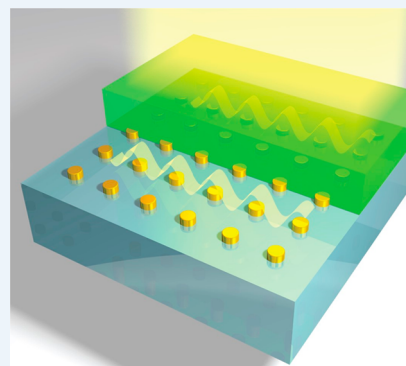


Plasmon Nanocavity Array Lasers: Cooperating over Losses and Competing for Gain

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ABSTRACT: Plasmon nanocavity array lasers leverage the combination of locally enhanced electromagnetic fields at localized particle plasmons with collective diffractive effects in periodic lattice geometries for low-threshold lasing with excellent coherence, line width, and directivity. This combination is enabled by the collective reduction of ohmic and radiative loss of plasmon antennas that hybridize to form surface lattice resonances. At the same time, candidate lasing modes compete for gain in the tight confines of the unit cell, where electromagnetic fields and population inversion are strongly structured in space, time, and polarization. This Perspective reviews the state of the art in understanding and manipulating this balance to combat losses and to optimize gain.



One of the kickstarters of the field of plasmonics was the proposal by Bergman and Stockman that surface plasmon resonances in nanoscale metal particles can be amplified by stimulated emission.¹ The original proposal for this polaritonic equivalent of a laser envisioned confinement of the electromagnetic field to nanometers in all three dimensions and spurned coupling to far-field radiation. In subsequent years, a sequence of plasmon-resonance-assisted lasers were reported that traded off confinement against thresholds and practical applicability as a bright device.² One of the foremost examples of a low-threshold practical lasing architecture that benefits from plasmons is the plasmon nanocavity array laser, first reported 7 years ago by the Odom group at Northwestern University^{3,4} and since followed up by several other groups.^{5,6} This geometry marries the benefits of distributed feedback (DFB) lasing with the near-field properties of metal nanoparticles, thereby going back to a first experiment by Stehr *et al.* on DFB lasing in metal nanoparticle gratings.⁷ In this Perspective, I describe the current status of plasmon nanocavity array lasers and the related work by Knudson *et al.* that is reported in this issue of *ACS Nano*.⁸ The Perspective highlights how the phenomenon that appears to be a plasmon-enhanced version of DFB lasing actually is critically dependent on, and controllable by, a near-field structure in the vector field of light on 20 nm length scales.

The success of the plasmon nanocavity array lasing architecture depends on the spectrally sharp, angularly selective resonances of the periodic arrays of plasmonic nanoparticles (Figure 1a,b). The nanoparticles themselves are interchangeably called “nanocavities”, or “nanoantennas”, indicating that they are at the same time resonant and strongly

scattering. As a consequence, the nanoparticles individually have quality factors of 5–30. However, “spectrally sharp” means that quality factors in lattices are in excess of $Q \sim 300$ (equivalent to a few nm line width), exceeding the resonance quality factor of the individual particles by at least a factor of 10.^{9,10} These ‘surface lattice resonances’ appear close to conditions for grazing diffraction orders. Although they are extended collective modes, they still have notable plasmonic character in terms of strong field concentration at each of the constituent plasmon nanoantennas. It is this marriage between confinement and Q that has made surface lattice resonances a staple tool in plasmonics for applications ranging from refractive index sensing, to Raman spectroscopy, to fluorescence enhancement in solid-state lighting contexts, to collective strong coupling.¹⁰ Figure 1c shows, for instance, that ensembles of fluorophores next to the particles do not emit as a simple Lambertian source but show enhanced emission at angles coincident with lattice resonances.¹⁰ In a suite of experiments that began in 2012, the Odom group demonstrated that surface lattice resonances are an excellent means to provide distributed feedback, meaning that plasmon lattices easily lase when optically pumped with (sub)-picosecond pulses of modest energy density ($< \text{mJ}/\text{cm}^2$).^{3–7,11} Evidence that lasing occurs is immediately observable from a dramatic spectral line width narrowing and a transition from Lambertian to highly directional output right at the threshold, which is apparent in input–output curves (output in just the lasing line *versus* pump power, Figure 1d). The lasing is highly directional and has subnanometer spectral

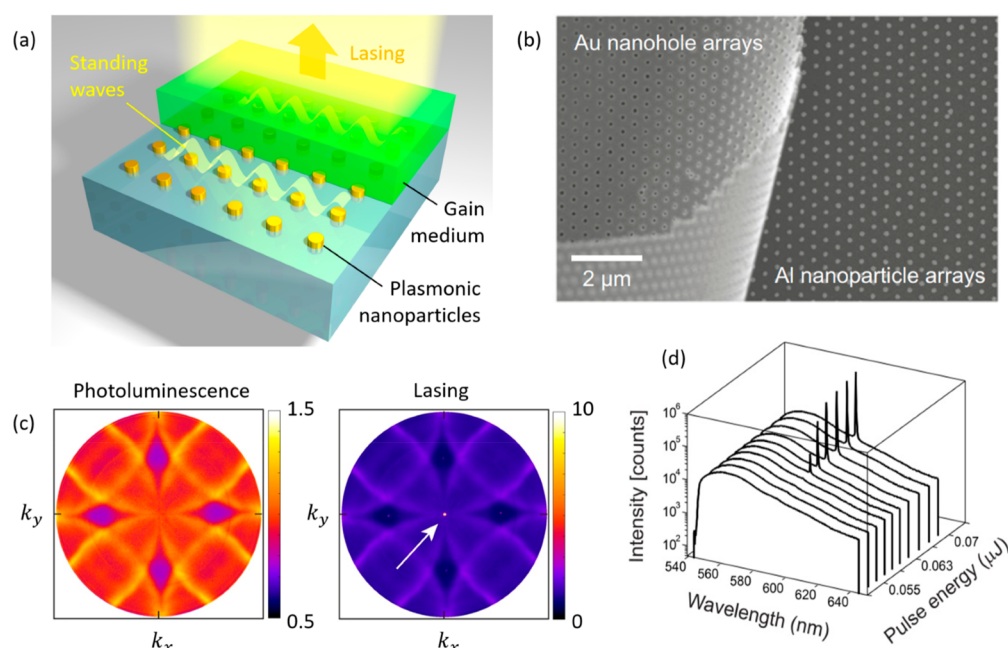


Figure 1. (a) Scheme of a typical plasmon lattice laser: a periodic metal nanoparticle array supporting high- Q surface lattice resonances coupled to a gain medium. (b) Example scanning electron microscopy image of an aluminum nanoparticle array that can be used for plasmon lasing. The array was made using a template stripping technique that is suited to covering large areas. Reprinted with permission from ref 12. Copyright 2016 National Academy of Sciences of the United States of America. (c) Example k -space photoluminescence (PL) enhancement patterns for fluorophores on plasmonic nanoparticle arrays below (left) and above (right) the lasing threshold. These are not real space images, but are recorded by Fourier imaging; they represent signal versus emission angle. Bright arcs indicate enhanced PL into the isofrequency contours of the repeated zone scheme lattice resonance dispersion. Lasing occurs at their intersection in the middle (second Bragg condition), such that the laser light is emitted in the normal direction. (d) Example emission spectra near the lasing threshold, showing the emergence of a laser peak from the broad luminescence band. These data are taken from research by the author. Reprinted and adapted with permission from¹³ Springer Nature: Springer, *Quantum Plasmonics*. Springer Series in Solid-State Sciences, vol 185, edited by S. Bozhevolnyi, L. Martin-Moreno, F. Garcia-Vidal, Chapter Plasmon Particle Array Lasers, by Y. Hadad, A. H. Schokker, F. van Riggelen, A. Alù and A. F. Koenderink 2017.

line width, thus showing the signatures of spatial and temporal coherence. Since diffraction is used to set the lasing conditions, the emission is easily tunable by periodicity and refractive index.¹¹ This functionality directly translates to tools for real-time dynamic control of plasmon lattice lasers, since they can be made on elastomeric stretchable substrates (tunable periodicity) and in liquid flow-cell geometries (tunable refractive index and gain medium).

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BASIC PRINCIPLES OF PLASMON LATTICE LASING

To appreciate the forefront of plasmon nanocavity array laser research, it is useful first to summarize the basic physical mechanisms that go into lasers built from plasmon nanoparticle arrays.¹⁰ The first step is to take a two-dimensional (2D) periodic lattice, usually taken to be square, and select a

pitch that is equal to the wavelength of the desired lasing output as measured in the medium surrounding the nanoantennas. This selection defines a band structure of grating anomalies that can be understood with the dispersion relation of the medium surrounding the nanoantennas folded back into the first Brillouin zone (Figure 2a). Hybridization of these anomalies with the polarizability of the particles forms surface lattice resonances, just slightly shifted in frequency from the grating anomaly conditions. Around high-symmetry points, anti-crossings appear, and lasing can occur at the edges of the resulting stop gaps, where standing modes arise and the bands are flat. As is standard practice for DFB lasers optimized for out-of-plane radiation, one chooses the pitch to operate at the second-order Bragg condition to ensure that the structure will have bright outcoupling at the same time as feedback.

As opposed to the usually weakly scattering dielectric corrugation in DFB lasers, the unit cell is filled with scatterers that offer a combination of plasmonic enhancement effects. First, plasmonic particles pack the largest possible optical cross section for scattering ($(3/2\pi)\lambda^2$ for dipole scatterers) in a small volume, meaning that the feedback per unit cell is strong and scattering is not perturbative. Second, plasmonic particles concentrate the optical pump field as well as the field of the lasing mode in small shells of ~ 20 nm thickness surrounding the particles. Third, the plasmonic particles at the metal interface provide substantial Purcell enhancements, promoting spontaneous emission rates and accelerating the time scales for buildup and decay of population inversion that would enter a

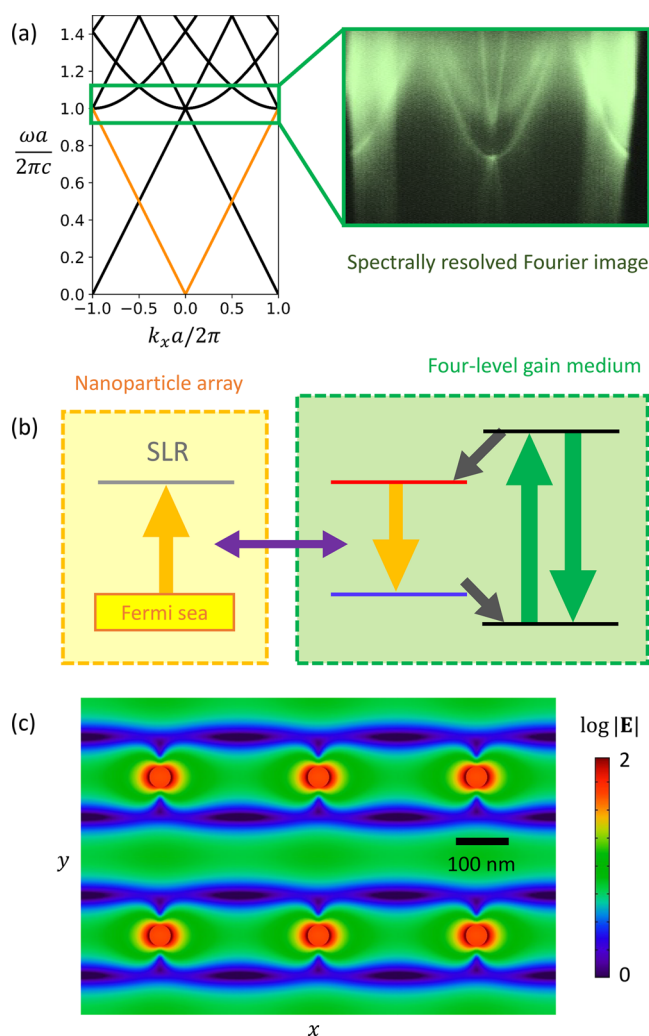


Figure 2. (a) Illustration of the expected dispersion band structure of a square lattice (left), simply repeating the dispersion of free space $k_{||} = \omega/c$ every reciprocal lattice vector. Modes that lie inside the light cone (orange) that hybridize with the plasmon particle resonance give rise to surface lattice resonance features in extinction and fluorescence (example measured band structure shown; lasing in this example occurs on the narrow, lower, band edge). (b) Energy diagram of a plasmon lattice laser proposed by Zhou *et al.*⁴ Plasmonic lattice modes (gray) can be excited at frequencies matched with the lasing transition (orange), providing an additional radiative decay channel for the inverted population (red). Copyright 2013 Springer Nature. (c) Electric field distribution associated with an optical eigenmode in an Ag particle array (COMSOL eigen mode solver, 40 nm silver disks in a dielectric) at the Γ point ($k_{||} = 0$). The main challenges include understanding and optimizing the interplay between the localized electromagnetic fields of the dipolar (this example) and higher order plasmon resonances and the spatiotemporal physics in the gain medium.

rate equation description. Thus, while the gain medium is usually applied as a homogeneous distribution of fluorophores in a solvent^{4,11} or polymer film,⁵ in fact, its participation in the lasing is spatially inhomogeneous. Therefore, the lasing is determined by physics in the gain media on the nanometer length scale and picosecond time scale.^{14,15}

OPEN QUESTIONS

One might extrapolate that plasmon nanocavity array lasers are similar to organic DFB lasers, simply with stronger feedback. Indeed, some characteristics are similar: laser wavelength directly tunes with pitch and refractive index *via* the Bragg condition, and plasmon lattice lasers have similar angular output and polarization properties. The strong feedback expresses itself as a remarkable resilience of these systems, for example, introducing disorder by randomly removing nanoparticles from the lattice, shrinking the laser to just a few unit cells across, or introducing variations in lattice geometry over a hierarchical set of length scales, such as in supercells and quasiperiodic arrays.^{6,16–19} This freedom to modify lasing geometry dramatically without immediately paying a price in lasing threshold enables researchers to program multimode lasing as well as multibeam output of phase-locked beams. These are all highly useful properties for applications as nanoscale light sources and sensors.

Yet, beyond the analogy to DFB lasers lies a vast field of open questions. Standard DFB lasers are well understood through the seminal coupled wave theory of Kogelnik and Shank, a perturbation theory for plane waves in the presence of losses and gain coupled with perturbative refractive index corrugation.²⁰ In real-space terms, such a model holds that the lasing mode simply traces out sinusoidal standing waves within the unit cell, and the only spatial structure of interest is on the length scale of many unit cells in the form of slowly varying mode envelopes that depend on the applied gain and the available scattering strength for feedback. Plasmon nanoparticle arrays, however, are not perturbative in terms of scattering strength, and the laser fields are highly structured on short length scales. These plasmon nanoparticle array characteristics lead to two interconnected puzzles.

The first puzzle relates to the long-range collective modes, that is, the nature of the band structure underlying lasing, and the long-range spatial structure of the lasing modes. Coupled wave theory only classifies the bands of weakly perturbative photonic band structures and, thus, does not apply near nanoantenna resonances, where the antenna scattering cross-section is as large as the unit cell and provides a nontrivial phase shift upon scattering. Figure 2a shows a representative measurement of bands in below-threshold fluorescence. Although the anti-crossing linear bands and parabolas are expected from folded grating anomaly cartoons, neither the magnitude of the splitting between bands, the fuzzy width of the bands, nor the contrast with which they appear in the data are easy to understand. Intuitively, one would expect the relevant parameter in this puzzle to be the particle scattering strength, as set by the complex-valued particle polarizability tensor in the vein of ref 21. However, at best, measured observables in ω, k can be reproduced in full wave simulations but cannot be cast in a simple understanding in terms of scattering strength, which also makes it difficult to estimate feedback from band structures consistently. According to Kogelnik and Shank's theory, stop gap widths and feedback strength are directly related.²⁰ In practice, measured stop bands in plasmon lattice band structures and measured shapes of slowly varying mode envelopes in finite-sized lasers are dramatically inconsistent in terms of extracting parameters for feedback strength.²² The underlying physics indicate that the feedback strength is heavily influenced by the local electromagnetic field distribution, which can have sharp gradients

over the particles (involving higher order multipoles) and gain medium.

The second puzzle essentially revolves around the imprint of the fine length scales of plasmonic field confinement on the population inversion and dynamics in the gain medium, a puzzle already recognized in the first publications in the field.⁴ A variety of effects are simultaneously at work here. Local enhancement of the pump light defines a spatially inhomogeneous pumping of the gain medium. Tight confinement of the lattice plasmon modes means that modes compete for the gain preferentially in hot spots ~ 20 nm across (Figure 2c). Finally, Purcell enhancement effects change the dynamics of spontaneous emission as another strongly spatially dependent factor entering the gain medium dynamics. In this second puzzle, the relevant parameters are decoupled from the nanoantenna scattering strength as measured by scattering cross-section, for example. Instead, the relevant quantities are the local field distributions associated with the antenna resonances at the pump and laser wavelengths and the local density of optical states in the spectral fluorescence bandwidth of the gain medium. Strictly speaking, these near-field effects are not just spatially dependent but also are *vectorial*. For instance, pump polarization will determine not only the location of hot spots where emitters are excited but also potentially which subset of emitters in the hot spots is excited through projection of the near-field pump polarization on the emitter transition dipole moments. Similarly, the various competing laser modes and the local Purcell enhancement factor can all have distinct polarizations encoded in them.

NEW PUZZLE PIECE BY KNUDSON *ET AL.*

Systematically dissecting the role of scattering strength and near-field enhancement effects in experiments is not easy. For instance, when asking how to probe which nanoantenna parameters matter, one would ideally *independently* vary scattering strength (cross section, polarizability), absorption, and near-field enhancement for pump and lasing modes. In a similar vein, to understand the imprint of near fields on the gain medium, it would be highly desirable to study a given antenna geometry while programming at will within the laser unit cell which subvolumes provide gain and which do not. If one could indeed control the gain in the unit cell spatially and vectorially, then one could understand and tailor the lasing through the controlled variation of the overlap of gain and plasmonic Bloch modes. Unfortunately, these abstract ideals are not straightforward to achieve. For instance, swapping out nanoantenna material (*e.g.*, Ag, Au, Al) modifies scattering strength, albedo, and near-field enhancement, but not independently. Similarly, programming the spatial and vectorial distribution of gain at will *within* the unit cell is difficult because the length scales are far below the diffraction limit.

In this issue of *ACS Nano*, Knudson *et al.* report an interesting experiment that cleverly exposes the possibility of programming the overlap between gain and Bloch modes.⁸ To this end, the authors study lattices of a low symmetry, essentially taking a squashed hexagonal unit cell with lattice angles up to 70° , instead of 60° . The consequence of the rhombohedral symmetry is that Bragg conditions that are degenerate for hexagonal lattices split, leading to two stop gaps that are close in wavelength. Not surprisingly, both Bragg conditions can be made to lase. In addition to turning to low lattice symmetry, the authors also changed the nanoparticle type from conventionally isotropic, cylindrical particles to

markedly elongated shapes. This change essentially ensures that within the gain bandwidth, the nanoantennas are resonant only along one axis and hardly respond for orthogonal polarization. The surprise of the work is that the authors can control *which* mode lases on the basis of the pump polarization. One would anticipate that by changing pump polarization, one could excite *spatially* different hot spots in the gain medium, thereby selecting through spatial overlap which of the two lattice modes gets sufficient gain to lase. However, simulations show that, in this work, the associated lattice modes have nearly identical real space distributions, with shared hot spots of their electromagnetic energy density essentially pinned by the short-axis resonance of each nanoantenna. Knudson *et al.* argue that the selection of which mode lases is *not* through spatial patterns in the gain, but rather through the vectorial nature of light. The competing modes may share the same hot spots spatially, but through different near-field polarization nonetheless couple to different populations of the gain medium. Streak camera imaging indicates that such a vectorially controlled preference for either of the two modes also directly imprints on the temporal dynamics that occur in the gain medium, giving an unprecedented glimpse into the polarization-resolved mode competition occurring on picosecond time scales in the plasmonic hot spots. Recently, theoretical techniques were developed that incorporate such intricate interplay between gain medium dynamics and electromagnetic fields.¹⁵ Still, theory has major challenges ahead if it is to catch up with experiment, since theory can only tackle single unit cells with periodic boundary conditions and deterministic driving to kickstart the system into amplification. These two assumptions impose the lasing mode instead of letting lasing start from noise and allowing the laser itself to select the winner.

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OUTLOOK

There are many interesting repercussions to the essential insight that one can construct intrinsically multimode nanolasers in which one can selectively elicit desired (superpositions of) modes by programming the pump field distribution in the unit cell and that one can do so without having any requirement for subdiffraction illumination techniques to structure the pump light. Indeed, the antennas themselves transduce the far-field pump light to near-field structure in the population inversion. Whereas in the present work, Knudson *et al.* used only pump polarization to control near-field polarized excitation of hot spots of gain medium, one can envision that angle of incidence, wavelength, polarization, beam orbital angular momentum, and, for pulsed pumping, temporal pulse envelope could also steer near-field distributions of pump light. This technique would provide a method for laser modulation that transcribes spatial properties of a control signal (pump) onto spectral and temporal laser characteristics. As Knudson *et al.* point out, the control parameters including the pump field

polarization also provide engineering cues to working with low-dimensional materials such as 2D semiconducting transition-metal dichalcogenides and perovskites, which have highly anisotropic linear and/or circular transition dipole moments.⁸ These ideas also have the potential to impact the emerging field of strong coupling plasmonics.²³ In this field, workers seek to hybridize excitons in dense molecular matter with plasmons, where sufficiently large vacuum Rabi splitting may lead to new collective effects such as Bose–Einstein condensation of exciton polaritons, modifications of photochemistry, and modification of transport properties of charge and excitons. Although the density of excitonic material is an order of magnitude higher in strong coupling studies than in lasers, the plasmonic part is similar. Hence, the precise understanding gleaned from building plasmon lattice lasers translates to strong coupling studies. In that community, experiments usually do not account for the huge heterogeneity of light–matter coupling strength, even if it likely has a major impact on observables.

A further impact of the work of Knudson *et al.* may be in the field of active metasurfaces. Metasurfaces are 2D nanostructured surfaces, often 2D arrangements of metallic or dielectric resonant nanoantennas, with the potential to reshape transmitted wavefronts at will. They could be seen as analog signal processors acting on 2D data sets, that is, incident beams.²⁴ Imbuing metasurfaces with gain by applying a pump but working below any lasing threshold, one could not only envision improving the efficiency of existing linear metasurface functions by having another degree of freedom to control refractive index and impedance independently, but also to create new, nonlinear function through amplification and gain saturation. Thus, spatially manipulated gain is one of several routes toward spatiotemporal control of metasurfaces.²⁵ Conceivably, the ideas of Knudson *et al.* could be generalized to programming gain in metasurfaces by programming the illuminating pump field. These notions also link to the potential of realizing metasurfaces and plasmon lattices with (broken) parity-time symmetry.²⁶

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Notes

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