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.

Nano-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 spaser[1] (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 issue[2].

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.

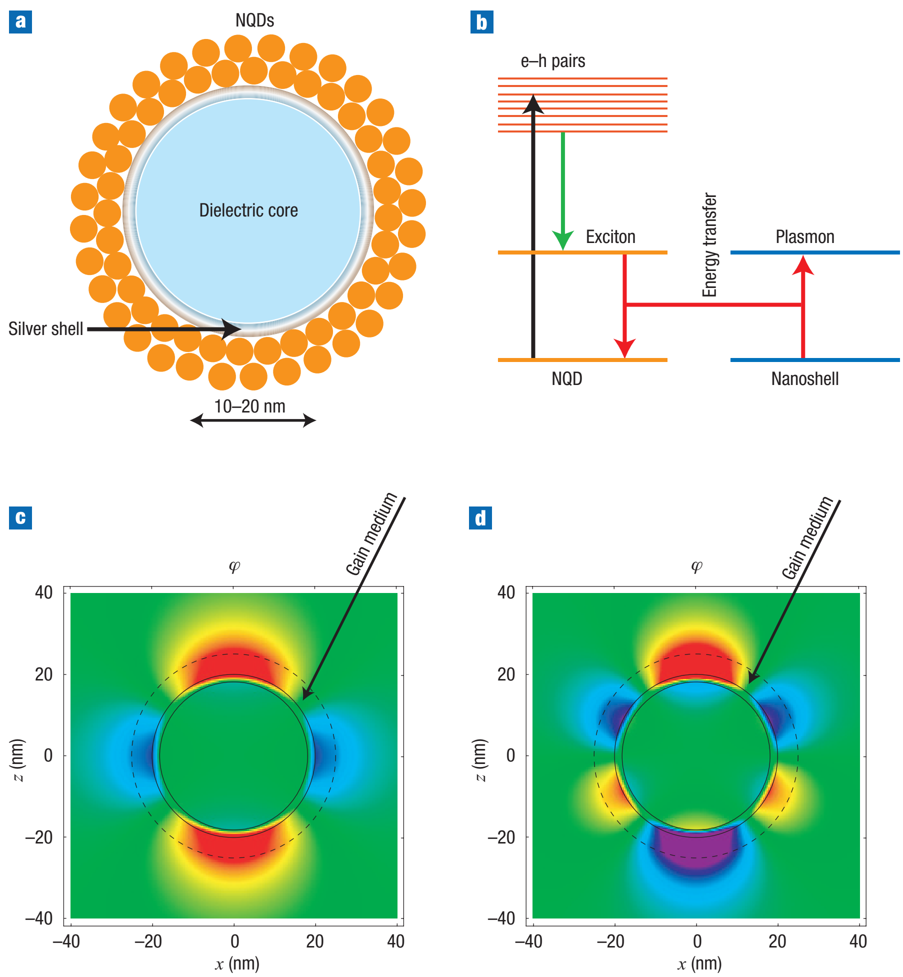

Figure 1 The spasing mechanism. **a**, Schematic of a spaser made from a silver nanoshell on a dielectric core (with a radius of 10–20 nm), and surrounded by two dense monolayers of nanocrystal quantum dots (NQDs). **b**, Schematic of levels and transitions in a spaser. The external radiation excites 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.

Box 1 Existing methods to nanolocalize optical energy

- 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 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.

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 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 microscopy[3] (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 source[1].

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 dots[6–8] (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 multiplication[8] (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 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 issue[2], 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 near-infrared, where the losses of noble metals are at a minimum.

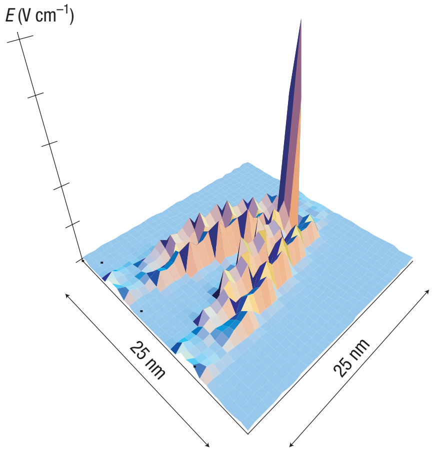

Figure 2 The electric field provided by the spaser described in ref. 1. Mode electric-field amplitude, *E*, is plotted over the surface of a V-shaped silver nanorod configuration that acts as a spaser resonator.

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 origin[1] 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 10^8 V cm^{-1}. For a spasing mode containing say 10^4 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 waves[10]. 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 nanostructures[11]. 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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