Towards the lasing spaser: controlling metamaterial optical response with semiconductor quantum dots

E. Plum,[1,*] V. A. Fedotov,[1] P. Kuo,[2] D. P. Tsai,[3] and N. I. Zheludev[1,]**

[1] Optoelectronics Research Centre, University of Southampton, SO17 1BJ, United Kingdom
[2] Institute of Physics, Academia Sinica, Taipei, 11529, Taiwan
[3] Department of Physics, National Taiwan University, Taipei, 10617, Taiwan
** erp@orc.soton.ac.uk*
www.nanophotonics.org.uk/niz/

Abstract: We report the first experimental demonstration of compensating Joule losses in metallic photonic metamaterial using optically pumped PbS semiconductor quantum dots.

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Metallic Joule losses in nano-structured metamaterials are the main obstacle in achieving optical negative index media and narrow resonance frequency selective surfaces for photonic applications. Several schemes have been suggested to overcome these losses including using gain media and parametric processes [1–9]. In particular it has been shown theoretically that through the local-field amplification mechanism a very small amount of gain can strongly

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change absorption and transmission of certain metamaterials [10,11] to the extend of creating conditions for a metamaterial lasing device, the "lasing spaser", a metamaterial version of the spaser [12]. In this paper for the first time we demonstrate experimentally that hybridization of a metamaterial with a layer of semiconductor quantum dots has a profound effect on its optical properties: first, a layer of quantum dots deposited on a metamaterial red-shifts its resonances, and second, when the quantum dots are optically pumped the resonant transmission is modified in a way corresponding to the reduction of Joule losses, thus providing the first step towards the demonstration of a metamaterial gain device and the lasing spaser.

Metamaterials supporting trapped-mode resonances, i.e. current oscillations weakly coupled to free space and thus exhibiting low radiation losses are of special interest from the standpoint of loss control. In photonic trapped-mode metamaterials the resistive Joule loss is the main mechanism of dissipation as radiation losses are small and may be controlled by design. Arrays of asymmetrically split ring resonators belong to this class of metamaterials where the quality factor of the trapped-mode resonance may be controlled by the degree of asymmetry of the split [13]. Moreover, strong interaction between the magnetic moments of the oscillating trapped-mode currents in arrays of asymmetrically split rings offer the intriguing opportunity of creating a coherent source of optical radiation, the lasing spaser, fueled by plasmonic current oscillations. In this case the gain substrate supporting the rings is the source of energy for the lasing spaser device.

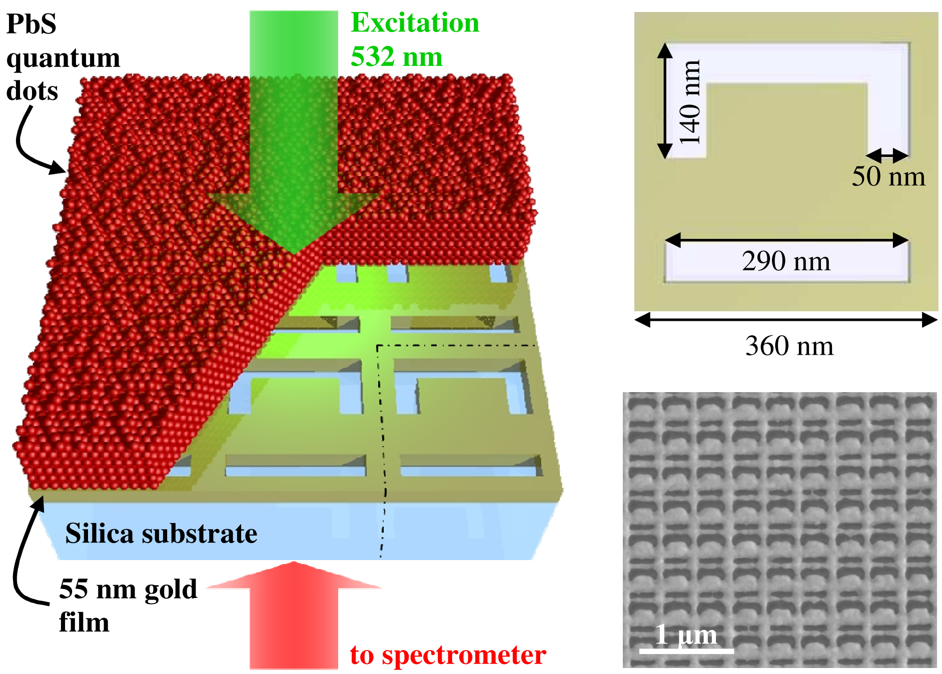

Fig. 1. Photonic metamaterial hybridized with semiconductor quantum dots. The insets show the metamaterial unit cell and an SEM image of the actual metamaterial structure.

In our experiment reported here we used a négative metamaterial array of split ring slits. The ring slits were cut from a 55 nm thick gold film supported by a silica substrate using focused ion beam milling (see Fig. 1). The unit cell of the structure (360 x 360 nm^2) contained a square "ring" with the following design parameters: the ring was 290 x 290 nm^2 in size and had a 100 nm wide asymmetric split. The overall size of the metamaterial array was 45 x 45 μm^2.

Transmission properties of the metamaterial array were characterized using a microscope-based spectrophotometer by CRAIC Technologies. Optical characteristics of the structure are

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polarization-sensitive. In a positive (complementary) design consisting of wire rings [13] the trapped mode would be excited by light polarized along the split. However, as dictated by the Babinet principle, in a négative structure of slits the trapped-mode resonance will be excited with the polarization perpendicular to the direction of the split. Indeed, according to Fig. 2(a) which shows transmission spectra of the array for two perpendicular polarizations, the transmission spectrum is featureless for light polarized along the split, while a resonant dip in transmission at 860 nm corresponds to the excitation of the trapped mode for the perpendicular polarization. While for microwave metamaterials consisting of a metal plate perforated with asymmetric ring slits the trapped mode resonance occurs when the wavelength is twice the average slit length [14], here it is slightly red-shifted due to the presence of the silica substrate and relative increase of the slit width.

To study the role of optical gain on characteristics of the metamaterial it was functionalized with semiconductor quantum dots (QDs). We used quantum dots with a 3.2 nm diameter PbS-core surrounded by a 2 nm thick shell of ligands, which were deposited on the metamaterial array as a suspension in toluene and then dried. The density of quantum dots was determined from the QD molar extinction coefficient (6×10^4 L mol^{-1} cm^{-1}) in control measurements on the glass substrate: the strength of exciton resonant absorption at 1050 nm corresponds to a density of quantum dots of approximately $4 \cdot 10^6$ μm^{-2} in a layer of approximately 1 μm thickness, see 2(c).

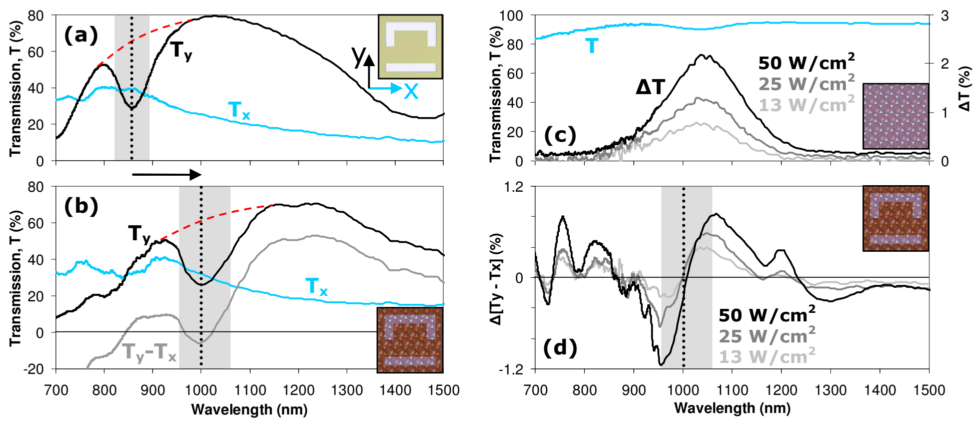

Fig. 2. Transmission spectra of (a) the metamaterial before deposition of quantum dots and (b) the metamaterial after deposition of PbS quantum dots. Blue lines correspond to x-polarization, black lines correspond to y-polarization and a differential spectrum T_y-T_x is shown in gray. The trapped-mode resonance for y-polarization is marked by a gray shaded region. (c) Transmission spectrum of PbS quantum dots on a glass substrate (blue line). The resonant curves show the change ΔT of the transmission signal due to optically pumping the quantum dot layer on the glass substrate. (d) Difference between pump-induced changes of the transmission signal for x and y-polarizations for different levels of pumping.

Resulting from deposition of the quantum dot layer we observed substantial red shift of the transmission spectrum: the trapped-mode resonance moved from 860 nm to 1000 nm, i.e. in a position overlapping with the emission peak of the QDs at 1050 nm. We argue that the red shift results from the shortening of the excitation wavelength due to increased effective permittivity of the dielectric environment. In addition we observed broadening of the resonance from about 70 nm to 105 nm, which is explainable by the additional resonant absorption losses brought about by the QDs.

The influence of the gain was studied when the QDs were optically pumped at 532 nm with intensities of up to 50 W/cm^2 using a frequency doubled CW YAG laser. The pump beam was delivered and concentrated by the microscope objective to a diameter of about 100 μm on the quantum dot layer, while the probe light was collected from the central 42 x 42 μm^2 area of the exposed metamaterial. We first investigated transmission of an optically pumped

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quantum dot layer on a glass substrate and saw a steady increase ΔT of the transmitted signal with increasing pump intensity which we attribute to the quantum dot luminescence (see Fig. 2(c)). Optical pumping of the quantum dots leads to a pronounced modification of the metamaterial's transmission spectrum at the trapped-mode resonance. This modification takes place on the background of QD luminescence. In order to separate the transmission spectrum modification from intensity-dependent quantum dot luminescence we present the pump-induced change $\Delta(T_y - T_x)$ of the differential transmission signal for two perpendicular polarizations (see Fig. 2(d)). Indeed, the luminescence signal does not depend on the polarization state of light used to probe transmission of the metamaterial and therefore is eliminated in the differential spectra. Thus, any observed difference $\Delta(T_y - T_x)$ is metamaterial specific and is due to gain. It must arise from strong interaction between the pumped quantum dot film and surface plasmon modes excited on the metamaterial surface, similarly to coupling observed between plasmonic field localizations and molecules in other periodic nanostructures [15,16]. The negative transmission peak seen in Fig. 2(d) which deepens with increasing pump intensity corresponds to a modification of the trapped-mode resonance of the metamaterial in the presence of gain provided by the optically pumped quantum dots. We argue that this is related to an increase of the quality factor of the trapped-mode resonance due to gain-compensated Joule losses in the metamaterial. Indeed, this observation perfectly corroborates with the results of numerical modelling of the wire split-ring array of the lasing spaser on a gain substrate below the lasing threshold [10]: the results imply narrowing and deepening of the resonance in reflection mode, so that following Babinet's principle the same behavior in general should be expected in transmission for the complimentary slit ring array.

In summary we have provided the first experimental evidence that presence of optically pumped semiconductor quantum dots compensates Joule losses in metallic metamaterial arrays in the optical part of the spectrum. This is a very substantial step towards demonstrating a lasing spaser device. Lasing will require much higher levels of gain which could be achieved by using nanosecond optical pump pulses and/or lowering the sample temperature.

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