PLASMONIC LASERS

A sense of direction

Lattice surface plasmons enable large-area unidirectional emission of coherent light generated at deep subwavelength scales.

Jorge Bravo-Abad and Francisco J. García-Vidal

When the laser was first developed in the 1950s it was labelled by some as "a solution looking for a problem"[1]. It is now, of course, recognized as one of the most important inventions of the twentieth century, and in the space of only a few decades, the technology has progressed from basic centimetre-scale ruby lasers to mass-produced semiconductor lasers. Recently, researchers have begun to turn their attention to the development of lasers with nanoscale dimensions and in the past ten years the field of nanolasers has made a number of important fundamental and applied breakthroughs. However, an essential ingredient has been missing from the technology: the ability to emit light in the form of a unidirectional beam. Writing in *Nature Nanotechnology*, Teri Odom and colleagues at Northwestern University now show that lattice plasmons in arrays of plasmonic nanocavities can be used to create such light beams[2].

In their most basic realization, lasers are based on two principles[3]. First, a photon travelling through a laser medium generates a second photon by stimulating a radiative decay of an emitter. Common emitters are a dye in an excited state or an electron–hole pair in the case of a semiconductor. The repetition of this stimulated emission process leads to optical amplification, and eventually, lasing action. Second, these photon multiplication events are fully coherent, that is, all emitted photons have the same frequency, direction and phase as the first photon. However, if light were allowed to propagate in any arbitrary direction in the gain medium, it would emerge with a non-well-defined directionality. To overcome this problem, most centimetre-scale lasers use an active medium that is enclosed between two highly reflective mirrors (a Fabry–Perot cavity), so that only electromagnetic waves with a specific directionality and phase are selected and lased out when they reach a certain intensity threshold level (Fig. 1a).

The development of photonic crystals — periodic optical nanostructures — has led to lasing action at much smaller scales. The versatility of photonic crystals to achieve simultaneous spectral and spatial electromagnetic mode engineering can be exploited for lasing action in two different ways. By introducing a defect in an otherwise perfectly periodic photonic crystal lattice, a microscopic counterpart to a Fabry–Perot cavity can be created[4]. Alternatively, instead of relying on localized cavity modes, the multidirectional distributed feedback effect occurring at frequencies close to the band edges in a defect-free photonic crystal slab can be used. The slow group velocity that characterizes these modes enables control

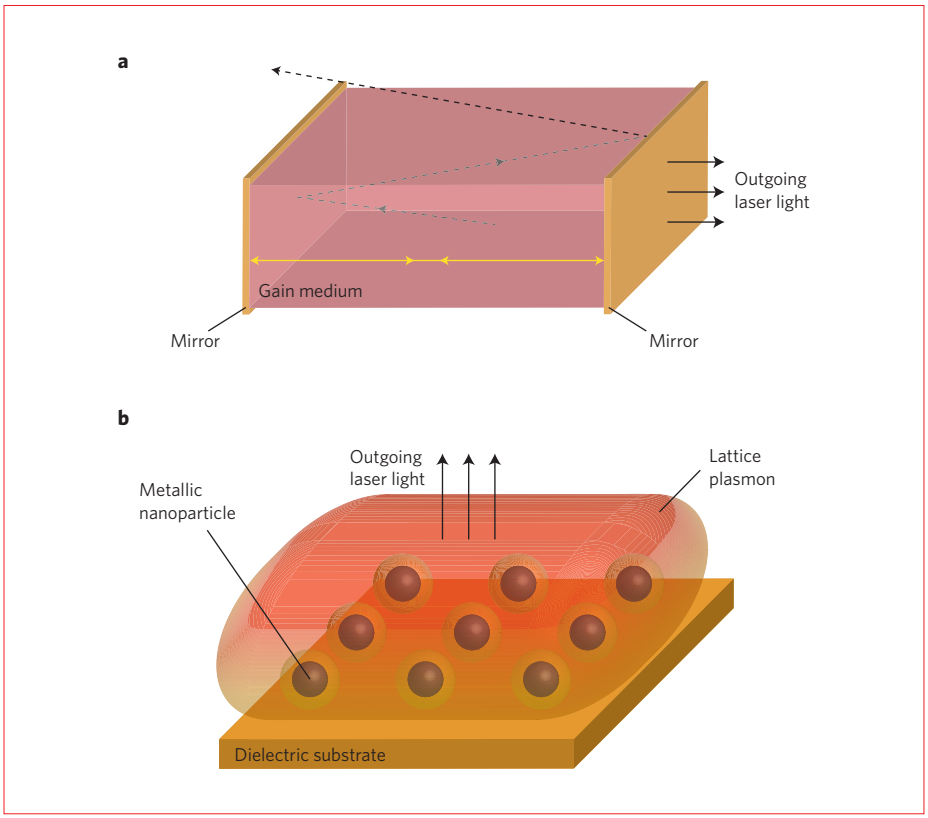

Figure 1 | Origin of directional emission. **a**, With a conventional Fabry–Perot cavity laser, optical waves with arbitrary propagation directions are generated in the gain medium. The ones that propagate in a direction that is not perfectly perpendicular to the mirrors escape from the cavity (black dashed line). Only the waves bouncing back and forth along the cavity axis (solid yellow arrows) spend enough time in the cavity to produce lasing action, therefore, accurately setting the directionality of the outgoing laser emission. **b**, Large-area lattice plasmon lasers are formed from a periodic array of metallic nanoparticles (grey spheres) patterned on a dielectric substrate and surrounded by a gain medium. The nanoparticles are tailored to support localized surface plasmon resonances. On optical pumping, these localized surface plasmons enable laser light to be produced in the nanoscale regions around each nanoparticle (halo around nanoparticles). Then, all generated laser light is 'collected' by lattice plasmons (extended modes that arise due to the electromagnetic coupling among nanoparticles). Lattice plasmons emit the generated laser light in the form of a highly directional beam. If it was not for the presence of lattice plasmons the laser light generated around each nanoparticle would be scattered in all directions.

of the lasing direction and polarization over large areas[5] (which represents the basic feature of these photonic-crystal surface-emitting lasers).

More recently, nanoplasmonics has offered the possibility of accessing deep-subwavelength regimes because surface plasmons are not restricted by the diffraction limit. The large local electric field associated with surface plasmons leads to a significant increase of light–matter interaction strength, which, in turn, dramatically boosts the optical amplification provided by a gain medium placed in the proximity of the plasmonic field. This allows the intrinsic losses typical of metals at optical frequencies to be fully compensated, and eventually, induce a self-sustained laser oscillation in the system[6–9]. However, despite their many advantages, plasmonic lasers have one fundamental drawback: the large wavevector difference between surface plasmons and electromagnetic radiation in free space, which means that when the generated light emerges it is scattered in all directions.

Odom and colleagues have now overcome this problem by merging the advantages of photonic crystals and plasmonics. They first pattern a two-dimensional periodic array of gold and silver metallic nanoparticles on top of a glass substrate and then immerse it in an active polymer medium, which is optically pumped at 800 nm. The electromagnetic coupling of the localized surface plasmon resonances from many nanoparticles gives rise to coherent lattice plasmon modes, which, in contrast to the typical modes of a photonic crystal, feature subwavelength lateral confinement without needing any additional waveguiding structure. These photonic modes display low group velocity well into the deep-subwavelength regime, which allows directional lasing emission over large areas. In simple terms, these extended modes collect all the lasing light produced at the nanoscale by each nanoparticle and emit it by a distributed feedback mechanism to the external world in the form of a directional beam (Fig. 1b). This allows a remarkably small angular divergence (<1.5°) of the emitted laser light to be achieved. Very recently, similar large-area plasmonic lasing has also been observed, but in this case using a holey metal film placed in the proximity of an optically pumped semiconductor gain layer[10].

The approach developed by Odom and colleagues also has the potential for further exploitation. From a fundamental standpoint, it is possible to imagine ways in which these systems could be used, for example to generate entangled light–matter states for quantum information applications. Furthermore, the systems could be useful in characterizing the coherent energy coupling between lattice surface plasmons and exciton quantum emitters. From a more applied perspective, the work could lead to the development of novel large-area devices that take advantage of the strong electric-field confinement of surface plasmons. Examples include more efficient energy-harvesting nanostructures, scalable Raman or fluorescence detection devices, and all-optical switches operated at ultralow power levels. However, several hurdles must first be overcome. One of the most critical is developing efficient electrically pumped plasmonic lasers. This will certainly require a number of innovative conceptual advances, but considering the speed with which lasing technology has already developed, it might not be that long before this is achieved. ❐

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SINGLE-MOLECULE DETECTION

Breaking the concentration barrier

The combination of a plasmonic nanoantenna and a nanoaperture has merged fluorescence enhancement and spatial confinement to enable single-molecule detection at biologically relevant concentrations.

Philip Tinnefeld

Detecting a single molecule is the ultimate goal in chemical analysis and constitutes the basis of innovative technological solutions in the biosciences, and for DNA sequencing in particular[1]. To detect a single molecule the observation volume must host only one molecule of interest during the measurement. The maximum concentration that can be detected by optical means is constrained by the size of the observation volume, which in turn is limited by diffraction. This implies that it is impossible to carry out single-molecule detection at concentrations exceeding the picomolar to low-nanomolar range, because the observation volume becomes populated by more than one fluorescent molecule as concentration increases. This concentration barrier has hampered the widespread use of single-molecule techniques for studying biomolecular interactions, such as protein–protein and enzyme–substrate interactions, whose dissociation constant is in the micromolar range (that is, reaction partners have to be present at that concentration to obtain a significant fraction of the biomolecular complex). Now, writing in *Nature Nanotechnology*, Jérôme Wenger and colleagues at Aix-Marseille Université in France, alongside collaborators at the ICFO-Institut de Ciencies Fotoniques and ICREA-Institució Catalana de Recerca i Estudis Avançats in Spain, describe a system that brings two complementary nanophotonic approaches together to achieve single-molecule detection with a high signal-to-noise ratio at a micromolar concentration[2].

In recent years, researchers have tried to lower the detection volume beyond the limit imposed by diffraction and overcome the concentration barrier of optical single-molecule detection by using two alternative nanophotonic approaches. In the first, depicted in Fig. 1a, light is confined to the near-field using nanoapertures of subwavelength dimensions (typically