Coherent Control of Nanoscale Light Localization in Metamaterial: Creating and Positioning Isolated Subwavelength Energy Hot Spots

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We show the strong optically induced interactions between discrete metamolecules in a metamaterial system and coherent monochromatic continuous light beam with a spatially tailored phase profile can be used to prepare a subwavelength scale energy localization. Well-isolated energy hot spots of a fraction of a wavelength can be created and positioned on the metamaterial landscape offering new opportunities for data storage and imaging applications.

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Precise control and manipulation of optical fields on a nanoscale is one of the most important and challenging problems in nanophotonics. Since optical wavelength is on a much larger microscale, it is impossible to employ conventional focusing for that purpose. In 2002 a method was suggested, which was based on tailoring phase modulation of ultrashort optical pulses in the time domain to achieve coherent control of spatial distribution of the excitation energy in complex inhomogeneous nanosystems [1]. In such systems the localized plasmon frequencies vary from nanoscale feature to nanoscale feature and thus can be correlated with their positions. The pulse phase modulation will cause the exciting field to take energy away from surface plasmons localized in those parts of the system where the oscillations are out of phase with the driving pulse and move it, with time, to the surface plasmon excitations in other parts where such oscillations occur in phase with the driving pulse. Later a similar idea based on tailoring of ultrafast pulses in the vicinity of silver nanostructures through adaptive polarization shaping has been successfully demonstrated [2,3]. It is also possible to control the near-field of nanoantennas by controlling polarization singularities and inhomogeneities of the incident field [4] and tailored plasmon interference [5]. Recently illuminating a subwavelength diffraction grating by an amplitude and incident angle optimized continuous-wave light beam has been suggested as a method to generate and control the location of light hot spots [6].

Here we show that strong interactions between regularly spaced plasmonic resonators, known as metamolecules, in a planar metamaterial, a two-dimensional array of nanoscale metamolecules, can result in a subwavelength localization of optical energy. A desired nanoscale light hot spot can be engineered simply by adjusting the far-field spatial profile of an incoming monochromatic coherent continuous light beam. Figure 1 shows artistic representation of the metamaterial array excited by the wave with a tailored phase profile creating a hot spot at the other side of the array, at its immediate proximity. We illustrate by using a simple model how the localization results from a cooperative response of the metamaterial array of asymmetrically split ring plasmonic resonators, providing physical insight into the energy localization effect. The essential requirement in the subwavelength localization process of the incoming wave is that individual metamolecules act as closely spaced discrete scatterers whose strong inter-metamolecular interactions modify the energy eigenspectrum of the system. This is in contrast with the homogenization approach to electromagnetic properties of

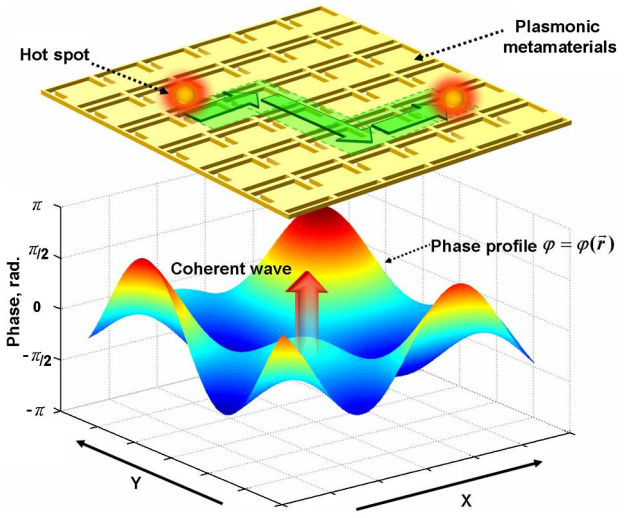

FIG. 1 (color online). Coherent control of light localization in an array of interacting metamolecules: artistic impression. A light beam with modulated phase profile excites a metamaterial array of interacting plasmonic resonators leading to the formation of the energy hot spot at the metamaterial. This spot may be moved from a metamaterial cell to another metamaterial cell by tailoring the phase profile of the incident beam.

metamaterials treated as a continuous medium instead of a collection of discrete metamolecules.

We show that evanescent field energy can be localized almost entirely at isolated metamolecules while groups of neighboring metamolecules remain unexcited. Now by simply adjusting the spatial phase profile of the incident beam the nanoscale hot spots can be moved from one metamolecule to another, providing an efficient technique for a subwavelength scale optical control and manipulation of a metamaterial system. In contrast with localization techniques relying on laser-pulse excitation [1–3] our method is based on a continuous-wave source and does not depend on a transient redistribution of energy between nano-objects. This makes the localization in a metamaterial system much simpler to implement. Moreover, it does not require the nanoscale system to be spatially inhomogeneous (such as a rough surface) and works with periodic, regular planar array of identical nano-objects opening the opportunities for microscopy and data storage applications which will be discussed below.

The idea of coupled resonators illuminated by a phase-modulated beam has a clear mechanical analogy in two coupled identical oscillators (modeling nano-objects) that may be driven to different amplitudes by setting up a phase delay between otherwise identical coherent mechanical forces driving them.

The spatial variation of phase required to generate the hot spots is slow and could take place on the scale longer than the wavelength of light. This can be routinely achieved experimentally using widely commercially available liquid crystal spatial light modulators (SLM). The choice of asymmetrically split ring array as the nanoscale landscape was motivated by the recent studies that established a key role of inter-metamolecular interaction in forming the electromagnetic response of such metamaterials [7]. We will see below that the existence of strong interactions between plasmonic resonators is the key requirement for nanoscale localization.

Our analysis of this system is based on direct quantitative numerical simulation using a 3D Maxwell solver illustrated by qualitative modeling the metamaterial as a regular array of discrete interacting dipolar scatterers. In all numerical simulations we concentrated on the visible and the near-infrared parts of the spectrum, assumed that metamaterial structures are constructed of gold plasmonic resonators and used realistic material parameters and Joule loss factors by using the well-established data for the dielectric parameters of the metal [8].

We demonstrate the idea of coherent control of light localization using the excitation beam with a periodic phase modulation across its wave front: even this most simple profile leads to a profound light localization at the array of interacting metamolecules at certain frequencies of the driving field. Figure 2 shows intensity maps at the metamaterial array driven by a monochromatic field manipulated such that, upon arrival at the metamaterial surface, the electric field has constant amplitude, a fixed polarization along the y direction, and a spatially varying phase $E(x, y) = E_0 e^{i\phi(x,y)} = \sum_{mn} C(m, n) e^{imkx} e^{imky}$. The incident wave has the wave front modulated along x and y with a period of six unit cells of the metamaterial array. In calculation only series components with $|m, n| \leq 2$ were retained to ensure that in the entire spectral range of interest mk, $nk < 2\pi/\lambda$ and thus the field $E(x, y)$ can be created by a far-field phase modulating device such as SLM.

The array is a metal gold film of 50 nm thickness. It is perforated with 25 nm slits arranged in a split square shape 200 nm × 200 nm in size constituting the metamolecule of the structure [see Fig. 2(h)]. The metamaterial's lattice period is 440 nm × 440 nm. This metamaterial nanostructure has a well-defined plasmonic absorption resonance at 880 nm corresponding to a local maximum in transmission and a minimum in reflection. We investigated field localization in the immediate proximity to the array at this wavelength of excitation and at two other wavelengths above (1150 nm) and below (700 nm) the resonance, at local minima of absorption and transmission.

The main characteristic features of the coherent control process are illustrated on Fig. 2, which shows (a)–(c) the intensity distribution at the array as a function of the amplitudes of spatial phase modulation. As expected, for a plane wave front of the incident beam ($\Delta\phi_{\max} = 0$) all metamolecules in the array are excited equally. With an increase of the modulation depth, energy tends to concentrate at two opposite quadrants of the 6 × 6 section of the array at a distance of $3\sqrt{2}$ unit cells from one another. For a fixed amplitude of phase modulation the localization is most pronounced at the plasmonic absorption resonance at 880 nm; detuning away from the resonance destroys the localization, as illustrated on Figs. 2(c)–2(e). Figure 3(a) shows intensity cross section of the isolated central peak of the resonant hot spot (880 nm) which has a footprint (at half maximum) of only 50 nm × 152 nm (0.0076 μm^2).

The main features of the coherent control may be illustrated by a simple model in which we treat current oscillations in individual metamolecules as pointlike dipoles. An incident driving field of a given wavelength excites the current oscillations, producing scattered radiation. These emitted fields in turn drive the corresponding oscillations in neighboring metamolecules. The system dynamics can be described by the Maxwell's wave equations, for instance,

$$(\nabla^2 + k^2)\vec{D}^+ = -\nabla \times (\nabla \times \vec{P}^+) - ik\nabla \times \vec{M}^+, \quad (1)$$

where $\vec{D}^+(\vec{r}, \Omega)$, $\vec{P}^+(\vec{r}, \Omega)$, and $\vec{M}^+(\vec{r}, \Omega)$ are the positive frequency components of the electric displacement, polarization, and magnetization densities, respectively, at frequency Ω [$\vec{P}^+(\vec{r}, t) = \sum_\Omega \vec{P}^+(\vec{r}, \Omega) e^{-i\Omega t}$]. For an array metamolecules labeled by an index j, at position $\vec{r}_j$, with electric (magnetic) dipole moments $\vec{d}_j$ ($\vec{\mu}_j$), the

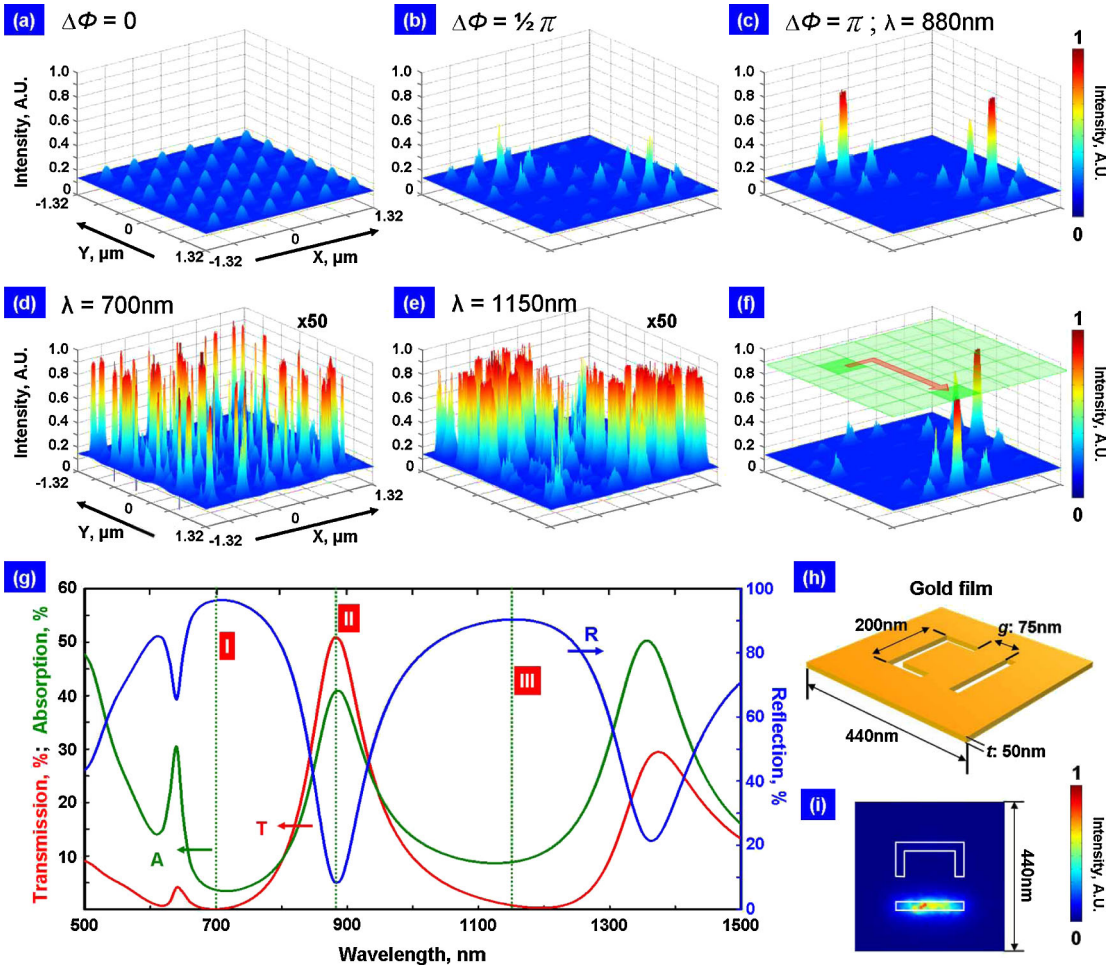

FIG. 2 (color online). Coherent control of energy localization at metamaterial array of interacting plasmonic resonators. Color maps (a)–(c) show the buildup of light localization with the increase of sinusoidal phase modulation of the driving field $\Delta\phi$. Note two intensity peaks formed at the opposite quadrants of the landscape at resonance conditions at λ 880 nm and $\Delta\phi = \pi$. Maps (c)–(e) show the light localization effect at different wavelength for $\Delta\phi = \pi$: tuning the excitation wavelength away from the resonance (d) $\lambda = 700$ nm and (e) $\lambda = 1150$ nm destroys the localization. Figure (f) illustrates how the hot spot can be moved across the metamaterial plane by shifting the phase profile of the incident beam by $(a, -5a)$. Figure (g) shows the far-field transmission, reflection, and absorption spectra of a plane wave ($\Delta\phi = 0$) impinging on the metamaterial array and its unit cell (h). Localization is most pronounced at $\lambda = 880$ nm corresponding to the absorption resonance II. Figure (i) shows the position of the hot spot within the metamolecule at resonance II.

polarization and magnetization densities are given by $\vec{P}(\vec{r},t) = \sum_j \vec{d}_j(t)\delta(\vec{r}-\vec{r}_j)$ and $\vec{M}(\vec{r},t) = \sum_j \vec{\mu}_j(t)\delta(\vec{r}-\vec{r}_j)$, respectively. Because of dipole-dipole interactions between metamolecules, the material displays collective modes of oscillation, each with a distinct resonance frequency and collective radiative damping rate. The majority of collective modes do not couple efficiently to the uniform driving field due to the spatial inhomogeneity of the modes. However, by allowing spatial variation in the driving field, one shall be able to excite a superposition of collective modes. A superposition of such modal excitations can lead to the formation of hot spots located only at a few isolated metamolecules.

The effect of strong inter-metamolecular interactions in the energy localization may be illustrated by a simple example of two electric dipoles with a separation much less than the wavelength of light. The collective state of the dipoles exhibits superradiant (enhanced radiative damping rate) and subradiant (suppressed radiative damping rate) eigenmodes. In the first eigenmode the two dipoles are in phase and in the second one they are π out of phase. If we could prepare an incident field that excites an equal superposition of the two modes, it could then localize the energy in one of the dipoles. The crucial part in the emergence of the subradiant and superradiant states in a two-dipole system are recurrent scattering events [9,10]—processes in which a wave is scattered more than once by the same dipole—which cannot be described by the standard continuous medium electrodynamics, necessitating a discrete scatterer model of the metamaterial system. We present a less trivial example in the supplemental material

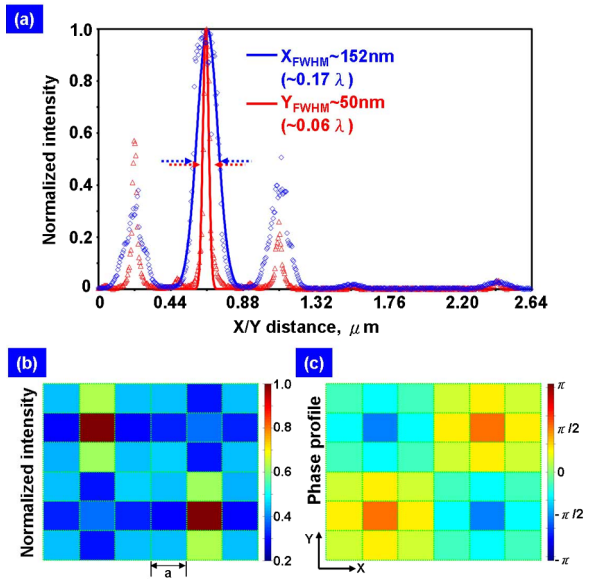

FIG. 3 (color online). The main cross sections of the energy hot spot (a) created at the metamaterial landscape corresponding to the conditions of Fig. 2(c) indicate a subwavelength localization with a footprint at the peak of about 1% of λ^2 while there is no excitation on the other quadrant. Color maps (b) and (c) show the intensity and phase of excitations within the model of individual interacting dipoles representing the metamolecules of array.

[11]. A numerical example of field localization achieved within the dipole model and showing all characteristic features of the coherent process control is illustrated in Fig. 3(b).

The opportunity to localize energy hot spots at isolated metamolecules provides for interesting applications. Indeed, by simply shifting phase profile function of the driving beam $\phi(x,y) = (\Delta\phi/2)\sin(\kappa x + \delta x)\sin(\kappa y + \delta y)$ on δx and δy, integers of the lattice period, one can reposition the hot spot across the metamaterial landscape, from one metamolecule to the other. This is illustrated in Fig. 2(f) by shifting the phase profile one lattice period along $\hat{x}$ and five periods along $-\hat{y}$. In what has been illustrated above, the hot spots can be positioned $3a\hat{x} + 3a\hat{y}$ apart and thus moved at chosen metamolecules in this space. Our computational capabilities do not allow for numerical modeling of driving fields modulated at a larger scale which would involve modeling of a larger array of metamolecules. We argue, however, that similar localization effects could take place in larger arrays where individual hot spots are localized in much larger dim areas. Moreover, we argue that localization will be improved by appropriate selection of metamaterial, unit cell, and the resonator size, and by controlling losses (for instance by using silver instead of gold) that will ensure long-range interactions between metamolecules.

Controlling the hot spot position on the metamaterial chessboard may be used for high-density data storage application, in particular, with phase-change nanoparticles [12] placed at the metamolecules and switched at the hot spot. Moreover, the light localization technique can be used in a new type of optical imaging instruments with subwavelength resolution: one can envisage that an object of study (say, a cell) is placed on the metamaterial surface with a light-collecting optics and a photo detector above it. Moving the hot spot from one place to another shall lead to the hot spot being exposed to different scattering or absorption regimes which will affect intensity at the photodetector. A map of such intensities can be build to reconstruct a subwavelength image of the object. Here the metamaterials lattice pitch defines imaging resolution and in some designs can be several orders of magnitude smaller than the wavelength [13].

In conclusion, we demonstrated coherent control of light localization in photonic metamaterial. Well-isolated energy hot spots as small as $0.06\lambda \times 0.17\lambda$ can be created and positioned at chosen metamolecules on the plasmonic landscape by illuminating an array of interacting plasmonic resonators with a coherent continuous light beam with a tailored spatial variation of the phase profile.

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