Metamaterial nanotips

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Nanostructured metamaterials, especially arrays of metallic nanoparticles which sustain the excitation of localized plasmon polaritons, provide excellent opportunities to mold the flow of light in the linear regime. We suggest a metamaterial structure whose properties are determined not only by its inner geometry but also by its entire shape. We call this structure a metamaterial nanotip. We evaluate the potential of this nanotip to control the size and the location of the field enhancement. Two-dimensional implementations of this metamaterial nanotip were comprehensively numerically simulated and confirm the expected, physically distinct regimes of operation.

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Abundant literature is devoted to the enhancement of local light fields using metallic nanoparticles. This effect is the basis of surface enhanced Raman scattering (SERS) (see e.g. \cite{1,2,3}). Nowadays it is also used for localizing and detecting different nanoobjects, including molecules (see e.g. \cite{4,5,6}), and even for the control of their fluorescence \cite{7,8}. Exciting results (local field enhancement by a factor of $10^6$) were obtained for nanoantennas realized as two closely spaced metallic nanoparticles \cite{9}. In extremely narrow gaps (1...2 nm) between two almost touching metallic nanoparticles the local field can grow even more significantly \cite{10}. In arrays of touching nanospheres with rather large diameters (when the plasmonic resonance is multipolar) the field enhancement in the 1 nm sized domain near the contact point may attain $10^6$ \cite{11}. It explains extraordinary experimental data for the Raman scattering enhancement (up to $10^{14}$) obtained in Ref. \cite{12}.

However, in some applications a trade-off between a maximal local field enhancement and a maximal area where the field is enhanced appears. In fact, in SERS studies situations often arise where the molecules to be detected float in a liquid or a gas \cite{13}. It is then difficult to steer these molecules into a 1 nm sized gap of an optical nanoantenna. In such situations it would be more favorable to create larger spot sizes (e.g. 50...100 nm) at the expense of a reduced field enhancement. Metamaterials (MM) based on metallic nanoparticles open the opportunity to realize and to tailor such spots. As it was shown, e. g., in Ref. \cite{14} arrays of small (a few nm) metal nanoparticles can be used to induce a strong dispersion of the effective permittivity with a Lorentzian line shape. The resonance is centered at that wavelength where the localized plasmon polariton is excited in the ensemble of metal nanoparticles. Such MM permits to accomplish a cavity from closely spaced metal nanoparticles having an effective permittivity not achievable with natural occurring materials. Choosing a proper design for the cavity and operating it in different spectral domains permits to control the location, where light localizes, as well as the field area and magnitude.

The purpose of this paper is to design a MM cavity that concentrates the incident light in a hot spot whose size and shape can be efficiently and reliably controlled by the design parameters of the MM. Two options for the hot spot are considered, namely being located inside or outside the cavity. Prior to further considerations we expect that our proposed MM cavity is subject to an optical analogue of the uncertainty principle. Namely, field enhancement and spot size remain intrinsically coupled. For example, it will be impossible to obtain a huge field enhancement in large spots. However, the intriguing advantage of the proposed MM cavity is that both quantities can be adjusted to the largest possible extent suggesting many potential applications. Examples are elements to couple light from propagating waves to optical nanocircuits or nanofilters \cite{15}, to obtain controlled photonic nanojets \cite{16,17}, as extremely robust tips for scanning near-field optical microscopes, or for biosensors.

We propose a MM cavity of submicrometer size implemented as a small cone made of metal nanoparticles. We call this structure a MM nanotip. The size of the cone’s base can be of the order of $\lambda/2$ or larger. A solid immersion lens or a conventional microcavity creating the conventional light nanojet can be used for focusing the primary light beam on top of the MM nanotip. The MM nanotip further squeezes the nanojet. The coupling of the MM cavity to the incident wave beam is strong since the incident light transmits through the flat cone base. The location of hot spots, their size and shape strongly depend on the MM design parameters (the distances between nanoparticles, their shape, and the size of the cone). Another control parameter is the cone truncation; transforming it to a frustum. Furthermore, the effective permittivity of the cone is determined by the array of nanoparticles and depends only weakly on the matrix. This provides unique features of the structure. First, the nanotip does not need to have a conical physical geometry. By creating a conically shaped array of nanoparticles in a dielectric slab (if we ensure that the apex of the cone coincides with an interface of the slab), effective field localization occurs around a singular surface point at the bottom of the slab. Second, the MM nanotip can be penetrable for molecules (the matrix can
be porous, alternatively one can make voids in the matrix. Both properties are useful for applications indicated above.

High reflection from the cone base can be prevented by gradually increasing the concentration of nanoparticles from the base to the apex of the cone. The light will experience an effective medium with a permittivity increasing without jumps between the base and the apex thus minimizing the reflection losses. In Fig. 1 a sketch of the MM nanotip together with a conventional tapered metal-coated SNOM tip with a subwavelength aperture is displayed. The figure illustrates the idea of the engineered high permittivity cone, our design strategy, and the regime of the nanojet. We emphasize that in this way one can achieve a wave impedance of the effectively tapered transmission line approximately uniform along the entire tip length. As a consequence this MM nanotip may be potentially employed to efficiently couple light into nanofilters, waveguides of metallic nanoparticles and nanoantennas.

At this first stage we simulate a 2D analogue of the suggested structure. Instead of nanoparticles we assume here nanocylinders. For the sake of saving computational time we consider a regular array i.e. no pitch (ratio period/diameter) variation along the nanotip yet. With this approach we cannot reduce spurious reflections at the base, though these reflections do not affect the main conclusions to be drawn and leaves this optimization for a future work. Therefore, the structure consists of periodically arranged metallic nanowires forming the tip. As metal material we have chosen silver. Material parameters were taken from literature. The dielectric matrix is assumed to be glass (n = 1.5). The diameter of the silver cylinders was D = 4 nm. The shape of the tip was chosen to be an equilateral triangle. The structure was illuminated by a TM polarized (electric field in the plane of incidence) plane wave with unit amplitude. The main parameters, modified in this work to identify the operational regimes of interest, are the wavelength and the period.

The effective permittivity of the MM made of closely spaced cylinders was obtained rigorously from the dispersion relation. The MM was reasonably assumed to have no spatial dispersion in the effective permeability. The dispersion relation was calculated with a appropriate plane wave expansion technique. The field distribution around the ultimate device was computed by using an extended Mie theory to treat the case of light scattering at an ensemble of non-penetrating cylinders.

To start with, Fig. 2 shows the effective permittivity of the MM made of nanocylinders as a function of wavelength and for various periods. It is evident, that strong dispersion with a Lorentzian line shape occurs near the wavelength where the localized plasmon polariton is excited in the nanocylinder. The smaller the period the larger is the resonance wavelength. The red-shift arises from the mutual coupling of the resonant fields in adjacent cylinders. This coupling and the larger filling fraction causes also the larger resonance strength for smaller periods. Two spectral domains of interest can be distinguished in general. For wavelengths below resonance the MM behaves metallic (ℜ(ε_{Eff}) < 0) whereas it has dielectric properties (ℜ(ε_{Eff}) > 1) beyond the resonance. We note that the effective permittivity exceeds by far that of any naturally occurring material at optical frequencies. Both regimes can be used to localize light. In the ‘metallic’ domain, surface plasmon polaritons can be excited in the tip. They will cause a strong field concentration at the tip apex in small volumes. In the ‘dielectric’ domain whispering gallery modes are excited in the tip leading to a diffraction-limited field concentration inside the tip. The diffraction limit is given by the ratio of the wavelength to the huge effective refractive index. If the operational wavelength is much larger than the resonance wavelength the field focus may be pushed off the tip. This leads to a strong field enhancement in close vicinity but outside the tip. This operational regime, where the formation of photonic nanocylinders can be observed, adds as a third one to those discussed above.

To identify spectral and parameter domains of interest, the electromagnetic field was computed close to the apex of the tip. The parameters we systematically inves-
tigated were the period $\Lambda$ and the size of the tip. Selected results for various periods are shown in Fig. 3. Strong enhancement and field localization can be seen. This data have been used to extract the parameters space of interest where any of the operational regimes defined are fully evolved.

Finally we have analyzed the entire field distribution for a selected number of relevant configurations. Results are shown in Fig. 4.

For the sake of comparison Fig. 4 (a) shows the electric field amplitude around a single nanocylinder. The chosen wavelength corresponds to the plasmon polarization resonance. Near the surface a field enhancement $\chi = |E^{loc}/E_0|$ of up to $\chi = 15$ can be observed. However, 1 nm off the cylinder surface it decreases to $\chi = 6$ and the averaged enhancement over the area 20 nm off the cylinder does not exceed $\chi = 1.1$.

Figure 4 (b) shows exemplarily a field distribution in the operational regime where the effective properties of the tip material are metallic and surface plasmon polaritons are excited in the tip. The field enhancement is strong at any apex and attains a maximum of $\chi = 7$. Reminiscent to the plasmonic properties of the entire tip is the rather constant field amplitude inside the tip, being comparable to the field amplitude of the single nanocylinder as shown in Fig. 4 (a).

Figure 4 (c) shows a field distribution in the operational regime where the effective properties of the tip material are essentially dielectric (only a minor imaginary part of the effective permittivity) and whispering gallery modes are excited. The field enhancement $\chi$ is strong at three hot spots inside the tip and can be further increased for $\Lambda \to D$.

Figure 4 (d) shows exemplarily a field distribution in the third regime where the effective properties of the tip material are essentially dielectric (negligible imaginary part of the effective permittivity) but the field is dragged out of the tip and a photonic nanojet appears. The amplitude enhancement inside the nanojet ($\chi \approx 1.7$) is a sign for an excellent optical coupling from the incident wave to the photonic nanojet. The effective width of the nanojet is approximately 120 nm (0.42$\lambda_n$, where $\lambda_n = \lambda/h$ is the wavelength in the host medium). We note that due to the wedge shape, the structure resembles a two-dimensional axicon. In a first approximation the evolving beam has two major spatial frequencies. The wedge shape structure therefore significantly suppresses diffraction, being much in favor for the photonic nanojets.

To sum up, we have suggested a new metamaterial structure, namely a MM nanotip, that can evoke field enhancement in comparatively large and controllable spatial domains and for molding the flow of light. Three operational regimes have been identified that permit for either field concentration inside the tip, directly at the apex or the formation of a highly subwavelength photonic nanojet emerging from the tip apex. The very basis of all these effects is the strong dispersion of the effective permittivity of the MM that forms the tip. Variation of the period and/or the shape of the nanocylinder array allow to control the size, shape and location of the hot spot. Based on these preliminary 2D simulations one can expect a rather strong field enhancement for 3D MM nanotips in comparatively large and controllable spatial domains.
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