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Photoemission electron microscopy, a tool for plasmonics

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ABSTRACT

A key challenge to plasmonics is the development of experimental tools allowing access to the spatial distribution of the optical near field at the nanometre scale. A recent approach for mapping the near optical field is the use of the photoemission electron microscopy PEEM. Indeed, photoemission can be strongly enhanced upon excitation of surface plasmons. By collecting the photoemitted electrons, two-dimensional intensity maps reflecting the actual distribution of the optical near-field are obtained. In the following a brief overview of the possibilities of the photoemission electron microscopy as a tool for plasmonics is given. Main focuses will be set on experimental results regarding the mapping of the near optical field at nanometer scale, the investigation of the spatio-temporal dynamics of plasmon-polariton waves and the manipulation at will of the near field.

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1. Introduction.

Light-matter interaction at the ultimate scale opens new opportunities for innovative applications. Specifically, when matter is structured at a scale below the natural wavelength of light, light-matter interaction can exhibit a physical behaviour at variance with what is commonly observed at the macroscopic scale. Optics at the subwavelength length scale is today known as nanophotonics. Plasmonics is a major part of nanophotonics dealing with the subwavelength interaction of an electromagnetic wave with the conduction electrons at a metallic/dielectric interface. The main ingredients of plasmonics are *localized surface plasmons* LSP and *surface plasmon polaritons* SPP. LSP modes are at the origin of strong local enhancements of the optical intensity in metallic nanostructures, with enhancement factors as large as several orders of magnitudes compared to the corresponding plane wave at the same frequency. SPP are propagating electromagnetic modes whose spatio-temporal characteristics promise what electronics and photonics individually do not, i.e. highly integrated devices working at mind-blowing (optical) speed. To make it brief, plasmonics is of primary interest for a number of targeted applications in line with low power and sustainable economy such as energy (lighting [1], displaying [2], photovoltaics [3]), sensors [4,5], information technology (data storage [6], integrated guiding and interconnection [7]), biotechnology [8], health (cancer therapy [9]) and new class of optical materials, i.e. metamaterials [10].

One key challenge to the advent of plasmonics is the access to experimental techniques allowing for the investigation of the spatial temporal distribution of the optical near field at submicrometre spatial and femtosecond temporal resolutions. Considering spatial aspect, this issue is currently addressed by *scanning near field optical microscopy* (SNOM) or related methods [11,12]. In respect to the expressed needs, SNOM has three major drawbacks: (i) the spatial resolution is insufficient (30–100 nm); (ii) the measurement proceeds by the insertion of a probe in the close vicinity of the object with possible parasitic tip/object coupling; (iii) making use of probe tips, the measurement reproducibility is low. As for the temporal side, SNOM belongs to the scanning probe technique family inheriting limited temporal investigation capabilities. Alternative methods are still few. For the interested readers, a recent panorama of the real-space microscopic imaging techniques devoted to plasmonics is available [13]. In the following, we will focus on the photoemission electron microscopy (PEEM) as a tool for plasmonics [14].

Nonlinear photoemission can be strongly enhanced upon excitation of surface plasmons. Among pioneering examples [15–22], Stuckless and Moskovits [23] early evidenced orders-of-magnitude enhancement of the two-photon photoemission process from rough silver films over those from smooth annealed films. Within the framework of plasmonics investigations, the photoelectric process to consider is nonlinear in nature and corresponds to a n-photon photoemission (nPPE). Indeed, for noble metals, plasmon energies correspond to near infrared/visible range,

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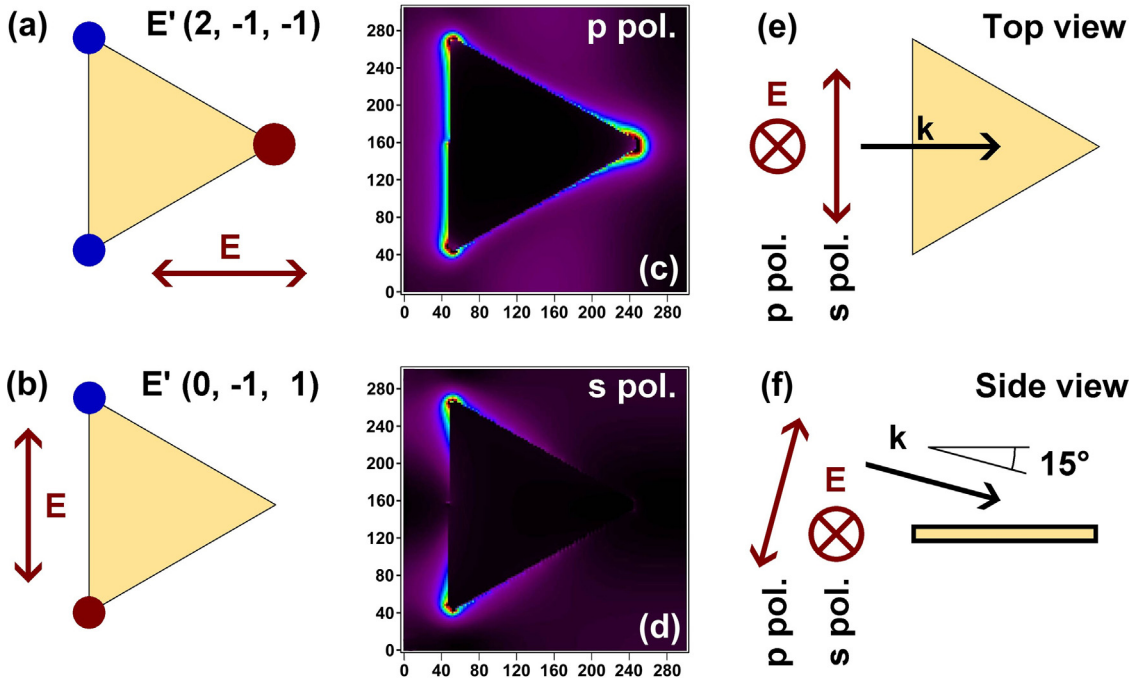

Fig. 1. Dipolar modes. (a and b) Symmetry adapted linear combinations (SALC) corresponding to the two dipolar eigenmodes of a solid triangle of D_{3h} symmetry. The charge patterns within the horizontal σ_h plane exhibit two orthogonal polarization states. (c and d) Maps of the out-of-plane $(E_z)^2$ near field computed by FDTD simulations of the dipolar eigenmodes for p & s light polarizations. In-plane height of the triangle is 200 nm, excitation wavelength 800 nm, light incidence angle 75°. (e and f) Top and side views of the illumination geometry.
Adapted from [36].

i.e. 380 nm (3.26 eV)–1400 nm (0.885 eV), while surface work functions amount to several eV (around 4.5–5.5 eV for Cu, Ag, Au [24,25]). As a consequence, plasmonics investigations conducted by direct linear electron photoemission are nearly impossible [26] and practical studies exploit photoelectric process of nonlinear orders of 2 and/or 3 [27]. The basic underlying mechanism tending the increase of the photoemission yield close to a plasmon energy is an n-step cascade photon absorption process [28], more refined models described it as a plasmon assisted multiphoton emission [22,29–31]. In short, the electron emission signal stems from the near field on the metal side, i.e. the electron reservoir and allows for both bright and dark plasmon mode detection [32]. By collecting the photoemitted electrons, two-dimensional intensity maps reflecting the actual distribution of the optical near-field are obtained. The PEEM imaging technique makes use of well-established electron optics, i.e. involves no physical probe altering the measure [33]. This approach provides full field spectroscopic images with a routine spatial resolution of the order of 20 nm (possibly down to 2 nm with recent electron kinetic energy resolved and aberration corrected instruments [34]). Making use of a true optical excitation scheme (photon in, electrons out), the natural degrees of freedom are the polarization and wavelength of the incident light, nPPE- PEEM give thus access to spatially resolved spectrometric investigations at the single nano-object level. Moreover, this technique exhibits state-of-the-art temporal capabilities inherited from the development of ultrafast optical spectrometries (pump probe experiment) [35].

So far, this technique has proved valuable in the investigation of several plasmonics topics, namely (i) high resolution (i.e. subwavelength length) mapping of the near-optical field of objects/structures of interest under light excitation, (ii) the dynamics of SPPs in space and time at sub micrometer spatial and femtosecond temporal resolution, and (iii) the manipulation of the near-field. through coherent control of the light excitation. In the following paragraphs, illustrative examples taken from author's research work as well as literature are presented.

2. High-resolution mapping of plasmonic modes.

A first characteristic example dealing with high resolution near-field mapping along with simple field manipulation is presented in [36]. The objects under interest correspond to Au equilateral triangles of nanometer edge sides. Two *localized surface plasmon resonances* (LSPR) are studied, namely the in-plane dipolar and quadrupolar plasmon excitations. Experimental field maps are interpreted within the framework of a group theory description and *finite difference time domain* (FDTD) simulations. Starting with the ($\ell = 1$) dipolar mode of E' symmetry, its irreducible representation is of degeneracy 2, so the physical charge distributions at resonance correspond to linear combinations of a pair of two eigenvectors. Both eigenvectors obtained as symmetry adapted linear combinations (SALC) of s-type orbitals mimicking electrical charges attached to the prism corners are displayed in Fig. 1 (a) and (b) together with the corresponding FDTD simulations (c and d). The SALC basis is constituted of the two linear combinations (2, −1, −1) and (0, −1, 1), whose perpendicular polarizations are respectively aligned on the altitude and on the edge of the considered equilateral triangle.

The (nPPE) – PEEM technique making use of a true optical excitation scheme (photon in, electron out), the polarization state of the excitation light can be easily controlled. When excited by an in-plane electric field vector aligned along one of its edges, a nanoprism displays a two spot near field resonance reminiscent of the SALC E'(0, −1, 1) state, see Fig. 2. Similarly, for excitation along triangle altitudes, dipolar resonances show one strong spot in qualitative agreement with the expected E'(2, −1, −1) SALC state. Similar conclusion conducted at shorter wavelength hold for the quadrupolar plasmon modes.

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