-space.

The photon scanning tunnelling microscope (**PSTM**) is actually not an "STM" but an AFM technique that employs a bare, tapered optical fibre to pick up nearfield optical signal and also guide it. It has a relatively low spatial resolution and in nanoplasmonics it is mostly used to study resonant cavities, wave guides, and other extended surface plasmonic structures.[15–21]

Better spatial resolution is achieved with subwavelength aperture probes – *e.g.*, metal-coated optical glass fibre tapers or hollowed AFM pyramidal tips. These kinds of techniques are commonly referred to as scanning nearfield optical microscopy (**SNOM** or **NSOM**). Their lateral resolution is limited to approximately the inner aperture diameter plus twice the optical penetration depth in the outer material. Only few attempts have been made to improve resolution by numerical post-processing,[22]

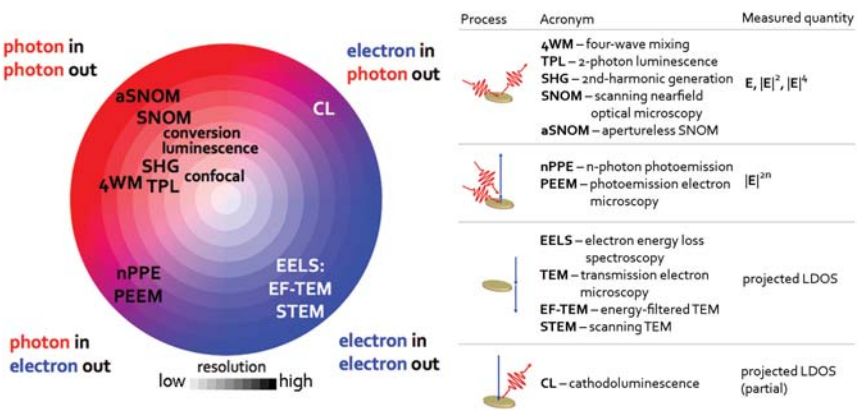

Fig. 1 Experimental techniques for real space imaging of nanoplasmonic resonances, schematically arranged according to incident/emitted particles, together with simplified depiction of the relevant process and the observed quantity.

chiefly because of a notorious aspect of all nearfield-probes: their possibly distorting influence on sample fields.[23,24] Its utilization as a desired effect is a rare exception in SNOM studies.[25] Usually, one tries to reduce this artefact by retracting the probe somewhat from the sample. An improving image fidelity has to be weighed against degrading signal-to-noise ratio and diminishing lateral resolution. Typically, only resonant field modes extend far and strongly enough into space to allow sufficient probe-sample separation.[26–29] On the plus side, SNOM readily offers spectroscopic capabilities,[30–34] the discrimination of two orthogonal polarization states[35–37] and time-resolved measurements.[38] With a careful preparation and independent characterization of the detecting probe it might become feasible to study unknown samples.[39] SNOM has also been successfully combined with TPL – valuable reviews of this approach have been published recently[40–42] – and second harmonic generation (SHG) in nano-ellipsoids and nanobars.[43–46]

To push the spatial resolution down to less than 10 nm, apertureless SNOM (**aSNOM** or **aNSOM**) uses point- or needle-like probes, whose apex radius largely determines the achievable lateral resolution,[47] essentially independent of wavelength. aSNOM is increasingly being applied to individual nano-plasmonic structures like disks,[13,23] holes,[48] triangles,[49] linear wire-antennas[14,50–52] (Fig. 2). Another interesting application are coupled nanoparticles,[53] for which plasmon mode hybridization is expected. Although very much possible, aSNOM is not frequently combined with non-linear optical effects in nano-plasmonics – a notable recent exception is a four-wave mixing study at coupled Au colloids.[54] As in farfield microscopy, inelastic fluorescence emission can also be used for imaging purposes. A properly suited particle, attached to an AFM tip, is readily able to record field intensity maps even three-dimensionally, as has been demonstrated above nanoslits.[55–57]

Like SNOM, aSNOM suffers from parasitic probe-sample coupling effects.[23,24] It has been shown in the past that replacing strongly scattering probes with weakly scattering probes (*e.g.*, nanotubes attached to regular AFM tips) can remedy this problem, but this might not be the easiest route to a routine, inexpensive method. An exciting alternative has been found in polarization controlled aSNOM.[13,14,23,51,58–60] Here, two optical states of a standard AFM probe are used with orthogonal polarization for excitation and detection. Thus, any spill-over signal is due to local polarization rotating effects at the sample. This allows, in effect, to map out particle plasmons (specifically, certain electric field vector components) with very good fidelity. *Ad hoc*, this is verified by the favourable comparison with simulations of the bare sample in the absence of any probe.[13,14,51]

3. Photon-in/electron-out methods

Photoemission (PE) of electrons (or "photo-electric effect") is not only one of the pillars of quantum-physics. In combination with modern electron microscopy (**PEEM**), it is also rapidly developing into a powerful approach to study nanoplasmonic nearfield excitations in both space and time.[61–65]

Its spatial resolution does not depend on the incident electromagnetic light-field but on the electron detection optics. The low kinetic energy of emitted electrons and specific, sample topography related artefacts impose a limit of some tens of nanometres,[61,64,66] with an (electron-) energy resolution of currently ~50 meV.[66]

Pulsed, multi-photon excitation, most notably two-photon photoemission (2PPE), has added options for time-resolved (TR) measurements[62–65,67] to allow direct mapping of the dynamics in nanoplasmonic systems in space and time. The temporal resolution largely depends on the width of the optical excitation pulses, reaching values well below a femtosecond. Adaptive shaping of the laser pulses opened another parameter space for control of PEEM experiments that has yet to be fully explored[61] (Fig. 3). Being a nonlinear, two-photon process, the sensitivity of 2PPE-PEEM is proportional to the time-integrated $|\mathbf{E}|^4$ of the local electric-field amplitude. Recent application of PEEM include localized and propagating SPPs in silver gratings[62,63] and wires of variable length,[68,69] gold wires,[70,71] and the identification of "hot spot" formation in symmetry broken samples.[72]

4. Electron-in/electron-out methods

Electron energy-loss spectroscopy (**EELS**) is similar in some ways to optical Raman spectroscopy. The quantum of energy

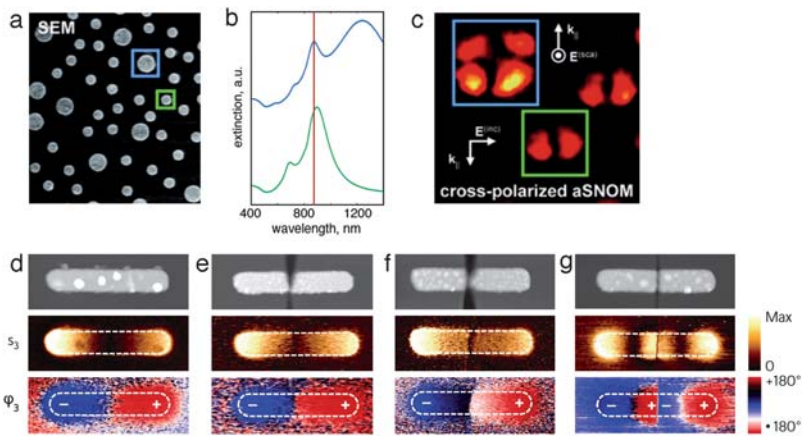

Fig. 2 Upper panel: topography, farfield spectra and nearfield optical images of two species of gold nanodisks (adapted from ref. 13 with permission of American Chemical Society, Copyright 2008). Lower panel: topography, optical field strength and phase of gold nanorod antennas, progressively being cut through at the centre (reprinted from ref. 14 by permission of Macmillan Publishers Ltd: Nature Photonics, Copyright 2009).

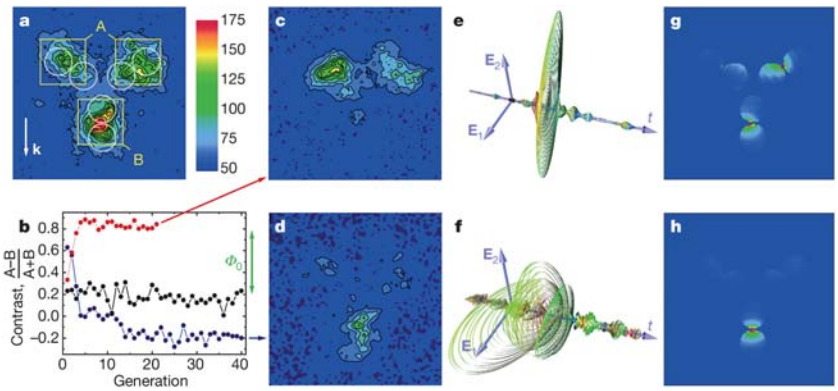

Fig. 3 PEEM monitoring of adaptive optimization of the controlled plasmonic response of a star-shaped nanostructure: (a) PEEM image (1.13 × 1.13 μm^2) for p-polarized excitation (b) optimization tracks, (c,e,g) predominant photoemission from region A, (d,f,h) predominant photoemission from region B (reprinted from ref. 61 by permission of Macmillan Publishers Ltd: Nature, Copyright 2007).

loss is the spectroscopic signature of an eigenmode being excited. Since the corresponding cross sections are many orders of magnitude smaller, the desired inelastic spectroscopic signatures ride on a huge instrumental background due to the elastic "zero-loss" (ZLP) or "Rayleigh"-peak. This is particularly troublesome in imaging techniques such as the combination of EELS with transmission electron microscope (**TEM**).

Thermal electron sources provide high currents, but have a ZLP width of at least 0.6 eV. In scanning TEM (**STEM**), therefore, one usually prefers cold field emitters to reduce the ZLP to ~0.3 eV. STEM-EELS provides access to surface plasmons above ~1.5 eV, as was demonstrated with plasmonic nanotriangles[73] (Fig. 4), and with coupled nanorods, spheres, and ellipsoids.[74–76] An alternative way to suppress the ZLP background combines thermal emitters with electron energy filtering TEM (**EFTEM**). It gained much appeal in the context of SPP microscopy a few years ago with the introduction of efficient, yet sharply discriminating electron monochromators for energy filtering, resulting in better than ~50 meV spectral width at ~200 keV kinetic energy. EFTEM has been applied down to SPP energies of ~0.5 eV in isolated and coupled nanowires,[77] and elongated nanoparticles,[78] triangular nanoprisms,[79] and even nanoholes in metallic films.[80]

In both STEM-EELS and EFTEM a "hyper-spectral" data cube (two spatial image dimensions and one spectral dimension) is obtained: STEM-EELS collects one-dimensional spectra sequentially from different sample regions, whereas EFTEM records two-dimensional images sequentially for different energy losses. Typically, the dimensional advantage makes EFTEM the faster technique, but STEM-EELS offers superior spectral sampling.

The details of the theoretical interpretation of experimentally acquired EELS maps are somewhat under discussion.[81,82] Being a measure of transition probability, an intimate relation with the (projected) local density of optical states (LDOS) seems a natural and successful description.[81] In general, though, the link between LDOS and EELS seems not to be a direct one-to-one mapping.[82]

Care must be taken also in assessing the spatial resolution in EELS experiments. It is not simply the diameter of the transmitted electron beam, which easily reaches atomic scales. The range of effective interaction with the sample (related to the impact parameter) can be much larger, particularly at low energy

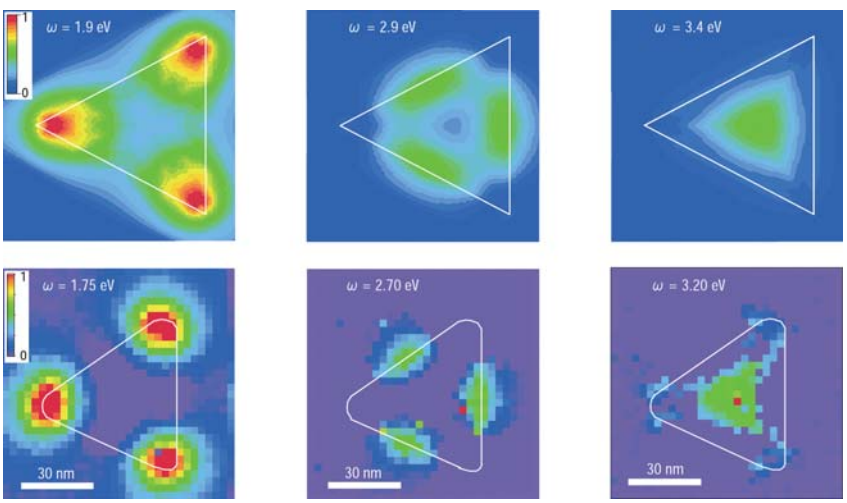

Fig. 4 STEM-EELS images of a triangular silver nanoprism at different loss energies. Upper row: simulated images. Lower row: measured images, after subtraction of the ZLP and Gaussian fitting. (reprinted from ref. 73 by permission of Macmillan Publishers Ltd: Nature Physics, Copyright 2007).

losses.[83] A recent comprehensive review of various resolution degrading effects in EELS experiments[84] estimates (for 100 keV incident energy) the inelastic delocalization due to long-reaching Coulomb forces as ~10 nm at optical energies. It is possible to push the spatial resolution somewhat through the use of additional annular dark field filtering – albeit, at the expense of lower signal-to-noise ratios.[85]

5. Electron-in/photon-out methods

The energy transfer from incident electrons to the plasmonic system can also be observed through subsequently emitted photons, *i.e.*, cathodoluminescence (**CL**). Being an irreversible process, though, it is not related by time-reversal to any of the processes from Section 3. Rather, it provides complementary information more closely related to EELS,[81] as it depends critically on the electron-plasmon interaction step. Its spatial resolution of ~10 nm is mainly given by the scanning electron microscope used for excitation.

Emission patterns and temporal structure (for pulsed excitation) of the emitted radiation are not as frequently studied as its spectroscopic properties. Arguably, it is the combination of optical monochromator resolution (easily below 1 μeV) with multichannel detection (for hyperspectral imaging) that provides the strongest impetus for the application of CL also to metallic nanostructures.[87] E.g., SPP modes and their symmetries have been studied by CL in noble metal wires[86,88] (Fig. 5), and triangular nanoprisms.[89] Intriguing are also the options for integrating CL with nanofabrication techniques. This provides *in situ* design, characterization, and optimization of plasmonic resonator structures such as annular,[90] linear ridge,[91] or two-dimensional Fabry–Pérot resonators.[92]

6. Conclusion and outlook

Not surprisingly, an ideal method for imaging SPPs does not exist; one which offers ultimate resolution in space, time, wavevector, and energy; which yields easy to interpret, complete information; which handles any substrate under any ambient condition. Rather, we are currently witnessing the emergence of specialist techniques that are suitable for specific purposes or sample types.

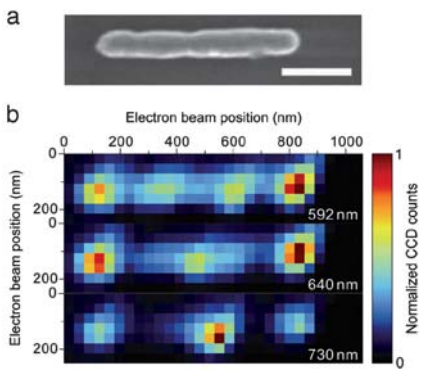

Fig. 5 Cathodoluminescence imaging of a 725 nm long gold nanowire. (a) Scanning electron microscope image; scale bar is 250 nm. (b) CL images of the nanostructure at different detection wavelengths. (reprinted from ref. 86 with permission of American Chemical Society, Copyright 2007).

For instance, only the interferometric detection of elastically scattered photons offers direct access to the optical phase of SPPs. Optical excitation produces maps of some power of the magnitude of coherent sum of all *excited* SPP eigenmodes, projected onto some direction. Electron excitation yields maps closely related to a projection of the local density of optical states, *i.e.*, the incoherent sum over *excitable* SPP eigenmodes.

Even a cursory glance at Fig. 1 reveals which quadrant has been emphasized historically in SPP microscopy. The variety of well-established, all-optical techniques outnumber the methods that use electrons for excitation or detection. In contrast, electron-based microscopy of SPPs is a rapidly evolving field, which promises to populate the other quadrants with exciting new techniques in the near future.

Perhaps the biggest driver of this development is the superior spatial resolution of electron microscopy. However, the overall spatial resolution of any technique is not only determined by the excitation and detection PSF, but also by the interaction volume. Even in STEM-EELS low-energy SPPs cannot be imaged with the atomic scale resolution achievable with elastic electron microscopy or inelastic electron microscopy at higher energy losses.[93] The interaction of an incident electron with SPPs imposes a subtle limit of ~10 nm at visible energies,[84] which is comparable to that of apertureless SNOM.

Optical nearfield methods have been notorious for parasitic coupling effects between probe and object, which often hinder even qualitative signal interpretation. Interestingly, the use of orthogonal probe modes for excitation and detection seems to emerge as a general way to tackle this problem, both in apertureless and aperture SNOM.

Some novel ideas have been proposed on the detection side of SPP imaging. For instance, electron phase retrieval maps have been suggested in holographic electron microscopy,[94] and the direct electrical detection of SPPs[95–97] promises to be highly efficient, though not yet readily applicable to imaging. Even more advances have been made in recent years on the excitation side. Beyond the adaptive temporal control of phase and polarization of single photonic pulses, any multi-particle excitation method offers in principle time-delay options for controlling the excitation and tracking the temporal evolution of desired surface plasmonic processes. An intriguing Fourier-spectroscopic variant of pump–probe techniques has been shown to resolve electromagnetic vector field components of THz-pumped SPP resonances with the spatial resolution of optical probe pulses.[98–104] During the writing of this Minireview, mapping plasmonic resonances with simultaneous electron and photon pulses has also been demonstrated.[105] It allows observation of higher-order EELS processes as well as electron energy gain spectroscopic (**EEGS**) imaging of plasmons.[106]

In summary, it appears that all the most powerful SPP imaging methods are converging on a real-space imaging scale of a few nanometres. There are strong indications of increasing attention to the time domain, which seems to hold the biggest promise for significant technology development in the near future. We expect the ongoing firm establishment of various techniques for real space imaging of SPPs will deepen the understanding of SPP phenomena – with accents also on quantum effects – and lead to sophisticated novel applications.

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