Spasers explained

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The spaser is a proposed nanoscale source of optical fields that is being investigated in a number of leading laboratories around the world. If realized, spasers could find a wide range of applications, including nanoscale lithography, probing and microscopy.

ano-optics is now undergoing a period of explosive growth where new ideas, developments and impressive results appear literally on a daily basis. It is concerned with the science of concentrating optical energy into regions with subwavelength dimensions (typically tens of nanometres). Yet despite all this progress, there is still the need for a coherent, intense, ultrafast (with pulse durations down to a few femtoseconds), source of optical energy concentrated to nanoscale areas, similar to the laser but on a much smaller scale. In 2003, David Bergman and I proposed such a source that is based on surface plasmons — the so-called spaser¹ (short for surface plasmon amplification by stimulated emission of radiation) - and researchers are now working to develop and exploit this idea. For example, Nikolay Zheludev and colleagues present their latest ideas regarding spasers on page 351 of this issue².

SURFACE PLASMONS

When introducing the concept of a spaser, it is first useful to explain how it is possible to beat the diffraction limit and focus electromagnetic waves to spots much smaller than a wavelength. The answer lies in the fact that on the nanoscale, optical fields are almost purely electric oscillations at optical frequencies, where the magnetic field component is small and does not significantly participate in the nano-optical physics. The ability of a nanostructured material to support and concentrate such fields is due to the existence of optical modes that are localized on dimensions much smaller than the optical wavelength.



Figure 1 The spasing mechanism. **a**, Schematic of a spaser made from a silver nanoshell on a dielectric core (with a radius of 10–20 nm), and surrounded by two dense monolayers of nanocrystal quantum dots (NQDs). **b**, Schematic of levels and transitions in a spaser. The external radiation excites a transition into electron–hole (e–h) pairs (vertical black arrow). The e–h pairs relax to excitonic levels (green arrow). The exciton recombines and its energy is transferred (without radiation) to the plasmon excitation of the metal nanoparticle (nanoshell) through resonant coupled transitions (red arrows). **c,d**, Field amplitudes, φ , around the nanoshell excited in two different plasmon modes.

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Box 1 Existing methods to nanolocalize optical energy

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- Within the context of SNOM, a single local-field hot spot can be excited either at a metal nanoparticle or a metal 4tip by a laser focus, or by an aperture in a metal film used as a light source.
- A nanoaperture in a metal-covered tapered optical fibre is a source of nanolocalized optical fields.
- Optical irradiation of random nanoplasmonic systems causes the appearance of spontaneous hot spots of local optical fields.
- Concentration of energy in tapered plasmon-polariton waveguides (typically silver or gold cones or

These energy-concentrating modes are surface plasmons (SPs). They are in essence the eigenmodes of a material system that correspond to oscillations, at optical frequencies, of the electron liquid with respect to the crystal lattice. It can be shown that for a system to support SPs, it should contain components with positive and negative dielectric permittivities. A material with negative dielectric permittivity does not support propagation of electromagnetic waves; instead the electromagnetic field decays inside the material within a certain skin depth and most of the incident radiation energy is reflected back. This behaviour is characteristic of a metal, where the skin depth is typically around 25 nm.

But for a nanoparticle with a size smaller than the skin depth, optical fields are able to penetrate its entire volume and drive SP oscillations. It is the skin depth that determines the characteristic length scale in nano-optics and makes the nanoscale (from a few nanometres to a few tens of nanometres) so important.

An SP mode is characterized by its quality factor, *Q*, which is the number of electron oscillations that occur coherently, and during which the mode is able to sustain its phase and accumulate energy from the external excitation field. The best plasmonic metals, for which *Q* typically ranges from 10 to 100, are the noble metals (silver, gold and platinum), aluminium and the alkaline metals. In response to an external excitation, a plasmonic nanoparticle can generate local fields that are enhanced by a factor *Q* with respect to the external field. The wedges) is used to transfer optical energy to the nanoscale.

- Nanofocusing of optical energy by so-called hyperlenses (layered metal-dielectric composite waveguides tapered to nanoscale curvature) is a promising method of nanolocalization.
- Optically nonlinear sources of nanolocalized hot spots are obtained by intense optical excitation of nanotips and nanoparticles.
- Specially designed nanoantennas or nanolenses create hot spots of nanolocalized optical fields.

region occupied by these enhanced fields is determined solely by the size of the nanoparticle. Another route to enhancing the local field is through geometric tricks. For instance, the sharp tip of a cone can create intense local fields (the so-called lightning-rod effect), and is exploited in scanning near-field optical microscopy³ (SNOM).

SPASING ACTION

It is critically important that sources of concentrated, intense optical fields localized on the nanoscale are available. Such sources will have benefits for fundamental nanoplasmonics and its numerous existing and potential applications, including: ultramicroscopy, (such as SNOM); ultrasensitive detection and spectroscopy of chemical and biological objects based on surface-enhanced Raman scattering (SERS); fluorescence imaging with single-molecule sensitivity; hyper-Raman (or two-photon-Raman) capabilities; coupling of light to semiconductor nano- and microstructures; and numerous biomedical applications. The problem of delivering optical energy to the nanoscale is a formidable one because the optical radiation (light) is limited by diffraction, and thus can only be focused into micrometre-sized regions.

Despite the availability of a wide variety of nanoscale optical sources (Box 1), none are ideal. Typically these sources have a halo of background scattered and delocalized optical fields around them, and do not offer the intensity needed to induce nonlinear processes, or the ultrafast speeds required for femtosecond spectroscopy. An ideal nanoscale source would also generate 'dark' optical modes that do not couple to far-field zones. None of the existing nanosources of localized optical fields possess all of these properties, but the spaser that was proposed in 2003 may be just such a source¹.

A spaser is the nanoplasmonic counterpart of a laser, but it (ideally) does not emit photons. It is analogous to the conventional laser, but in a spaser photons are replaced by SPs and the resonant cavity is replaced by a nanoparticle, which supports the plasmonic modes. Similarly to a laser, the energy source for the spasing mechanism is an active (gain) medium that is excited externally. This excitation field may be optical and unrelated to the spaser's operating frequency; for instance, a spaser can operate in the near-infrared but the excitation of the gain medium can be achieved using a UV pulse.

The reason that SPs in a spaser can work analogously to photons in a laser is because their relevant physical properties are the same. First, SPs are bosons: they are vector excitations and have spin 1, just as photons do. Second, SPs are electrically neutral excitations. And third, SPs are the most collective material oscillations known in nature, which implies they are the most harmonic (that is, they interact very weakly with one another). As such SPs can undergo stimulated emission, accumulating in a single mode in large numbers, which is the physical foundation of both the laser and the spaser.

One of the simplest and potentially most promising types of nanoparticles to function as a spaser resonator is a metal-dielectric nanoshell. Such nanoshells have been introduced by Naomi Halas and collaborators and have since found a very wide range of applications^{4,5}. A possible design of a nanoshell-based spaser is illustrated in Fig. 1a. It consists of a silver nanoshell surrounded by a few monolayers of nanocrystal quantum dots6-8 (NQDs). A schematic of the energy levels and transitions in this spaser is shown in Fig. 1b. An electron-hole pair excited by an initial photon from the external excitation field (black arrow in Fig. 1b) relaxes to an excitonic (or possibly multi-excitonic) state due to carrier multiplication⁸ (green arrow in Fig. 1b). In a free NQD the excitons would recombine to form photons. However, when the NQD is sitting on the surface of a resonant nanoparticle, the excitonic

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energy is transferred, without any significant emission of radiation, to the resonant SPs of the nanoparticle (coupled red arrows in Fig. 1b), a process that has a much larger probability by orders of magnitude. The SPs stimulate further transitions in the gain medium, leading to the excitation of more identical SPs in the same SP mode, driving the action of the spaser. Examples of two SP modes that can be excited in the nanoshell are shown in Fig. 1c,d.

DARK MODES

One of the advantages of the spaser compared with existing sources of local fields, which also sets it apart from the laser, is that it can generate dark modes that do not couple to the far-zone optical fields. In other words, a spaser generates coherent, strong local fields, but does not necessarily emit photons. This is a potentially great technological advantage because it offers a source of nanolocalized optical fields that does not emit any background radiation. The source can still act on molecules in its near field and excite their radiation (such as fluorescence and Raman effects) as conventional nano-optical sources do. This is because a small molecule is affected by the field at the point of its position and is blind to the overall (global) symmetry of the entire local field that determines whether the mode is dark or luminous (bright).

Another important advantage of using dark SP modes is that they do not undergo radiative losses. Such losses for the luminous modes would lead to a higher threshold for the spaser operation. Interestingly enough, if the symmetry of the spaser's nanoplasmonic particle is slightly broken, a dark spasing SP mode will become luminous. A collection of such spasers may then start to emit light, just like a laser. This idea has been proposed by Nikolay Zheludev and colleagues on page 351 of this issue², where they suggest using a metamaterial containing a planar array of spasers, each of which has a slightly perturbed symmetry. This array then becomes a very efficient planar laser emitting light normally to its plane.

So what are the ideal operating conditions for a spaser? Well, the NQD packing density around the metal nanoparticle should be as high as possible. And the spectral width of the spasing SP mode should be as small as possible (that is, its dephasing time should be as long as possible). These conditions are best met in the



Figure 2 The electric field provided by the spaser described in ref. 1. Mode electric-field amplitude, *E*, is plotted over the surface of a V-shaped silver nanorod configuration that acts as a spaser resonator.

near-infrared, where the losses of noble metals are at a minimum.

In sharp contrast to the laser, the specific shape of the nanoparticle at the heart of the spasing mechanism does not affect its gain at a given frequency, provided that the spasing SP mode overlaps with the gain medium. This seriously limits the possibilities for engineering favourable spasing conditions. On the other hand, the spaser gain depends on its operating frequency, which can be shifted into the desired spectral range by engineering the geometry of the nanoplasmonic particle, for instance as a nanoshell, as in Fig. 1a.

Spasers can generate pulses of localized optical fields with durations on the femtosecond scale that can be as short as 5 fs. The magnitude of the electric field in the pulse at the spaser surface is of quantum origin¹ and an example is illustrated in Fig. 2. The field amplitude is shown for a spaser whose 'nanoparticle' consists of two silver nanorods connected at one end to form a V-shape. It can be seen that the field forms a hot spot at the tip of the V-shape where its amplitude reaches 10⁸ V cm⁻¹. For a spasing mode containing say 10⁴ SP quanta, the resulting field is extremely strong, approaching atomic values.

The spaser is a nanoplasmonic device with dimensions that are less than the skin depth, and it generates only nanolocalized SPs. There is, however, a spectrum of related effects based on surface plasmon polaritons (SPPs), which are waves localized in the direction normal to a surface that propagate along it for distances much greater than the wavelength. Researchers have reported the stimulated emission of SPPs (ref. 9). Although this is an important step towards the realization of a spaser, in contrast to the spaser, SPPs on a flat metal surface propagate freely and are not subjected to the cavity feedback necessary for lasing (spasing).

In other work, scientists have reported a quantum cascade laser that operates at a frequency of 17 THz, whose resonator is based on surface electromagnetic waves¹⁰. There have also been studies into the different gain media to compensate for optical losses, which could be used in spasers. Researchers have suggested using the effect of gain on the scattering of light from metal nanostructures¹¹. But these experiments used dye molecules that may not be suitable for spasers, owing to the low dipole oscillator strength of the fluorescent transition and problems with achieving sufficiently large molecular concentrations without excited-state quenching. The most promising gain media for spasers remain NQDs at present.

Very intense, ultrafast, temporarily coherent pulses of nanolocalized optical fields will find numerous applications in both fundamental science and engineering. It is one of the unique properties of spasers that the spasing may be dark, producing strong local optical fields that do not emit light by themselves into the far field unless there are excitable molecules at the surface of the spaser. The big advantage of spasers is the possibility of using them to perform background-free spectroscopy. A spaser that is electrically pumped would be particularly valuable, although this possibility still needs to be explored. It may be no exaggeration to say that the spaser, when finally operational, will do for nano-optics what the laser has done for conventional optics.

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Lasing spaser

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In 2003 Bergman and Stockman introduced the spaser, a quantum amplifier of surface plasmons by stimulated emission of radiation¹. 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 spaser¹. Here we introduce a further development of the spaser concept. We show that by combining the metamaterial and spaser ideas one can create a narrowdiversion coherent source of electromagnetic radiation that is fuelled by plasmonic oscillations. We argue that a twodimensional 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 twodimensional 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.

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

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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.

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 μ m), 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 μ m 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



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.

(lossy substrate) the metamaterial attenuates electromagnetic radiation. Gain in the substrate exceeding a threshold value of $\alpha_{\rm th} = 70 \ {\rm cm}^{-1}$ is sufficient to overcome losses at a frequency of about 35 THz ($\lambda = 8.4 \,\mu\text{m}$), and signal attenuation in the metamaterial then becomes signal amplification (see Fig. 4). This level of gain corresponds to small-signal amplification of only \sim 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 $\alpha = 125$ 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 $\Delta v = 2$ 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 $\alpha_{\rm th}$ to ~1,800 cm⁻¹ and reduce the maximum level of achievable amplification to about 35 dB (a factor of ~3.2 × 10³) (Figs 4 and 5). Here amplification peaks at $\alpha = 2,550$ 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

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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.

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 factors³. 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 μ m. 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 nanoshell⁶ and horseshoe resonant⁷ 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



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 .

coherent regime of the lasing spaser and Bose–Einstein condensation of magnons under pumping should be considered⁸.

METHODS

In the mid-IR version of the planar metamaterial the unit cell had a lateral dimension of 1.5 μ m, the split ring had a radius and linewidth of 0.6 and 0.05 μ m, respectively, and $\beta_1 = 160^{\circ}$ and $\beta_2 = 151^{\circ}$. The thickness of the active layer on the support substrate is 2 μ m 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 μ m) 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 $(2\pi/\lambda) \text{Im}(\sqrt{(\varepsilon' + i\varepsilon'')})$. 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.I.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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