A rocky road to plasmonic lasers

To the Editor — Miniaturization of lasers down to nanoscale volumes well below the classical diffraction limit is of major interest for numerous applications, ranging from all-optical computing to biomedical imaging. To realize such nanolasers, resonators comprising metallic nanostructures that support highly confined surface plasmon polariton (SPP) modes are very promising. However, demonstrating plasmonic nanolasers remains challenging, despite massive research activity in this rapidly evolving field. Furthermore, there is little agreement within the community about what is required to prove unambiguously that a structure represents a plasmonic laser, or 'spaser'. Within the plasmonics community (and in this Correspondence) the term spaser is used as synonym for a nanoscale laser making use of SPPs. However, according to the original definition in the metamaterials community, a spaser is an SPP quantum amplifier that does not necessarily generate a radiative output[1]. Here I wish to outline some distinctive characteristics of spasers and point out related phenomena that can potentially be misinterpreted as plasmonic lasing.

Just like conventional, macroscopic lasers, spasers comprise three principle components — a gain medium, a resonator and a pump. However, in contrast to other lasers, the resonator of a spaser provides feedback for SPP modes rather than optical modes. Typical resonator geometries include nanoflakes[2] and nanowires[3] on metal surfaces, metallic core–shell nanoparticles[4] and metal-coated nanoridges[5] or nanotori[6]. The characteristics of lasing are to some extent similar in spasers and conventional lasers: lasing sets in above a certain pump threshold and is evidenced by spectral narrowing and increased directionality, polarization and temporal coherence of the output. Whereas testing these criteria is straightforward for macroscopic lasers, plasmonic structures modify the spontaneous emission of the gain medium. Even without substantial stimulated emission, this can cause spectral narrowing that might be misinterpreted as lasing. If the resonator and gain medium support multiple SPP modes, distinguishing this effect from true spaser action is relatively straightforward: for spasing, the modal growth rate with pump power varies drastically between modes as a result of mode competition.

Plasmonic resonators (providing strong field confinement) have high dissipative loss and relatively low Q-factors, typically less than 100. This places great demands on the gain medium and pump. Structures offering true subdiffraction-limited confinement (that is a mode-volume $V_{mod} << (\lambda/2n)^3$, where λ and n are the wavelength and the refractive index in the medium) require gain coefficients of the order of (or even greater than, depending on the particular structure and confinement) 10^3–10^5 cm^{-1}. Simple accounting, that is, comparing the accumulated loss with the available gain, can indicate whether a spaser is in principle able to reach threshold. To substantiate claims of spasing, it is helpful to include such comparisons in reports on spasers. In this context, estimating losses through optical simulations and quantifying the available gain by amplified spontaneous emission[7] or pump–probe[8] measurements provides valuable information. Furthermore, to make most efficient use of the available energy, well-designed spaser resonators exploit the Purcell effect and direct most of the spontaneous emission into the desired laser mode[9]. Optimization of the device geometry and fabrication process provides considerable scope for further reduction of loss[3]. However, energy dissipation in the metal fundamentally limits the maximum Q-factors that can be attained.

Another frequent problem is properly defining the borders of the investigated system: subwavelength confinement of the electromagnetic field is not guaranteed by using merely nanoscopic metal structures. Instead, a certain geometry, such as a metal–insulator–metal waveguide or a metal-capped structure[10] is required and the actual mode-volume should be quantified relative to $(\lambda/2n)^3$ (where it should be noted that the medium is not always vacuum). If structures are not metal capped, strong confinement may still occur at appropriate frequencies but loss can be even more significant. Moreover, owing to the large optical gain present in structures designed for plasmonic feedback, even weak unintentional optical feedback, for example back-reflection from the sample surface, can trigger conventional, optical lasing. Confining the optical gain to the immediate surroundings of the plasmonic resonator helps to suppress this effect.

Probing individual spasers is challenging owing to their nanoscale dimension. Instead, scientists often study spaser ensembles, for example dispersions of core–shell nanoparticles, each potentially constituting a separate spaser. In these cases, spaser-to-spaser variance blurs threshold behaviour and broadens the emission spectrum, complicating the comparison with optical models. Moreover, resonance effects arising from interaction within such ensembles — such as random lasing from nanoparticles in a continuous gain medium[11] — are possibly misinterpreted as genuine spasing. Modelling the modes supported by the spaser and comparing these against experimental output spectra is an efficient plausibility check to verify the presence of SPP-based lasing. Finally, it is useful to engineer spasers such that their resonant wavelength is spectrally separated from standard laser lines, like the Nd:YAG harmonics; then bleed through from an upstream pump laser cannot be misinterpreted as a spaser signal.

In conclusion, careful modelling and experimental design are critical to substantiate spaser claims but will also guide the development of future, more efficient and ever smaller lasers.

References

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