Lasing spaser

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In 2003 Bergman and Stockman introduced the spaser, a quantum amplifier of surface plasmons by stimulated emission of radiation[1]. They argued that by exploiting a metal/dielectric composite medium it should be possible to construct a nanodevice, where a strong coherent field is built up in a spatial region much smaller than the wavelength[1,2]. V-shaped metallic structures, combined with semiconductor quantum dots, were discussed as a possible realization of the spaser[1]. Here we introduce a further development of the spaser concept. We show that by combining the metamaterial and spaser ideas one can create a narrow-diversion coherent source of electromagnetic radiation that is fuelled by plasmonic oscillations. We argue that a two-dimensional array of a certain class of plasmonic resonators supporting coherent current excitations with high quality factor can act as a planar source of spatially and temporally coherent radiation, which we term a 'lasing spaser.'

In the lasing spaser, identical plasmonic resonators impose the frequency at which the device will lase. They draw energy from a supporting gain substrate. This combination of artificial classical electromagnetic resonators plays the role of the active medium in the lasing spaser, just as an assembly of essentially quantum inversely populated atoms plays the same role in a conventional laser. In a conventional laser the direction of emission is dictated by the external resonator, and its coherence is underpinned by the stimulated emission of atoms in the gain medium. In the lasing spaser the direction of emission is normal to the plane of the array, where strong trapped-mode currents in the plasmonic resonators oscillate in phase. The coherence in this case arises from the fact that in-phase collective oscillations of antisymmetric currents have the lowest radiation losses and are therefore the easiest to excite. A deliberate small asymmetry in the plasmon resonator, which breaks the non-radiating nature of the trapped-mode oscillation, will allow a fraction of the energy accumulated in current oscillations to be emitted by the array into the free space. This is analogous to the leakage of radiation through the output coupler of a laser resonator. Therefore, in contrast to the optical quantum generator, the lasing spaser is a classical device at all key levels apart from the provision of gain to the substrate active medium.

To create a lasing spaser a special type of metamaterial array of plasmon resonators is required. It should support high-Q (high quality factor) current oscillations that have the lowest total emission losses when all currents in the array oscillate in phase. We call such media coherent metamaterials. We recently demonstrated that a high-quality mode of intense antisymmetric current oscillations may be excited in split-ring resonators with weak asymmetry (ASRs)[3]. Strong oscillations in the rings will build up and exhibit long decay time only if the ring asymmetry is weak and the resonators are arranged into a regular two-dimensional array. This is because the radiation losses associated with the electric and magnetic dipole emission of the oscillating antisymmetric currents are cancelled if the resonators are placed in an infinite regular array, as illustrated in Fig. 1. Thus, the high-Q resonator is formed not by a single ASR plasmonic resonator, but by the entire array. Weak coupling of this current mode to free space occurs only due to the asymmetry in the split ring and may be controlled by design (smaller asymmetry gives lower coupling and higher Q-factor). The behaviour of the weakly asymmetric split-ring arrays is in sharp contrast with that of metamaterials, where radiation losses are strong and the response depends weakly on mutual interactions of individual elements of the structure. We argue that laser action fuelled by trapped-mode spaser current oscillations can be achieved by making use of the coherent nature and high-Q feature of the oscillations in an array of ASRs and will result in light emission with high spatial coherence.

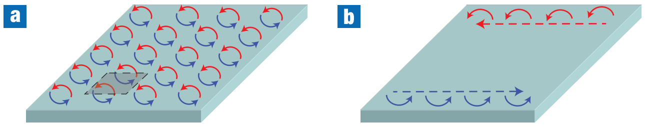

Figure 1 Schematic showing why an array of antisymmetric currents does not radiate. a, Electric and magnetic dipole emissions of a pair of opposite oscillating currents (inside the dashed line frame) in the split-ring array are cancelled. **b**, All such current pairs can be removed from the metamaterial structure without affecting its radiation in the far-field, and in a finite array only peripheral currents cannot be cancelled. With increasing size of array, losses due to peripheral currents become increasingly negligible, leading to higher quality factors of the trapped-mode resonances.

If the array of resonators is in contact with a gain medium, for example when it is supported by a thin slab of gain material (see Fig. 2), then radiation losses and Joule losses in the metal can be overcome. Various gain media such as optically and electrically pumped semiconductor structures or quantum-dot-doped dielectrics may be suitable for this purpose. We show below that on reaching the threshold value of gain, the intensity of the resonant wave reflected and transmitted through the structure increases dramatically. By combining a thin layer of a gain medium with a high Q-factor ASR array, it is possible to achieve orders of magnitude enhancement of single-pass amplification

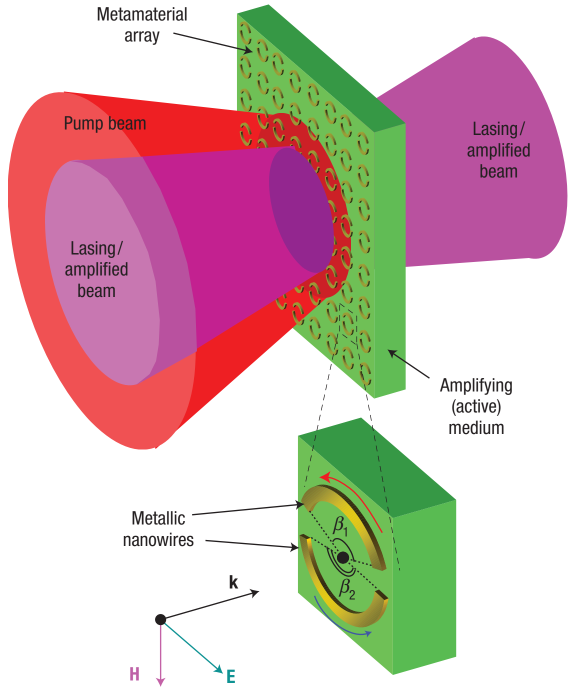

Figure 2 Lasing spaser. The structure consists of a gain medium slab (green) supporting a regular array of metallic asymmetric split-ring resonators. The dashed box indicates an elementary translation cell of the array, and the arrows along the arcs of the ring in the enlarged schematic of the cell illustrate the antisymmetric currents of plasmonic oscillations. In-phase plasmonic oscillation in individual resonators leads to the emission of spatially and temporarily coherent light propagating in the direction normal to the array.

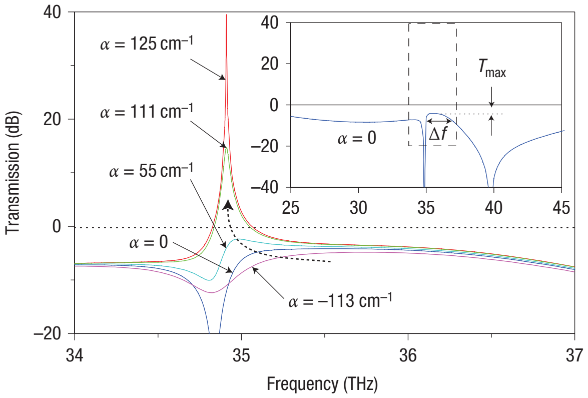

Figure 3 Transmission spectra of the mid-IR resonator array. Spectra of the planar ASR metamaterial are provided in the vicinity of the trapped-mode transmission resonance for different values of gain α. The dashed arrow follows the transformation of the transmission resonance. The inset presents the transmission spectrum of the metamaterial with no losses/optical gain ($\alpha = 0$) over a much wider frequency range, the dashed box indicating the spectral domain that is covered by the main plot.

in comparison with the amplification of the bare gain medium layer.

We illustrate this concept by providing a numerical analysis of amplification in the array of ASRs combined with a gain dielectric substrate. Two cases are considered. In the first case, resonant amplification is achieved in the mid-infrared (mid-IR) part of the spectrum (at a wavelength of about 8 μm), where Joule losses in the metals are neglected and only losses and gain in the isotropic dielectric substrate are taken into account. In the second case, we consider amplification at a wavelength of 1.65 μm and take into account Joule losses in the metallic wires. In both cases, losses and gain in the substrate are assumed to be independent of frequency. This simplifying assumption is valid when the metamaterial resonance is narrower than the gain line of the substrate and inhomogeneous spectral hole burning is insignificant. We also assume no depletion of gain in all operational regimes.

The unit cell of the modelled metamaterial structures is presented in Fig. 2. It consists of a planar subwavelength asymmetric metallic ASR horizontally split into two wire segments of different lengths corresponding to arc angles β_1 and β_2, where the ends of the segments are separated by equal gaps. The ASR is brought into direct contact with a dielectric slab, which could be a gain medium supporting the array. Arrays of such metal structures can be manufactured by e-beam or photolithography.

Figure 3 shows the transmission characteristics of the infrared ASR array for different levels of bare substrate gain presented in terms of the gain coefficient α. For negative values of α (lossy substrate) the metamaterial attenuates electromagnetic radiation. Gain in the substrate exceeding a threshold value of $\alpha_{th} = 70\ \mathrm{cm}^{-1}$ is sufficient to overcome losses at a frequency of about 35 THz ($\lambda = 8.4$ μm), and signal attenuation in the metamaterial then becomes signal amplification (see Fig. 4). This level of gain corresponds to small-signal amplification of only ~2.7% in a 2-μm-thick active layer of the bare substrate. A further increase in the bare substrate gain leads to a rapid increase of resonant amplification in the metamaterial, reaching a level of 42 dB (a factor of $\sim 1.6 \times 10^4$) at $\alpha = 125\ \mathrm{cm}^{-1}$. In a bare film such levels of gain will only lead to small-signal amplification of about 5%. As well as the increase in bare substrate gain, the width of the amplified spectrum collapses from 1.2 THz at zero gain to $\Delta\nu = 2$ GHz at the amplification maximum. A further increase in the bare substrate gain leads to a rapid decrease in small-signal amplification of the metamaterial structure. This is because gain broadens the resonance in the same way that losses broaden absorption resonances, and achieving antiphase oscillation of currents in the split-ring arcs of the plasmonic resonator becomes more difficult as radiation losses increase. Such behaviour may also be found in an externally driven ensemble of three coupled lossy mechanical oscillators, where an increase of driving force leads to an increase of the amplitudes of their oscillations until a critical value of driving force is reached, after which the amplitudes decrease. In an infinite array, the width and magnitude of the amplification peak are only limited by radiation losses, and are controlled by the asymmetry of the split-ring resonators. In a realistic case the finite size of the array (see Fig. 1) and fabrication tolerances will limit amplification.

Similar analysis has also been performed for a structure resonating at 1.65 μm, where losses in the metal increase α_{th} to $\sim 1{,}800\ \mathrm{cm}^{-1}$ and reduce the maximum level of achievable amplification to about 35 dB (a factor of $\sim 3.2 \times 10^3$) (Figs 4 and 5). Here amplification peaks at $\alpha = 2{,}550\ \mathrm{cm}^{-1}$, which corresponds to amplification of 5.5% in the bare substrate film, and the spectral width of the amplification resonance reduces from 3 THz to about $\Delta\nu = 500$ GHz.

We argue that the current oscillation will self-start coherently in all the rings of the array if sufficient gain is provided. This is because the radiation losses in the metamaterial are at a

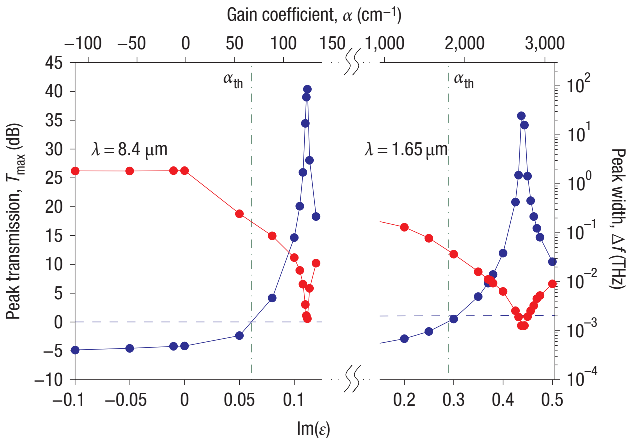

Figure 4 Transmission properties of near- and mid-IR resonator arrays. Small-signal amplification (blue) and spectral width (red) of the resonant transmission peak as a function of gain level in the substrate.

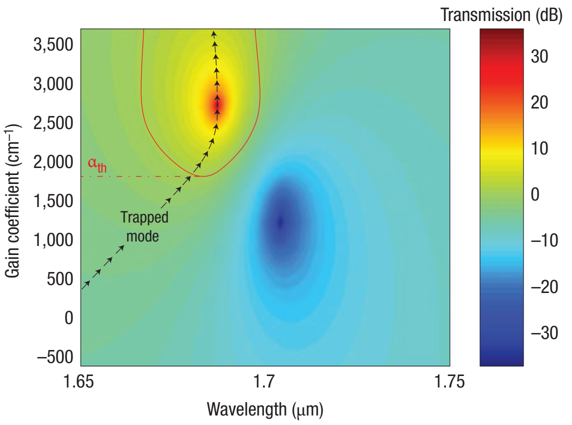

Figure 5 Transmission spectra of the near-IR resonator array. Spectra of the planar ASR metamaterial are provided in the vicinity of the trapped-mode transmission resonance corresponding to different values of bare substrate gain α. Solid contour: region of unity transmission. The line of arrows shows the evolution of the trapped-mode resonance frequency with increase of α.

minimum for a collective mode formed by in-phase oscillations of the antisymmetric currents in all the rings of the array, and any other modes will scatter strongly, having much lower quality factors[3]. Such oscillations will produce a spatially and temporally coherent diffraction-limited beam of optical emission normal to the array, transforming an optical amplifier into a lasing spaser. This will happen without the need for an external resonator; coherence and narrow diversion of the output will be ensured by the low-loss condition. From the properties of the metamaterial array as an amplifier we can expect that, on reaching a threshold gain, the system will start lasing coherently across the whole array. With increasing gain the output intensity will increase rapidly and its spectrum will narrow dramatically. In reality, the output intensity of the lasing spaser is likely to be limited by saturation in the gain medium and heat management problems.

The small scattering losses of the current in the metamaterial array make the levels of threshold gain and gain needed to achieve a peak amplification of 35–40 dB practically attainable. For instance, quantum-well structures can provide gain of the order of 1×10^3 cm^{-1} (ref. 4), which is similar to the threshold value required for an ASR array operating at 1.65 μm. Furthermore, quantum cascade amplifiers can readily provide the gain values needed in the mid-IR case, because attainable gain coefficients in this wavelength range exceed 100 cm^{-1} (ref. 5). This easy-to-achieve threshold gain condition gives a key advantage over recent suggestions to combine amplifying media with nanoshell[6] and horseshoe resonant[7] elements to create a compact plasmonic nanolaser. In such arrangments the high dipole radiation losses of the plasmonic resonator make the threshold gain level difficult to achieve.

The lasing spaser allows high amplification and lasing in a very thin layer of material with a more modest gain level, making it a very practical proposition. The thin-layer geometry is a desirable feature for some highly integrated devices and from the point of view of heat management and integration. Here the amplification/lasing frequency is determined by the size of the ring and may be tuned to match luminescence resonances in a large variety of gain media. This therefore makes the lasing spaser a generic concept for many applications. Finally, the ring currents in the metamaterial array can be seen as classical analogues of magnon quasiparticles, and the striking similarity between the coherent regime of the lasing spaser and Bose–Einstein condensation of magnons under pumping should be considered[8].

METHODS

In the mid-IR version of the planar metamaterial the unit cell had a lateral dimension of 1.5 μm, the split ring had a radius and linewidth of 0.6 and 0.05 μm, respectively, and $\beta_1 = 160^\circ$ and $\beta_2 = 151^\circ$. The thickness of the active layer on the support substrate is 2 μm and its dielectric constant (real part) $\varepsilon' = 10.9$. The optical response of such a metamaterial structure was analysed in the 20–50 THz frequency range (6–15 μm) using the method of moments. This numerical method involves solving an integral equation for the surface currents induced in the metallic pattern by the incident electromagnetic wave, then calculating the scattered fields produced by the currents as a superposition of partial spatial waves. The metallic pattern is therefore treated as a very thin perfect conductor (which is acceptable for most metals in the mid-IR region), and the gain (losses) in the substrate is introduced through the imaginary part of its dielectric constant ε and assumed to be isotropic.

In the metamaterial structure designed for the near-IR domain, the diameter of the ASR resonator was 140 nm, with a unit cell of 210 × 210 nm. The angular lengths of the metallic wire segments corresponded to angles $\beta_1 = 160^\circ$ and $\beta_2 = 125^\circ$, and they had cross-sections of 20 × 50 nm. The metal of the nanowires was assumed to be silver with a dielectric constant described by the Drude model. The substrate was 100 nm thick with $\varepsilon' = 9.5$, and gain was introduced through the imaginary part ε'' of the substrate's dielectric constant, which is related to the gain/attenuation coefficient α by $(2\pi/\lambda)\mathrm{Im}(\sqrt{(\varepsilon' + i\varepsilon'')})$. The transmission properties of this active nanostructure were numerically modelled using a true three-dimensional finite-element method for solving Maxwell's equations.

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Author contributions

The idea of the lasing spaser belongs to N.I.Z. who also wrote the paper. S.L.P. and N.P. performed infrared and near-infrared numerical experiments correspondingly. V.A.F. contributed to the analysis of data.

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