Materials Today • Volume 17, Number 10 • December 2014

Nonlinear plasmonic antennas

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Contrary to traditional optical elements, plasmonic antennas made from nanostructured metals permit the localization of electromagnetic fields on length scales much smaller than the wavelength of light. This results in huge amplitudes for the electromagnetic field close to the antenna being conducive for the observation of nonlinear effects already at moderate pump powers. Thus, these antennas exhibit a promising potential to achieve optical frequency conversion and all-optical control of light at the nano-scale. This opens unprecedented opportunities for ultrafast nonlinear spectroscopy, sensing devices, on-chip optical frequency conversion, nonlinear optical metamaterials, and novel photon sources. Here, we review some of the recent advances in exploiting the potential of plasmonic antennas to realize robust nonlinear applications.

Introduction

Surface plasmon polaritons (SPPs) refer to the oscillation of charge density waves resonantly coupled to the electromagnetic field at a metal–dielectric interface [1]. SPPs stand out in comparison with light in conventional all-dielectric optical elements for their confinement is not restricted by the conventional diffraction limit [2]. Therefore, SPPs can be localized and guided deeply sub-wavelength, which promises a viable route toward the realization of ultra-compact waveguides and antennas for integrated optics [1,3,4].

In addition to realizing compact optical elements, the deep sub-wavelength confinement of SPPs is accompanied by huge near-field amplitudes of the electromagnetic fields. This permits the observation of nonlinear effects at moderate power levels even though the interaction volume might be very small. Mathematically this means that the induced polarization in the medium ceases to be linearly related to the electric field. Restricting our consideration for the moment to a non-resonant interaction of light with homogenous matter, the resulting nonlinear polarization can be described by higher order Taylor terms as [5]

$$P_i(t) = \varepsilon_0 \int_0^\infty R_{ij}^{(1)}(\tau) E_j(\vec{r}, t-\tau) d\tau + \varepsilon_0 [\chi_{ijk}^{(2)} E_j(t) E_k(t) + \chi_{ijkl}^{(3)} E_j(t) E_k(t) E_l(t) + \ldots], \quad (1)$$

where the indices run over the Cartesian coordinates, ε_0 is the permittivity of free space, $R_{ij}^{(1)}(t)$ the response tensor whose Fourier transform yields the susceptibility tensor $\chi_{ij}^{(1)}(\omega)$ of a linear, dispersive medium, whereas $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$ are the non-dispersive second- (quadratic) and third-order (cubic) nonlinear susceptibility tensors, respectively. This substantial simplification of vanishing nonlinear dispersion can be made if all frequencies involved are far below any resonance frequency of the nonlinear medium and Kleinman symmetry holds [5]. In this case both tensors are real-valued. While the cubic nonlinearity is present in all media in varying strength, the quadratic term vanishes in bulk centrosymmetric media [5]. The nonlinear polarization (Eq. (1)) includes the most prominent effects as parametric frequency mixing, including parametric amplification, and self-phase modulation [5–9].

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http://dx.doi.org/10.1016/j.mattod.2014.05.009

For dielectric optical media these nonlinear effects have been extensively investigated and experimentally observed in the past [5]. However, much remains to be explored in plasmonic systems, which can enhance the nonlinear response both in the surrounding dielectric media (sometimes called extrinsic response), and in the metal itself, which forms the plasmonic systems (sometimes called intrinsic response). This is evidently caused by the substantial field enhancement for moderate input pump powers. Prominent examples are quadratic nonlinear effects such as second harmonic generation (SHG) due to symmetry breaking at the surface of centrosymmetric media [10–12] as well as a quadratic response in metals which emerges from a full hydrodynamic model for the electron gas [13–16]. In particular, the latter aspect has attracted a considerable deal of research interest because metals being centrosymmetric media are not expected to generate a bulk second order nonlinear response. However, if both intrinsic and extrinsic sources of nonlinearity are present, usually the latter turns out to be stronger. Moving beyond classical limits, nonlinear effects also arise when quantum mechanical effects become prominent. This generally occurs when plasmonic antennas are placed sufficiently close enough that quantum tunneling effects become significant [17–19] or while considering the resonant interaction of plasmonic antennas with quantum systems [20,21].

Although all the aforementioned issues are scientifically interesting and subject to current research, we will restrict the scope of this review to nonlinear effects in plasmonic antennas that can be described by the phenomenological model of Eq. (1). In particular, we will be interested in the opportunities offered by plasmonic antennas to tailor and electromagnetic fields at nanoscale for significantly enhancing the nonlinear effects. To provide a broader overview, we will begin with a brief introduction to the resonant localization of light by plasmonic antennas. A snapshot of recent trends and developments pertaining to the observation nonlinear effects in various systems will follow this.

Plasmonic antenna resonances

The localization of light evoked by plasmonic elements is best introduced by the referential example of a small sphere. According to Mie theory, the field scattered by a metallic sphere of radius much smaller than the wavelength of the incident light corresponds to that of an induced electric dipole moment inside the sphere. The induced dipole moment can be resonantly excited when the so-called Fröhlich condition is satisfied which requires the permittivity of surrounding dielectric to be opposite in sign and twice as large in magnitude as the permittivity of metal [22]. This results in localization and enhancement of light at the interface between a sub-wavelength metallic sphere and the surrounding dielectric medium. Further modification to the basic spherical geometry such as spherical core–shells [23], ellipsoids, cylinders [22], discs [21] or even coupled spheres [24] have also been analytically described. More advanced shapes such as bow-tie structures [25] lead to similar resonant scattering of light with different degrees of freedom to tune the resonance frequency. However, a description of their properties usually requires rigorous numerical treatment [26–29]. To circumvent all constraints imposed by specific geometries, a recent study proposed to employ an evolutionary algorithm to devise plasmonic particles of arbitrary shapes in order to achieve resonances at frequencies on demand [30].

Apart from the particles discussed above, the so-called plasmonic wires can also achieve plasmonic localization of light. They are essentially metallic waveguides of finite length. As such, they form a Fabry–Perot (FP) cavity where the excited propagating SPP mode sustained by the metallic waveguide oscillates back and forth between the waveguide terminations [31,32]. Being a cavity, the structure resonates when the resonance condition of a FP cavity is met. This requires the round-trip phase accumulated by the guided plasmonic mode to be an integer multiple of 2π. Like plasmonic particles, plasmonic wires also come in different flavors such as cylindrical [31], center-fed [33] and also in more complicated geometries such as split-rings [34,35]. This provides various degrees of freedom to tailor their response according to specific needs.

Nonlinear frequency conversion

Departing from the ability to tailor the electromagnetic response in linear optics, the most common manifestation of nonlinear effects in plasmonic antennas is the conversion of the frequency of light to second harmonic (SH) in a quadratic medium and third harmonic (TH) when the material exhibits a cubic nonlinear response. A natural strategy for harvesting the nonlinear potential of plasmonic antennas is to tune the input pump to the antenna resonance, although non-resonant pumping can also enhance nonlinear conversion if the generated harmonic meets the antenna resonance [36]. Most commonly used antennas for nonlinear frequency conversion include spheres [37–39], core–shells [40–43], nano-wires [44–46], bow-ties [6,8,9], asymmetric L-shaped particles [47,48] and split-rings [14,49–51], composite geometries such as dimers [52–54], and clusters configurations [55,56].

From a theoretical standpoint, plasmonic antennas also present an interesting platform to study an artificially induced quadratic nonlinear response in metals. As mentioned in the introduction, the second order term in Eq. (1) vanishes in centrosymmetric bulk media. Symmetry breaking at the surface can nevertheless act as a weak SH dipole source [10–12]. In structured media such as spheres, it is also possible to induce a bulk nonlinear quadratic response due to the excitation of higher-order multipole moments such as the magnetic dipole or the electric quadrupole [39,57]. Asymmetric plasmonic elements, such as L-shaped [58] and split-ring [35,59] antennas, can also be engineered to exhibit an induced non-centrosymmetric response [47,48,50,51,60,61]. In case of split-ring resonators, for instance, the source of bulk quadratic effects has been long suspected to be the magnetic and associated electric quadrupolar moments [49–51,61,62].

We consider spherical plasmonic core–shell (CS) particles as an example of a practical scheme for realizing antennas that serve as a source for high harmonic generation. Their localized resonance can be easily tuned by modifying the ratio between inner and outer radii of the core and shell, respectively [23]. This allows greater flexibility compared to spherical particles whose resonance in the sub-wavelength regime can only be engineered by the physical properties of the surrounding medium [22]. The nonlinear frequency conversion from CS particles has been investigated both theoretically and experimentally [40–42]. Figure 1a shows the CS geometry made up of a gold shell and a $BaTiO_3$ core that exhibits a strong quadratic nonlinear response which was used in Ref. [40] as a tunable source of SH generation (SHG). Figure 1b