

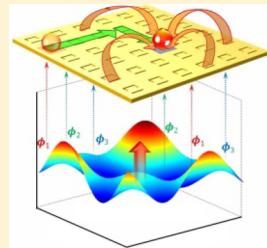
"Digitally" Addressable Focusing of Light into a Subwavelength Hot Spot

T. S. Kao, E. T. F. Rogers, J. Y. Ou, and N. I. Zheludev*

Optoelectronics Research Centre and Centre for Photonic Metamaterials, University of Southampton, Southampton SO17 1BJ, United Kingdom

 Supporting Information

ABSTRACT: We show that a plasmonic metamaterial can act as a far-field to near-field transformer that focuses a free-space beam of light into a subwavelength energy hot spot at a prescribed location with a spot size only a small fraction of the wavelength. The hot spot position on the metamaterial can be prescribed and moved at will from one metamolecule of the array to another in a "digital" fashion simply by modulating the input phase profile, thus providing new opportunities for imaging and optical data processing.



KEYWORDS: *Light localization, metamaterials, coherent control, nanophotonics*

It is commonly believed that a light beam cannot be focused to a spot that is significantly smaller than the wavelength of light in free space. However, strictly speaking, focusing light into a small hot spot is possible by precisely tailored interference of multiple optical beams, the phenomenon known as superoscillation.^{1–3} However practical use of superoscillations is complicated by the presence of high intensity halos surrounding the hot spot, an issue that only recently has been overcome.^{4,5} Another approach, based on the negative-index metamaterial lens, also offers a subwavelength focusing capability, however this technology still remains at the development stage, trying to overcome the severe obstacles of energy losses and challenging nanofabrication.^{6–10} In terms of near-field focusing, nanoscale light concentration may be achieved by illuminating a subwavelength diffraction grating with optimized light beams,¹¹ adiabatic transformation of a plasmonic field,^{12–15} coherent control of ultrafast pulses^{16–18} and arranging constructive interference in plasmonic and nanoantenna systems,^{19–22} and exploiting mode interference in arrays of wires.²³

Here we experimentally demonstrate a different approach of focusing a free space light beam into a nanoscale near-field hot spot placed with "digital" precision on a nanostructured landscape. The crucial difference in our approach, first described theoretically in ref 24, is the use of intense interactions and energy exchange between the building blocks of a plasmonic metamaterial to focus a free-space wave with a tailored phase profile into a nanoscale hot spot on the metamaterial surface. This idea of coherent control of light localization is illustrated in Figure 1. Indeed, if a regular array of metamolecules is illuminated by a plane wave, all the molecules will be excited to identical levels, as required by the translational symmetry of the system. However, if plasmonic excitations are driven by a wave with a spatially modulated phase profile, and thus metamolecules are driven with relative

phase lags, energy may be redistributed within the metamaterial through interactions between metamolecules, creating hot spots located on certain metamolecules. Here the position of the hot spot is precisely anchored to the metamolecule, and, due to the interactions, the modulation of near-field can be achieved at a finer scale than the scale of modulation of the incident light beam. Moreover, a desired nanoscale hot spot can be moved from one metamolecule to another in a "digital" fashion simply by adjusting the far-field spatial profile of an incoming monochromatic coherent continuous-wave light beam. The idea has a clear mechanical analogue in coupled identical oscillators (modeling metamolecules) that may be driven to different amplitudes by setting up a phase delay in between the driving forces (see Figure 2).

Interactions are crucial in allowing redistribution of energy on the surface of the metamaterial. Without these interactions it will not be possible to transform large scale modulation of the incident beam into an isolated nanoscale localization in the near-field. For that reason we have chosen a plasmonic metamaterial consisting of arrays of asymmetrically split-ring nanoscale resonators as the far- to near-field transformer in our experiments. This metamaterial has been shown in recent studies to have strong interactions that are key to forming its electromagnetic response.²⁵ The metamaterial with a lattice period of 440 nm was fabricated by focused ion beam milling in a gold film of approximately 50 nm thickness deposited on a transparent silica substrate. The pattern consisted of 25 nm wide slits arranged in a split square shape $200 \times 200 \text{ nm}^2$ in size. Figure 3d shows a scanning electron microscope (SEM) image of a 6×6 section of the array. The metamaterial was

Received: December 9, 2011

Revised: March 21, 2012

Published: March 28, 2012

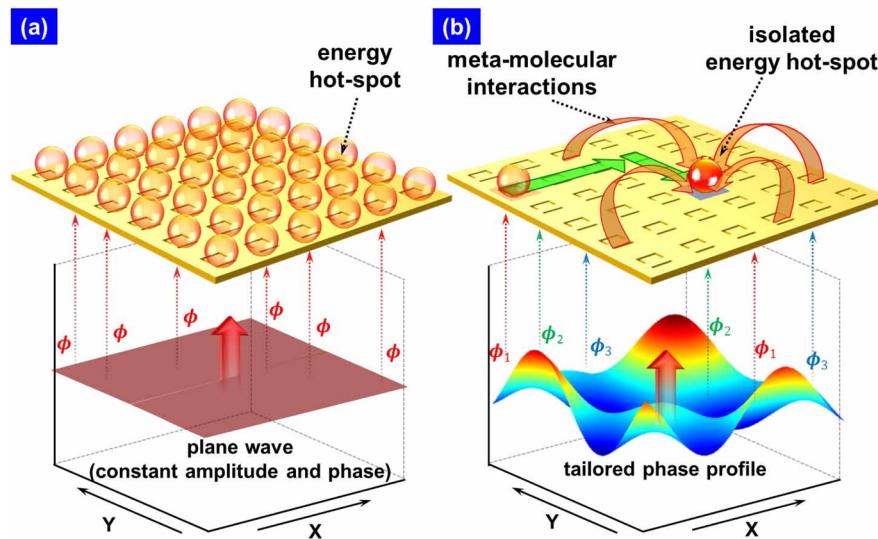


Figure 1. Plasmonic metamaterial as near-field focusing device. Artistic impression. (a) For plane wave illumination, the optical energy on the metamaterial landscape is homogeneously distributed. (b) By spatially tailoring the phase profile of an incident light beam, the strong optically induced interactions between metamolecules in a metamaterial system can lead to the formation of a subwavelength hot spot located on a certain meta-molecule. This spot may be moved from one metamaterial cell to another by tailoring the phase profile of the incident beam.

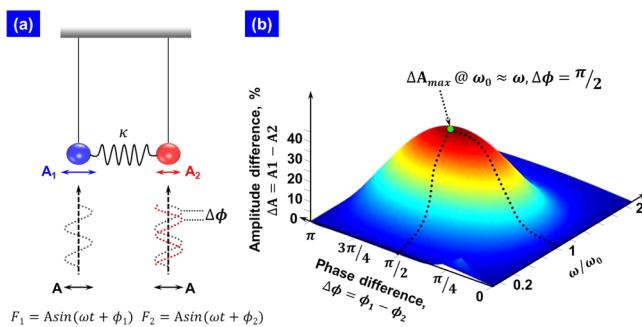


Figure 2. A mechanical analogue illustrating energy localization by coherent (phase) control. Identical coupled oscillators driven at the same frequency with the same amplitude will be excited to different levels if they are driven with a phase delay (a). Here oscillators represent individual metamolecules of the metamaterial structure. Plate (b) shows the difference between the amplitudes of oscillation as a function of frequency ω and phase difference ϕ between driving forces. Here ω_0 is the resonant frequency of individual oscillators.

illuminated by a laser beam shaped by a liquid crystal phase spatial light modulator (SLM). The light distribution in the metamaterial was monitored using an optical microscope and mapped with a scanning near-field microscope (SNOM) employing 80 nm aperture polarization-insensitive metal-coated tapered fiber SNOM probes.

Light localization in the metamaterial was studied with the spatial light modulator introducing a harmonically modulated phase profile $\varphi(x,y) = (\Delta\varphi_{\text{max}}/2)\sin(\kappa x)\sin(\kappa y)$ to the incident wave. Here, $\kappa = 2\pi/(6a)$ and a is the period of metamaterial pattern. As has been illustrated by computer simulations, such a spatially modulated phase profile leads to a concentration of the near-field mainly at two diagonal metamolecules of the 6×6 section of the grid. The effect has a resonant character and can only be observed at a wavelength at which interaction between metamolecules is strong, corresponding to the plasmonic absorption peak.²⁴ The wavelength of $\lambda = 852$ nm used in our experiments is within the plasmonic absorption resonance envelope of this nanostructure that is centered at $\lambda = 930$ nm. It

should be noted that due to the finite numerical aperture of the imaging system the application of the phase modulation to the input beam may result in small unintended intensity modulations. These do not significantly affect either the principle of operation or the performance of the system.

The main characteristic features of the effect are illustrated in Figures 3 and 4. We first studied light localization as a function of depth of the free space wave spatial modulation. In Figure 3, the green grid indicates the boundaries of the metamolecules. As expected, for a flat wavefront of the incident beam ($\Delta\varphi_{\text{max}} = 0$), all metamolecules in the array are excited equally. Figure 3a shows a nearly homogeneous intensity distribution on the metamaterial landscape. With increasing modulation depth (Figure 3b,c), energy progressively concentrates at two opposite quadrants of the 6×6 section of the array at a distance of $3\sqrt{2}$ unit cells from one another. In the images in panels a–c, the perceived size of the hot spot is limited by the microscope resolution. To investigate the actual size of the energy hot spots, near-field optical mapping has been conducted. Figure 3f is the SNOM mapping of the metamolecule at which the energy hot spot is located. It shows that in fact the hot spot is $184 \text{ nm} \times 147 \text{ nm}$ ($0.22\lambda \times 0.17\lambda$) in size and thus the combination of interaction between metamolecules and resonance within a single metamolecule allows the near-field to be modulated at a scale that 18 times smaller than the scale at which the incident free space field is modulated. This conforms to full 3D Maxwell calculations (inset to Figure 3d) that place the hot spot at the middle of the short slit of the metamolecule and indicate that its intensity is about 120 times stronger than the intensity of the incident beam, when the input beam is modulated as in Figure 3c. Figure 3e shows the topography of the metamolecule.

At resonance conditions, light passes through the plasmonic metamaterial predominantly through the center of the short part of the split-ring slit, as illustrated on Figure 3d and confirmed in the SNOM scan in Figure 3f. Figure 3c shows a microscope image of the light coming through the metamaterial that is diffraction limited, as would be appropriate for a single isolated subwavelength hot spot. We see this as unambiguous

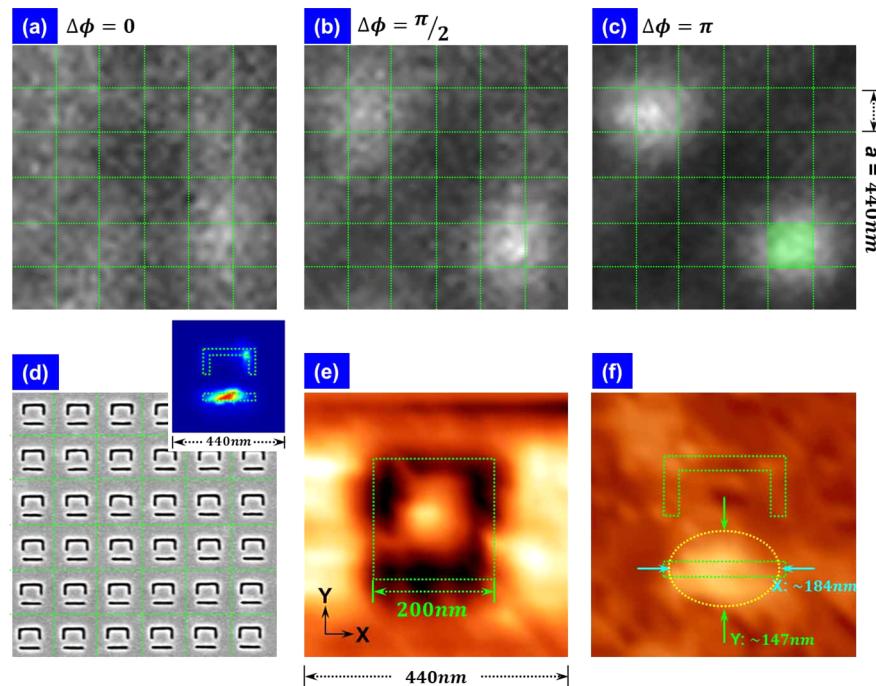


Figure 3. Experimental demonstration of coherent control of light localization in a plasmonic metamaterial observed with optical microscope and scanning near-field microscope. Panels a–c show transmission optical microscope images of the split-ring nanostructure illuminated with coherent laser radiation at $\lambda = 852$ nm at different wavefront distortions: plane wave illumination, reference (a); harmonic phase modulation with period of six lattice cells ($2.64 \mu\text{m}$) and $\Delta\phi = \pi/2$ (b) and for $\Delta\phi = \pi$ when the maximum localization is achieved (c). Note that the optical microscope does not resolve the actual size of the hot spot. Panels d and e represent the scanning electron microscope image of the plasmonic metamaterial array (gold film with groove pattern) and atomic force microscope image of the individual unit cell ($440 \text{ nm} \times 440 \text{ nm}$), respectively. The intensity distribution measured by the scanning nearfield microscope (f) reveals the subwavelength hot spot ($184 \text{ nm} \times 147 \text{ nm}$) located at the short groove, exactly as predicted by full 3D Maxwell calculations, see inset to panel d.

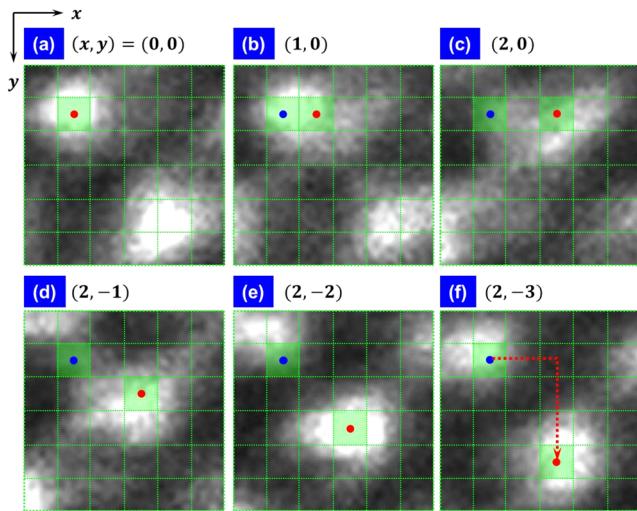


Figure 4. Moving the light hot spot on the metamaterial chessboard at will. Control of the hot spot position is achieved by modulating the phase profile of the incident beam. The red dot indicates the targeted position of the hot spot. Images b–f show sequential movement of the hot spot along the trajectory indicated by the red dashed arrow, that is, between the coordinate points $(0,0)$ and $(2,-3)$.

evidence that, indeed, an isolated metamolecule is excited due to intermolecular interactions.²⁴ If, however, a tightly focused beam centered on a single cell were used instead, it would create a spot of $\sim 708 \text{ nm}$ fwhm and would also illuminate the surrounding cells (spaced by 440 nm) to a significant level,

creating in the microscope image a broader structured spot, that we do not see.

Figure 4 illustrates how the energy hot spot can be repositioned on the metamaterial “chessboard”. By simply shifting the phase profile of the driving beam $\varphi(x,y) = (\Delta\phi_{\max}/2)\sin(kx + \delta x)\sin(ky + \delta y)$ by δx and δy , integer multiples of the lattice period, the hot spot is repositioned across the metamaterial landscape, from one metamolecule to another. This is illustrated in Figure 4a–f as a sequence of translations, two periods along \hat{x} and then three periods along $-\hat{y}$. This illumination shift is achieved by applying a small phase tilt to the pattern displayed on the spatial light modulator.

The experiments reported above confirm the viability of nanofocusing through coherent control on a 6×6 fragment of the metamaterial array. We note that metamolecular interactions helped localize light at a much smaller scale than the required wavefront modulation. Using this approach, localization can be controlled in the neighborhood of metamolecules that is covered by the extent of the interactions. Previous measurements conducted with arrays of asymmetric split-rings of different size²⁶ indicated that the interaction neighborhood increases with the reduction of Joule losses in the system. One therefore can expect that the nanofocusing mechanism will work even better in the near-infrared part of the spectrum (where plasmonic Joule losses are smaller) and also with lower loss plasmonic metamaterials such as silver. These improvements will offer a much bigger “photonic chessboard” for manipulation of the near-field.

In contrast with the recently reported plasmon interference technique that offers diffraction limited hot spots,¹⁹ our method

generates hot spots that are substantially subwavelength (the hot spot footprint reported here is $\sim 0.04 \times \lambda^2$). Having a subwavelength hot spot is an extremely important advantage not only for imaging but also for nonlinear and switching applications. A very distinct feature of our approach is that the hot spot may be moved and repositioned in a quantized fashion, from one meta-molecule to another, in steps of the lattice period (about $\lambda/2$ in our case). We expect that an increase of the positioning resolution will be possible with further progress in photonic metamaterials where development of resonant nanostructures with a small ratio between the lattice period and the resonant wavelength is a priority (metamolecules as small as $\sim 0.15\%$ of the resonant wavelength have already been demonstrated at radio frequencies²⁷). Such quantization of the hot spot location makes this method very attractive for “digital” applications. At this stage we have demonstrated the ability to activate two hot spots on the array of 36 metamolecules and move them across the metamaterial landscape. We argue that since the area of metamaterial over which the integration between the metamolecules takes place increases with the reduction of Joule losses and improves at resonance,²⁸ a similar tendency shall be expected for the size of the area where interaction between metamolecules can affect localization. A challenge remains to develop algorithms that will allow writing arbitrary patterns of near-field hot spots into the metamaterial array. The ability to write an arbitrary pattern of optical hot spots on the photonic chessboard will provide substantial new opportunities for optical data storage for instance, with phase-change nanoparticles²⁹ placed at the metamolecules, and also for addressing of nanophotonic circuits and nanoluminescent bioassay labeling.

In conclusion, we experimentally demonstrated energy hot spots with a substantially subwavelength footprint that are created and positioned at a chosen metamolecules on the metamaterial array with a continuous-wave light beam with a spatially tailored phase profile using a far-field to near-field plasmonic metamaterial transformer.

■ ASSOCIATED CONTENT

Supporting Information

Supporting Information with further details of the experimental arrangement and the mechanical analog of the coherent control process is available for this letter. This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: n.i.zheludev@soton.ac.uk.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors would like to thank J. Ruostekoski and S. Jenkins for fruitful discussions and to acknowledge the financial support of the Engineering and Physical Sciences Research Council, U.K. and the Royal Society.

■ REFERENCES

- (1) di Francia, G. T. *Nuovo Cimento, Suppl.* **1952**, 9, 456–438.
- (2) Berry, M. V.; Popescu, S. *J. Phys. A: Math. Gen.* **2006**, 39, 6965–6977.
- (3) Zheludev, N. I. *Nat. Mater.* **2008**, 7, 420–422.
- (4) Rogers, E. T. F.; Lindberg, J.; Roy, T.; Savo, S.; Chad, J. E.; Dennis, M. R.; Zheludev, N. I. *Nat. Mater.* **2012**, 11, 432–435.
- (5) Kosmeier, S.; Mazilu, M.; Baumgartl, J.; Dholakia, K. *J. Opt.* **2011**, 13, 105707.
- (6) Pendry, J. B. *Phys. Rev. Lett.* **2000**, 85, 3966–3969.
- (7) Shelby, R. A.; Smith, D. R.; Schultz, S. *Science* **2001**, 292, 77–79.
- (8) Fang, N.; Lee, H.; Sun, C.; Zhang, X. *Science* **2005**, 308, 534–537.
- (9) Liu, Z.; Lee, H.; Xiong, Y.; Sun, C.; Zhang, X. *Science* **2007**, 315, 1686.
- (10) Valentine, J.; Zhang, S.; Zentgraf, T.; Ulin-Avila, E.; Genov, D. A.; Bartal, G.; Zhang, X. *Nature* **2008**, 455, 376–379.
- (11) Sentenac, A.; Chaumet, P. C. *Phys. Rev. Lett.* **2008**, 101, 013901.
- (12) Stockman, M. I. *Phys. Rev. Lett.* **2004**, 93, 137404.
- (13) Schroer, C. G.; Lengeler, B. *Phys. Rev. Lett.* **2005**, 94, 054802.
- (14) Durach, M.; Rusina, A.; Stockman, M. I.; Nelson, K. *Nano Lett.* **2007**, 7, 3145–3149.
- (15) Gramotnev, D. K.; Bozhevolnyi, S. I. *Nat. Photonics* **2010**, 4, 83–91.
- (16) Stockman, M. I.; Faleev, S. V.; Bergman, D. J. *Phys. Rev. Lett.* **2002**, 88, 067402.
- (17) Schnell, M.; Garcia Etxarri, A.; Huber, A. J.; Crozier, K.; Aizpurua, J.; Hillenbrand, R. *Nat. Photonics* **2009**, 3, 287–291.
- (18) Aeschlimann, M.; Bauer, M.; Bayer, D.; Brixner, T.; Garcia de Abajo, F. J.; Pfeiffer, W.; Rohmer, M.; Spindler, C.; Steeb, F. *Nature* **2007**, 446, 301–304.
- (19) Gjonaj, B.; Aulbach, J.; Johnson, P. M.; Mosk, A. P.; Kuipers, L.; Lagendijk, A. *Nat. Photonics* **2011**, 5, 360–363.
- (20) Volpe, G.; Molina-Terriza, G.; Quidant, R. *Phys. Rev. Lett.* **2010**, 105, 216802.
- (21) Volpe, G.; Cherukulappurath, S.; Juanola Parramon, R.; Molina-Terriza, G.; Quidant, R. *Nano Lett.* **2009**, 9, 3608–3611.
- (22) Koenderink, A. F.; Hernández, J. V.; Robicheaux, F.; Noordam, L. D.; Polman, A. *Nano Lett.* **2007**, 7, 745–749.
- (23) Lemoult, F.; Lerosey, G.; de Rosny, J.; Fink, M. *Phys. Rev. Lett.* **2010**, 104, 203901.
- (24) Kao, T. S.; Jenkins, S. D.; Ruostekoski, J.; Zheludev, N. I. *Phys. Rev. Lett.* **2011**, 106, 085501.
- (25) Papasimakis, N.; Fedotov, V. A.; Fu, Y. H.; Tsai, D. P.; Zheludev, N. I. *Phys. Rev. B* **2009**, 80, 041102.
- (26) Plum, E.; Tanaka, K.; Chen, W. T.; Fedotov, V. A.; Tsai, D. P.; Zheludev, N. I. *J. Optics* **2011**, 13, 055102.
- (27) Kurter, C.; Abrahams, J.; Anlage, S. M. *Appl. Phys. Lett.* **2010**, 96, 253504.
- (28) Fedotov, V. A.; Papasimakis, N.; Plum, E.; Bitzer, A.; Walther, M.; Kuo, P.; Tsai, D. P.; Zheludev, N. I. *Phys. Rev. Lett.* **2010**, 104, 223901.
- (29) Soares, B. F.; Jonsson, F.; Zheludev, N. I. *Phys. Rev. Lett.* **2007**, 98, 153905.