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9 Plasmon particle array lasers

Y. Hadad, A. H. Schokker², F. van Riggelen², A. Alù^{1,2} and A. F. Koenderink²

¹Department of Electrical and Computer Engineering, The University of Texas at Austin,
1616 Guadalupe Street, UTA 7.215, Austin, TX 78712, USA
Tel.: +1 (512) 471.6530 / Email: alu@mail.utexas.edu

²Center for Nanophotonics, FOM Institute AMOLF, Science Park 104, 1098 XG Amsterdam,
The Netherlands
Tel: +31 754 7189 / Email: f.koenderink@amolf.nl

Abstract Diffractive arrays of strongly scattering noble metal particles coupled to a high-index slab of gain material can form the basis for plasmonic distributed feedback lasers. In this chapter, we discuss recent theoretical and experimental results describing the electromagnetic properties of these structures. Particularly, we investigate bandgap topology versus detuning between the plasmonic and Bragg resonances. We examine the complex dispersion relation, accounting for the fact that the particles are electrodynamic scatterers with radiation loss, that couple via a stratified medium system supporting guided modes. From the complex dispersion of this array we can deduce loss and outcoupling properties of the various Bloch modes, giving a handle on its lasing properties. From the experimental side, we show how to measure the dispersion relation using fluorescence microscopy, and systematically examine the array dispersion for realized plasmonic lasers as function of detuning between particle and lattice resonance. We conclude the chapter with a vision towards employing disordered, quasiperiodic and random plasmonic arrays to induce different optical responses, and experimentally demonstrate the exceptional robustness of lasing to disorder in these systems.

9.1 Introduction

Lasing takes place when gain and feedback are combined. In conventional laser systems the feedback mechanism is typically implemented in Fabry-Perot resonators with partially reflecting mirrors that enable a fraction of photons to escape the cavity after several roundtrips [1]. Alternatively, in **distributed feedback lasers**, the feedback is achieved through distributed resonances, such as **Bragg** resonances [2], or, when dealing with random systems, multiple scattering [3]. In general, similar to conventional lasers, the efficiency of the distributed feedback, together with the structure's Ohmic and radiation losses, plays an important role in deter-

mining the lasing threshold, i.e., the required gain above which the excited mode is self-sustained. Commonly, distributed feedback lasers are based on periodic dielectric gratings, and therefore they present inherently low loss, but also weak scattering cross sections, and thereby low feedback efficiency. For example, organic distributed feedback lasers have been widely studied since the mid-nineties for their ability to provide large-area lasing upon optical or electrical pumping, while being cheap to fabricate [4]. Such lasers generally consists of an organic **gain** medium that is deposited as a thin layer on a periodically corrugated dielectric surface, with a periodicity chosen such that it offers an in-plane Bragg diffraction condition within the gain window [2],[5]. A wide range of emission wavelengths can be chosen through the availability of a vast range of organic fluorophores and fluorescent polymers, while the relevant, usually weak, perturbative, corrugations can be realized, e.g., through optical lithography, or soft-imprint lithography [6],[7].

More recently a different class of lasers was proposed based on plasmonic materials. Plasmonics revolves around the fact that free electrons in metals can support collective resonances at optical frequencies [8]. This causes metal nanoparticles or nanostructured surfaces to provide highly enhanced and strongly localized electromagnetic fields upon irradiation, and to provide large spontaneous emission rate enhancements when coupled to nearby fluorescent sources[9]-[11]. This notion has triggered the development of several classes of plasmon lasers or ‘**spasers**’ [12]. Implementations include hybrid-mode nanowire lasers, in which light is tightly confined in the gap between a metal and II-VI gain medium, metal-insulator-metal lasers in III-V systems [13]-[15], and long-range surface plasmon systems coupled to organic gain media [16]. Here we focus on plasmonic distributed feedback lasers: when **plasmonic particles** are placed in two-dimensional **diffraction** periodic arrays, they not only can provide large field enhancement, but further provide control over emission directivity and brightness due to a hybridization of localized plasmonic resonances, and grating anomalies associated with the array [17]-[19]. In particular, these systems have been studied as substrates for Surface Enhanced Raman Scattering (SERS) [20], for sensing [21],[22] and for solid-state lighting [23]. Recently, several groups [24]-[26] have shown distributed feedback lasing in such plasmonic periodic systems. A main difference with conventional dielectric feedback lasers is that, while the dielectric perturbation is weak and non-resonant, for plasmonic systems the scattering strength per unit cell of the lattice can be made very strong, based on a resonance. One practical advantage is that strong scattering implies that any stop gap that arises in the band structure of the waveguide-particle system will be broader. In turn, **broader stop gaps** correspond to smaller Bragg scattering lengths [27] or, equivalently, much smaller required device sizes for lasing, and large robustness to disorder. However, this stronger coupling doesn’t come without a price: plasmonic materials are lossier compared to dielectrics. Therefore a systematic study of these new systems is required.

This chapter gives an overview of recent theoretical and experimental efforts towards better understanding this subject. We begin with an overview of the typical experimental measurements in this field that support lasing effects, leading to observations of the underlying band structures. Next we present a theoretical **dipole** model description used in order to account for near- as well as far-field interactions between the particles as mediated by the waveguide structure. As opposed to a periodically corrugated dielectric waveguide with a weak periodic grating, the "nearly-free photon" numerical approach to calculate the band structure with very narrow stop gaps [28] is not valid here, and a more adequate model is developed here. This theoretical model is followed by a comparison with an experimental study of the **band structure** underlying lasing as the plasmon resonance is tuned to the lasing condition. Moreover, we demonstrate that, as the plasmon resonance crosses through the lasing condition, the loss characteristics of bands interchanges, and as a consequence also the stop gap edge at which lasing condition occurs moves from the low to the high end of the gap. We compare these findings with the theoretical electrodynamic point dipole model. The chapter concludes with a brief discussion of very recent results regarding quasiperiodic and quasi-random plasmonic systems, showing that the strong scattering offers unique opportunities for studies in the fields of aperiodic (dis)order.

9.2 Experiments on plasmon lattice laser

9.2.2 *Samples and experimental methods*

In this section we review experimental methods to probe plasmon particle array distributed feedback lasers. All experiments to date report optically pumped, pulsed-excitation, laser action, typically measured either in a low-NA spectroscopy set up, or in a microscope. We have found it advantageous to use fluorescence microscopy techniques [25] so that in one instrument we can measure spectra, input-output curves, real space output and wave vector resolved output (Figure 1). In essence, our method revolves around an infinity-corrected inverted fluorescence microscope with a very high-NA microscope objective (NA=1.45, Nikon Plan Apo 100x). By focusing a pump laser (532 nm in our case) in the back aperture of the microscope objective, one can pump a reasonably-sized area on the sample, of typically 20-100 μ m across, the only difference with regular epi-illumination being that the excitation is pulsed (0.5 ns, μ J energy pulses in our case). Wide-field fluorescence, captured by the same objective and separated from the pump light by a dichroic beamsplitter, can subsequently be analyzed in various ways. As one typically uses low-repetition rate pulses, single-shot large area detection is called for, such as offered by current CCD cameras with effective sensitivities of about 5

photons per pixel count, at *effective* dynamic ranges of better than 2^{12} . First, by imaging fluorescence directly on a CCD camera, one can make real space images of sample output. Second, using a Bertrand, or conosopic lens, one can also perform back-aperture imaging [29][30][31]. High-NA back-aperture images quantitatively map angular-emission profiles, with resolution in the order of 0.2 degrees, over the entire angular acceptance range of the used objective (70 degrees). Owing to the “Abbe-sine” rule these ‘**Fourier**’ images are direct maps of $k_{\parallel}$, parallel-momentum-space. While on a regular 2D CCD array (Andor, Clara Si CCD) one obtains wavelength-integrated intensity versus k_x and k_y , one can also relay the Fourier image to the entrance slit of an imaging spectrograph [32] (Shamrock 303i, with Si iVAC CCD). If this is equipped with a CCD camera, one can obtain direct maps of energy-momentum space for one chosen linecut through momentum space. The most important feature that this single-shot imaging approach lacks is the ability to assess temporal photon statistics, for which one would need continuous-wave operation, and time-correlated single photon counting.

Fig.1.(a) Diagram of an inverted fluorescence-microscope adapted to study plasmon lasers. Epi-illumination is provided by a 532 nm pulsed laser (0.5 ns, μ J energy). Collected light from single pump shots is collected either on a camera or spectrometer, where a Fourier lens can switch between real-space and k-space imaging. Right: diagram explaining back-focal plane imaging and the Abbe sine condition. The objective lens is represented by its reference sphere of diameter f NA (with f the objective focal length). Physically the back focal plane is located near the baffle at the mounting thread of the objective.

9.2.2 Input-output curves, thresholds and Fourier space

In this section we discuss the typical features of the data one obtains when measuring plasmon lattice lasers, illustrated by the example of systems studied in our group that consist of particles in a waveguiding layer that supplies gain. It should be noted that several other works, notably of the group of Odom at Northwestern [24],[26],[33] have studied systems that share many of these broad features, yet also differ on important other aspects. These are the topic of section 9.5.

As a typical example, we consider systems of Ag and Au particles in periodic square grids, fabricated using electron beam lithography and lift off, and deposited on glass. These are covered by a 400 to 450 nm thick layer of a high index polymer ($n=1.6$ for SU8) doped with 0.25 wt% of the organic dye Rh6G. The thickness and index of the layer means that a single TE and a single TM waveguide mode are supported, with a mode index of around 1.54. With this particular choice, the gain window will be at around 590 nm, while the plasmon resonance for strongly scattering, i.e., large (100 nm diameter) metal disks will be further to the red. For lattice periodicities in the range of 360 to 400 nm, the kinematic 2nd order Bragg diffraction sweeps from 550 to 615 nm, through the gain window. Figure 2 presents a typical below- and above threshold spectra, as well as an input output curve collected normal to the sample, on a lattice with a pitch of 380 nm. Typical findings in this system are a clear threshold at pulse energies of order 50 nJ or 1 mJ/cm². At threshold, the spectrum under normal incidence sharpens to a peak of sub-nm width lasing peak that is to within a few percent accuracy at the kinematic 2nd order Bragg condition for the waveguide mode. This wavelength is hence tuneable by lattice pitch, and by the effective index associated to the diffractive resonance, which can be tuned by varying the waveguide or cladding index [26].

Fig. 2. (a) Emission spectra for various pump energies showing the typical emergence of a narrow laser line at the frequency of 2nd order Bragg diffraction of the gain-slab waveguide mode (pitch 380 nm). (b) Input-output curve plotting the integrated energy in just the spectral window of the laser peak versus pump power. (c) Panchromatic Fourier image just below threshold. (360 nm) pitch. (d,e) Photoluminescence enhancement for the same data, just below and just above threshold, obtained by dividing out the radial dependence for fluorescence in the same geometry without particles..

Broadly, this is the typical behavior for 2D distributed feedback lasing, also commonly observed in dielectric structures [2]. The threshold energies are comparable to those reported by Suh, Zhou and Yang [24],[26],[33], although they used a gain medium shifted to the near infrared (IR140 dye), and are quite comparable to thresholds of more optimized polymer DFB lasers [2],[4].

While the physics is similar to that of 2D dielectric **DFB lasers**, we propose that, if there are differences, they must be encoded in the underlying periodic-system band structure that depends on the strong, resonant scattering of the plasmon particles. Figure 2 shows a raw back focal plane image below threshold, as well as the corresponding photoluminescence enhancement below and above threshold. We remind the reader that **back focal plane** images form a map of parallel momentum $k_{\parallel}$, or more precisely of $k_{\parallel}/k_0 = n \sin \theta [\cos \phi, \sin \phi]$ where (θ, ϕ) are respectively the angle of emission relative to the sample normal, and the in-plane angle, and where n is substrate index. Viewed from the glass side, emission from any thin fluorescent layer at the air/glass interface will appear as a comparatively dim disk for $k_{\parallel}/k_0 < 1$ surrounded by a bright ring for larger NA. For layers that support a waveguide mode, in fact most emission is expected to occur at $|k_{\parallel}|/k_0$ equal to the waveguide mode index, which is by definition inaccessible to any objective. Due to the lattice periodicity, however, the waveguide dispersion folds back, causing the appearance of bright rings with radius of curvature 1.54, and centered at the reciprocal lattice vectors (or more precisely, at vectors $\mathbf{G}/k_0 = \lambda_0/d (m, n)$). Normalizing emission to that from an unpatterned slab, we find typical photoluminescence enhancements up to 1.5 times.

Fig. 3. (a) By relaying the Fourier image to the entrance slit of a spectrometer, one can disperse one slice of k -space over frequency. A band structure becomes apparent in the fluorescence. Panel (b) shows that for high-index dielectric particles (titania disks of 150 nm diameter), the band structure hardly shows an avoided crossing. In contrast for silver disks (110 nm diameter) a distinct band anticrossing appears near the 2nd order Bragg condition (at 590 nm, set by the 380 nm pitch).

These enhancements are identical in mechanism to, but in magnitude far smaller than those, obtained by Lozano et al. [23], and Rodriguez et al [34] in the framework of plasmon-lattice enhanced remote phosphors with optimized alignment of

LSPR and diffractive waveguide coupling. As we cross the threshold, a bright and narrow beam emerges that has a donut profile. Polarization analysis evidences this lasing beam to be TE, i.e., s-polarized. From this we conclude that laser light is mainly outcoupled through the TE mode, which has a strong spatial overlap with the gain and with the particles. While the picture of band folding of the free photon / waveguide dispersion is generic to any periodic system, the distinct physics of the system is contained in fine features, such as the avoided crossings that are expected at Bragg conditions, i.e., precisely at the intersections of the free photon circles. Resolving such features requires spectral resolution at the same time as wave vector resolution. While results in Figure 2 on a 2D CDD chip report the sum over all emission wavelengths, wavelength information can be obtained by dispersing one slice of k-space on a grating. Figure 3 show typical below-threshold spectrally resolved Fourier image. The folded waveguide dispersion appears as narrow bright features organized in four bands. Indeed, at second order Bragg diffraction in a square lattice four diffraction orders couple, namely diffraction along $2\pi/d(\pm 1, 0)$ and $2\pi/d(0, \pm 1)$ giving respectively straight lines crossing at the Γ -point, and two parabola. Due to strong scattering at the plasmon particles, these four bands display a splitting that strongly depends on the plasmonic nature of the particle. In particular we have found that even for very high-index dielectric particles (TiO₂, disks 150 nm in diameter were required [25] to obtain lasing) no observable stop gap is found. In contrast, for silver particles, stop gap widths up to 20 nm, or $\Delta\omega/\omega=3\%$ occur. For Au particles, we have found only narrow stop gaps and high lasing thresholds, showing that more absorptive metals give weak scattering yet high loss

9.3 Theory of plasmon lattices coupled to stratified media

Now that the reader is familiarized with the general characteristics of 2D plasmon lattice lasers we turn to a theoretical model able to capture the main phenomena in an analytical framework. Starting from the **nearly free photon approximation** as intuition, we develop a point dipole model for the **complex band structure**.

9.31 Two-dimensional periodic arrays, folded dispersion, and the “nearly free-photon” approximation

Plane waves are solutions of the wave-equation in infinite homogeneous media, governed by a conical dispersion $\omega = kc$, where $k = (k_x^2 + k_y^2)^{1/2}$ and c is the speed of light. If in the medium we introduce a periodic 2D array of identical particles, then coupling between the plane waves due to the particles will lead to new

forms of solutions. These are termed Bloch or Floquet harmonics. The dispersion of these solutions is periodic in the (k_x, k_y) plane, the so-called reciprocal or momentum plane that is directly imaged by Fourier microscopy. For example, if the lattice in the physical domain is square with lattice constant d then in the k -space, the reciprocal lattice will also be square and with reciprocal lattice constant $2\pi/d$, as shown in Fig. 4(a-b). The area defined by, $(k_x, k_y) \in [-\pi/d, \pi/d] \times [-\pi/d, \pi/d]$ in the (k_x, k_y) plane is termed the first Brillouin zone (BZ). In the limit of negligibly scattering particles the dispersion of the waves supported in the periodic array will consist of infinite replicas of the fundamental cone, as shown in Fig 4 a. Also highlighted in Figure 4 are two common cuts through this diagram. On one hand, Fig. 4b shows an equifrequency slice, similar to taking a constant energy (Fermi) surface in k -space for electrons. Figure 4c, instead, shows a cut through the dispersion relation at fixed $k_x = 0$, thus representing an ω vs k_y bandstructure.

Fig. 4 Sketch of the nearly free photon dispersion in a 2D system (a) the conical dispersion relation $\omega=c|k|$ is repeated every reciprocal lattice vector (black dots). The cone centered at $|k|=0$ is plotted in orange. (b) Constant frequency slice, showing the circular dispersion relation repeated every reciprocal lattice vector. The cut is taken at a frequency just below 2nd order Bragg diffraction – with increasing frequency the circles grow, and intersect at the origin of k -space. (c) ω - k_y cut for $k_x = 0$, showing the well known band diagram in the first Brillouin zone (blue box in (a,b)).

If the interaction between the particles and the plane waves is weak but not negligible, then the new solutions can be described as a combination of just a few plane waves. In this case the new dispersion will be very close to the dispersion in case of negligibly scattering particles. Exceptions are frequency points where phase-matching between counter propagating plane waves takes place and thereby relatively efficient coupling between the plane waves occurs, namely, right at the intersection points of different dispersion branches. In Fig. 4c this concerns the crossing of four cones at the origin in k -space, visible in the ω vs k_y slice as two intersecting lines, and a (degenerate) parabola. This point is the second order

Bragg condition (the first being at the edge of the Brillouin zone). This description in terms of a folding of a dispersion relation is termed the “**nearly free photon**” approximation, and it is used to approximately describe the wave solutions in weak perturbation scenarios. This is similar also to the case of conventional dielectric-grating distributed feedback lasers. In this chapter, however, we study a rather different problem, namely that of a periodic lattice of plasmonic particles embedded in a stratified dielectric system that supports waveguide modes and provides gain. Plasmonic particles are strong scatterers, and therefore, although it may give the basic physics, the simplified, “nearly free photon” picture does not convey the full bandgap physics of such a system. In particular, distinct stop gaps are expected to open up. Indeed, if we compare the conceptual Fig. 4 with the measurements in Fig. 2,3 it is directly clear that there is a close correspondence, yet at the same time that theory must account for stop gaps, and the width of the bands.

9.3.2 Surface lattice resonances

Now we turn into a more detailed description of our system of interest, namely a particle lattice inside a 2D dielectric waveguide system. Consider a square lattice of plasmonic cylinders with diameter D and height H with lattice constant d positioned within a dielectric slab that supports a guided mode as illustrated in Fig. 5(a) (top and bottom). The guided slab mode wavenumbers k_m are related to the free-space wavelength λ_0 via the dispersion relation $k_m(\omega = 2\pi c / \lambda_0)$. A typical dispersion of the fundamental TE and TM modes supported by a slab with thickness of $h = 450$ nm that is made of relative dielectric constant $\epsilon_2 = 2.79$, and sandwiched between two half-spaces with relative dielectric constants $\epsilon_1 = 1$ and $\epsilon_3 = 2.25$ is shown in Fig. 5(b). The periodic lattice exhibits Bragg resonances whenever the wavelength of the waveguide mode $\lambda_m = 2\pi/k_m$, rather than the vacuum wavelength, meets the Bragg condition

$$d = m \frac{\lambda_m}{2}, \quad m = 1, 2, \dots \quad (1)$$

Eq. (1) describes the case of Bragg diffraction by rows of particles parallel to a Cartesian axis, for waves with wave vector perpendicular to it. By way of example, with $d = 400$ nm, a second-order ($m = 2$) Bragg resonance will take place at $\lambda_{m=2}^{TE} \approx 640$ nm for the TE mode with mode index around 1.55. A similar TM resonance will take place at a slightly shorter free-space wavelength, owing to the somewhat different mode index. Note that additional Bragg resonances will take place for diffraction at differently oriented particle rows (i.e., other Miller indices

l, m). As these fall outside the gain window of most reported laser studies, we ignore these.

Fig. 5. (a)Top: Illustration of the slab and the plasmonic array. Bottom: Single plasmonic cylinder. (b) Typical dispersion of fundamental TE and TM modes in relatively thin slab.

9.3.3 Semi-analytical approach: polarizability and lattice sums

Ultimately, the lasing threshold for the Bloch modes that arise due to periodicity will be determined by the balance between the provided gain and the intrinsic loss. The net gain depends on the gain coefficient and the spatial overlap of the mode with the gain medium. The intrinsic propagation loss of the Bloch modes is due to Ohmic damping and radiative outcoupling. Therefore, dominant lasing will take place at or near frequencies where the lowest loss Bloch modes for feedback arise. In other words, to understand lasing we need to determine the dispersion relation of the plasmon-waveguide system, and seek the frequency regions in which the imaginary part of the complex dispersion (in the absence of gain) is minimal. Hence, in the following we calculate the passive array dispersion. We employ a dipolar model for the scatterers, taking into account Ohmic loss, as well as the coupling between the dipolar excitations in the array and the far field radiation. Thereby, we account for the fact that lasing will take place near the low loss points in the complex dispersion, while at the same time requiring that experimentally observable lasing needs outcoupling to radiation, and hence not a *fully* dark mode. Due to the strong scattering of the plasmonic particles, coupled mode theory and the “nearly free photon” assumption that are commonly used in the analysis of conventional dielectric distributed feedback lasers will lead to incomplete results for plasmonic arrays. Therefore we use the **discrete dipole model** that accounts for near as well as far field interactions between scatterers that are modelled as polarizable points that can carry a large dipole moment [35]. This method can give relatively accurate predictions particularly in the vicinity of the plasmonic particle resonance frequency. In this frequency range the single particle

scattering behavior can be accurately modeled as dipolar, so that essential features are correctly captured.

Polarizability model

The dipolar dynamics of each particle is encapsulated in its polarizability $\alpha(\omega)$. For a strongly and resonantly scattering particle in free space, such as in the case of a metal sphere described by a Drude model for its dielectric constant [36], the **quasistatic polarizability** is given by

$$\alpha_{\text{static}}(\omega) = \frac{V\omega_0^2}{\omega^2 - \omega_0^2 - i\omega\gamma} \quad (2)$$

(in CGS units, with ω angular frequency, ω_0 the particle resonance, γ an ohmic damping rate, and V an (effective) particle volume) To turn this static polarizability into that of a physical scatterer one must include **radiation damping** [35]

$$\frac{1}{\alpha} = \frac{1}{\alpha_{\text{static}}} - i\frac{2}{3}k^3 \quad (3)$$

with $k = n\omega/c$. As we are considering particles located inside an inhomogeneous medium, this “radiation correction” must be adapted, to account for the fact that both radiative damping and the resonance frequency are normalized. Moreover, although the particles we used in the experiments discussed in the following sections are electrically small they are not much smaller than the wavelength, thereby causing additional red-shifting in their resonance frequency due to phase retardation effects. While strictly speaking, the radiation damping term in Eq. (3) can be *ab initio* corrected using the Green’s function of the stratified system [37], here we adapt a more practical approach by fitting ω_0 and V in Eq. (2) and Eq. (3), to agree with full wave simulations of the scattering cross section shown in Fig. 6a below. Following this approximate approach, for metal disks of diameter $2r$ the fitted resonance frequency is given by $\omega_0 = 2\pi c / \lambda_{\text{LSPR}}$ with $\lambda_{\text{LSPR}} = 334 \times 10^{-9} + 3.6 \times 2r[m]$ and the damping rate by $\gamma \approx 0.05\omega_0$, valid for particle diameters in the range 40 to 110 nm.

Lattice sum: Bloch wave assumption and dipole polarization

The structure is shift invariant in both x and y , and to derive a dispersion relation for the particle-waveguide system, we can hence assume that the dipolar excitation on the lattice has a Bloch form with dipolar moment

$$\mathbf{p}_{mn} = \mathbf{p}_{00} e^{id(nk_x + mk_y)} \quad (4)$$

where m, n denote the index real-space particle sites and (k_x, k_y) is the wavevector of the excited collective plasmonic mode. In this work we focus on polarization of $\mathbf{p}_{mn}$ only in the layer plane, containing components only in the x and y directions. This follows from the fact that the reported experiments work with flat silver disks, which are essentially only polarizable in the plane.. Furthermore, second order Bragg resonance that corresponds to the square lattice pitch d has to take place in two orthogonal directions, parallel to the lattice primitive vectors. Generally, the response of any particle in the lattice will be given by its polarizability and the field that it experiences due to incident field, plus the field of all the particles in the lattice. If we denote with $G(\omega, \mathbf{r}', \mathbf{r})$ the electric field **dyadic Green function** (meaning $G(\omega, \mathbf{r}', \mathbf{r}) \cdot \mathbf{p}$ specifies the field at $\mathbf{r}'$ due to a dipole moment $\mathbf{p}$ located at $\mathbf{r}$), the lattice response is

$$\mathbf{p}_{00} = \alpha \left[\mathbf{E}_{in} + \sum_{m,n \neq 0} G(\omega, \mathbf{r}_{00}, \mathbf{r}_{mn}) \mathbf{p}_{00} e^{id(mk_x + nk_y)} \right],$$

or in other words [35]

$$\mathbf{p}_{00} = \left[\alpha^{-1} - \sum_{m,n \neq 0} G(\omega, \mathbf{r}_{00}, \mathbf{r}_{mn}) e^{id(mk_x + nk_y)} \right]^{-1} \mathbf{E}_{in}.$$

In this expression, the summation of the single-dipole Green function over the real space lattice is known as **lattice sum**, and it contains all the physics of dipolar near- and far field coupling, to all multiple scattering orders. We note that if one looks for a dispersion relation, one considers the structure in absence of any driving, setting $\mathbf{E}_{in} = 0$ and looking for poles in the prefactor, which plays the role of a polarizability renormalized by interactions in the lattice. In our work we are interested in lasing near 2nd order Bragg diffraction only. In that case, since the x and y direction are equivalent, we can focus on the $k_x = 0$ slice of wave vector space, and can set the dipole polarization to $\hat{x}$. In this case, the modal matrix problem reduces to the simplified scalar equation

$$\Delta(\omega, k_x, k_y) = \alpha(\omega)^{-1} - \sum' G_{xx}(\omega, \mathbf{r}_{00}, \mathbf{r}_{mn}) e^{id(mk_x + nk_y)}. \quad (5)$$

In Eq. (5) the symbol *primed-sum* $\sum'$ denotes summation over all indices except for $(m, n) = (0, 0)$, and G_{xx} is the xx component of the electric Green's function tensor.

Fig. 6. (a) Extinction cross section of silver disks of 30 nm high and diameters 60, 90 and 110 nm, in SU8 ($n=1.65$) on glass ($n=1.5$) calculated using FDTD, assuming normal incidence from the glass side, and using tabulated optical data for silver [38] (b,c) Illustration of the lattice sum G_{xx} (summation in Eq. 5) for free space [35]. The real and imaginary part both show strong resonances exactly at the folded free dispersion, which gives rise to surface lattice resonances in the lattice polarizability.

Figure 6b shows G_{xx} for the case of a lattice in free space. Clearly, the lattice sum is strongly structured, with diffractive resonances occur right at the nearly free photon dispersion relation. Indeed, these are the surface lattice resonances on which lasing was reported by Zhou et al. [24], who operated at exact index matching between substrate, superstrate and gain slab. We also refer to Chapter 7 in this book for an overview of results one obtains in extinction and emission on basis of the same formalism.

Approximation of the Green's function in layered media

Taking the full spectral content of the Green's function into account for the three-layer system of a substrate, a gain medium, and a superstrate, in the infinite summation in Eq. (5) is numerically challenging, and not essential. In order to simplify the analysis, we take the following physical considerations into account that are valid for systems in which the gain originates from a high index slab, as in our work [25]:

- Particle-particle interaction will mainly arise through the waveguide modes
- As the TE and TM mode indices are very close in the organic gain systems typically studied [24]-[26],[33],[39] significant TE-TM coupling is expected.

Due to these physical reasons we may replace the full Green's function G_{xx} by its modal part, G_{xx}^m including both TE and TM mode contributions, i.e. $G_{xx}^m = G_{xx}^{TE} + G_{xx}^{TM}$, where the TE and TM contributions are separately given by

$$G_{xx}^{TE} = 2A_{TE} \left[H_0^{(1)}(k_{TE}\rho) + \frac{\partial_{x'}^2 H_0^{(1)}(k_{TE}\rho)}{(k_{TE})^2} \right] \quad (6)$$

$$G_{xx}^{TM} = -2A_{TM} \left[\frac{\partial_{x'}^2 H_0^{(1)}(k_{TM}\rho)}{k_{TM}^2} \right] \quad (7)$$

where $\rho = \sqrt{(x-x')^2 + (y-y')^2}$, and k_{TE}, k_{TM} are the wavenumbers in the transverse direction of the guided slab mode in the absence of the array, and are given by a solution of the corresponding mode transcendental equation. The amplitudes A_{TE}, A_{TM} are given by

$$A_X = \frac{k_0^3}{4\pi\epsilon_0} \frac{i}{2\eta_0} 2\pi\xi_X g(z, z, \xi_X), \quad X = TE, TM \quad (8)$$

where $\xi_X = k_X / k_0$, and g is the 1D Green's function given in appendix A. The infinite summation in Eq. (5) is slowly converging due to the inverse square root dependence of the Hankel function with respect to its argument. However, its convergence can be significantly accelerated by using the so called Ewald summation technique, adapted to the problem of interest. More details can be found in Appendix B. Note that taking nearest neighbor interactions only would be very inaccurate, since the propagators that couple the particles are dominated by slowly decaying slab modes that decay as $1/\sqrt{\rho}$. While far-field interactions are critical, since the particles are separated by roughly one guided wavelength from each other, the singular $1/r^3$, $1/r^2$ terms of the Green's function are practically irrelevant.

9.3.3 Theoretical model – results

Coupling of collective plasmon resonances to far-field radiation

The solution of Eq. (5) provides the complex dispersion of the collective plasmonic excitation of the array. In other words, at each frequency-wavenumber combination where Eq (6) approaches zero, the lattice shows a strong response at weak, or zero, excitation. We therefore evaluate the complex dispersion relation by solving Eq. (6) at real frequencies, yet complex wave vector. The lowest loss mode correspond to zeros with the smallest imaginary part of the complex dispersion. In addition to evaluating the **complex dispersion** of Bloch modes, we also evaluate the outcoupling of Bloch modes. In analogy with conventional laser systems, a resonator based on perfect, rather than partially reflecting, mirrors will yield field amplification but no outcoupled laser light. In order to obtain the out-

coupling efficiency for a given combination of real ω and k_y , we invoke reciprocity. The excited dipolar moment p_{00} due to an impinging x-polarized plane wave with amplitude E_0 at ω and with $(0, k_y)$ is given by $p_{00} = E_0 / \Delta(\omega, 0, k_y)$. By reciprocity, also the radiated field at given dipole moment p_{00} at ω and with $(0, k_y)$ will be proportional to $1 / \Delta(\omega, 0, k_y)$. Fig. 7 reports this quantity, i.e., the outcoupling at fixed dipole strength, as a function of real frequency ω and parallel momentum, as greyscale images. Here, white (black) represents efficient (poor) coupling. Panels (a)-(c) correspond to three frequency detuning cases $\lambda_B^{TE} < \lambda_{LSPR}$, $\lambda_B^{TE} \approx \lambda_{LSPR}$, and $\lambda_B^{TE} > \lambda_{LSPR}$, respectively, where λ_{LSPR} is the wavelength corresponds to the plasmonic particle resonance frequency, and λ_B^{TE} is the free space wavelength at which the 2nd order TE mode Bragg resonance takes place according to Eq. (1). The most notable features in Figure 7 are that the repeated zone scheme dispersion becomes apparent as sharp features. However, clear stop gaps open up between bands. Moreover, it should be noted that some bands appear dark exactly at or near $k_y=0$, indicating that outcoupling is forbidden, for instance by symmetry. Finally, the zero-detuning case shows a markedly distinct behavior.

Fig. 7. Coupling between x-polarized dipolar excitation and x-polarized far-field. (a) $\lambda_B^{TE} < \lambda_{LSPR}$, (b) $\lambda_B^{TE} \approx \lambda_{LSPR}$, (c) $\lambda_B^{TE} > \lambda_{LSPR}$.

Mixed TE-TM and the effect of resonance detuning

In Fig. 8 we zoom-in on the frequency-wavenumber range where the TE-Bragg condition is satisfied and plot the outcoupling efficiency of Fig. 7 as grayscale alongside the complex dispersion of the collective plasmonic excitation, obtained as a solution of Eq. (5). The three panels, a b and c, of the figure correspond to the three detuning cases $\lambda_B^{TE} < \lambda_{LSPR}$, $\lambda_B^{TE} = \lambda_{LSPR}$, and $\lambda_B^{TE} > \lambda_{LSPR}$, respectively. The curves (blue only or blue and brown) in each figure represent the complex disper-

sion. Shown in the left (right) side of each of the panels is the dispersion of the imaginary (real) part of $k_{\parallel} = k_y$ (recall $k_x = 0$). There are additional dispersion branches with much higher imaginary part that are not shown, as only the lowest-loss branches are important for lasing. In the left panels the frequencies that correspond to the TE and TM Bragg resonances are also marked with dashed lines. Distributed resonance and signal amplification in the presence of gain will take place at the points of minimum imaginary part. In all three detuning cases considered in Fig. 8 there are two frequencies at which the imaginary part of $k_{\parallel}$ has a minimum. Having two, and not one, condition with minimal loss is a direct consequence of TE-TM coupling. If the two polarization families were completely decoupled, each would separately give rise to a stop band, where one of the two stop band edge would correspond to minimal loss (mode with nodes at the scatterers), and one would correspond to large loss (mode with antinodes at the scatterers). In the lasers studied in experiments, however, the TE and TM modes are very close (waveguide systems) or even identical (surface lattice resonance of Zhou [24]) in dispersion. Scattering by the particles can couple TE and TM modes with in-plane momentum at right angles to each other.

Fig. 8. Lasing vs detuning. (a) $\lambda_B^{TE} < \lambda_{LSPR}$. Left: dispersion of imaginary part of k_y , distributed resonances takes place at the minimum points. Right: dispersion of the real part of k_y . Greyscale image: coupling between the excited mode to radiation. (b) as (a) but with $\lambda_B^{TE} = \lambda_{LSPR}$. (c) as (a) but with $\lambda_B^{TE} > \lambda_{LSPR}$.

The minimal loss points occur at or near $k_{\parallel} = 0$, corresponding to 'distributed' standing-wave resonances, and thereby will be amplified if sufficient gain is supplied. To observe lasing, at the same time outcoupling is required. For that, the real $k_{\parallel}$ that corresponds to the point of minimal $\text{Im}\{k_{\parallel}\}$ should co-locate with strong outcoupling, i.e., a whiter region in the underlying grayscale map. Following this idea, in Fig. (4a), $\lambda_B^{TE} < \lambda_{LSPR}$, the only point where we have simultaneously a minimum imaginary part of $k_{\parallel}$ and a corresponding real part of $k_{\parallel}$ in a high coupling region in the grayscale map, is right above the TE Bragg condition.

In the second case (4b), $\lambda_B^{TE} = \lambda_{LSPR}$, we find two points, one above and one below the TE Bragg condition, where we have low imaginary part of $k_{\parallel}$ and simultaneously the real part of $k_{\parallel}$ in a regime of good outcoupling. Finally, in the last case (8c), $\lambda_B^{TE} > \lambda_{LSPR}$, we see that laser output is expected only just below the TE Bragg condition. These observations implies that for different detuning we would expect to see lasing from different frequency regions, either only below, both above and below, or only above the TE Bragg resonance frequency.

Fig. 9. Measured band diagrams for three distinct cases, namely when the plasmon resonance is red detuned from the lasing wavelength as given by the Bragg condition, when the two are on resonance, and when the localized plasmon resonance is blue-detuned from the lasing condition. The first case is achieved using Rh6G as gain medium, taking particles of 110 nm diameter at 380 nm pitch (590 nm lasing wavelength). The other two use a Rh6G-Rh700 dye mixture as gain medium and a pitch of 460 nm (lasing at 710 nm). The plasmon is tuned by working with large (117 nm diameter) resp. small (80 nm diameter) particles. These diagrams are taken just at (left) or below the lasing threshold. Lasing occurs on the lower band edge in the left diagram ($\omega = 3.19 \cdot 10^{15}$ rad/s, note CCD over-exposure artefact), and on the upper band edge ($2.67 \cdot 10^{15}$ rad/s) in the right diagram. Colorscale is linear in intensity.

9.3.4 Stop gap and band crossing

With the theory in hand, we return to the magnitude and the topology of the band structure and lasing observable in recent experiments [24]-[26]. As the bandwidth of plasmon resonances is quite wide relative to the gain bandwidth even of organic dyes, it is not easy to measure band structures for all different detuning cases $\lambda_B^{TE} > \lambda_{LSPR}$ and $\lambda_B^{TE} < \lambda_{LSPR}$ without also changing the gain medium. To this end, we combine results from two sets of samples, one using Rh6G and a pitch of 380 nm to obtain lasing $\lambda_B^{TE} = 590$ nm, well to the red of the plasmon resonance (near 700 nm for 110 nm diameter disks), and one using a different dye (Rh700) and pitch (460 nm) to obtain lasing at 710 nm, well to the red of the plasmon resonance (occurring at 650 nm, for smaller, 80 nm diameter disks). Figure 9 shows representative measured band structures. Commensurate with the theory predic-

tions, for these large detunings the band structure can be clearly interpreted as a perturbed free photon dispersion, with noticeable stop gaps. For the on-resonance case (replacing the 85 nm by 120 nm disks), the dispersion is markedly different. Importantly, we find that lasing occurs on the red edge of the measured stop gap in the case $\lambda_B^{TE} < \lambda_{LSPR}$, while it occurs on the blue edge in the case $\lambda_B^{TE} > \lambda_{LSPR}$. This stands in good agreement with the predictions of the dipole model.

9. 5 Open questions for periodic plasmon lasers

In this section we provide a discussion of open questions, and crucial differences between the various experimental reports of plasmon array lasers[24],[25],[33]. The generic conclusion common to all works in this field is that plasmon particles allow distributed feedback lasing at thresholds of about 10 mJ/cm², quite comparable to organic DFB lasers. Two limiting factors for this threshold are: first, only about 10% of pump light is absorbed and, second, that feedback via multiple scattering suffers from absorption loss. Indeed, for Au particles, thresholds are about 20 fold-higher. As regards which plasmonic properties make these DFB lasers from their dielectric counterparts, a few distinct physical mechanisms have been proposed. First, the effect of stronger scattering per particle than in the case of dielectric systems is responsible for the large stop gap, which indicates that only few lattice spacings (in the order of 30) are needed for strong feedback [25]. Second, strong local field enhancement and **Purcell factors** have been proposed as facilitating lasing action [24],[26],[33]. Here we note that so far studies have been performed in two very distinct limits. On one hand, Schokker et al. [25] have studied lasing action in gain media with intrinsically high **quantum efficiency** (>90%). For these cases it is expected that local field enhancement and Purcell factors have no strong role. Indeed, Purcell factors near plasmon particles are typically enhanced only in very small volumes, estimated to encompass no more than a few percent of the unit cell. A very recent stochastic **superresolution-imaging** map of LDOS in periodic plasmon arrays confirms this estimate [40]. For media with high internal quantum efficiency, this means that > 95% of fluorophores participate in lasing without actually enjoying particularly strong local field enhancements, yet benefitting from feedback by strong scattering. On the other hand, in the work of Suh and Zhou,[24][33] very low internal quantum efficiency fluorophores were used (<10%). Since Purcell factors can increase the internal efficiency of low efficiency fluorophores, lasing action is expected to occur preferentially on basis of gain medium close to the metal. Indeed, Suh et al and Zhou et al. [24][33] have performed ultrafast dynamics measurements to evidence that large Purcell factors of order 300 play an important role in bow-tie lattice lasers. To systematically elucidate the relative importance of scattering, plasmonic absorption, and Purcell enhancement, it would be very useful to perform measurements with systems in which the internal quantum efficiency of

the gain medium could be tuned at will, or in which the spatial distribution of the pump beam *within* the unit cell can be controlled so as to preferentially excite emitters close to, or far from, the metal [41]. Moreover, from the photonic side, it would be ideal to perform measurements on a series of samples in which physical particle size is kept constant, yet **polarizability** and **albedo** are varied. Unfortunately, this is difficult to realize in practice. Finally, we note that in these systems the gain medium and laser output will show rich spatiotemporal dynamics. Dynamic modelling and measurements are hence required.

Fig. 10. (a) 2D plasmon arrays (pitch 380 nm, Rh6G gain medium) with particles randomly removed still show distinct lasing at the 2nd order Bragg condition (590 nm), even for extremely large dilution of the lattice. When just 1% of the particles is left, lasing persists. In this regime also random-lasing type emission spikes appear (b) Input-output curves.

9.6 Scattering, aperiodic and finite lasers

The fact that plasmon lattice lasers have comparatively wide stop gaps, means that the feedback needs only a few lattice constants (of order $(\Delta\omega/\omega)^{-1}$) as compared to their dielectric counterparts. On this basis, one would expect plasmon lattice lasers to be very robust against finite-size effects, random removal of particles, or shuffling of particles. Indeed, in our group we have found that oligomers of below 10 by 10 lattice constants already provide lasing output at the 2nd order Bragg diffraction condition. A particularly interesting finding [39] is that the lasing persists for extreme perturbations of the lattice in terms of random particle removal. We have studied structures for which as many as 95% of particles were removed from the lattice, while the remaining 5% were left in place. Remarkably, for this type of **randomized plasmonic structure** the threshold remains essentially unaffected when up to 80% of particles are removed, while the slope efficiency is about twice better for lattices with disorder, than for lattices without disorder (Fig. 10). The lasing wavelength remains pinned to the 2nd order Bragg diffraction wavelength of the

system. When even more particles are removed, the threshold does increase strongly, and laser output becomes weak.

One of the main reasons we identify for this robustness is that the planar wave-guiding geometry is itself very amenable to amplification - indeed slab waveguides have been widely studied for **Amplified Spontaneous Emission** (ASE) sources [1]. Since scattering per particle is very strong compared to the scattering per unit cell in dielectric DFB lasers (taking the ratio in stop gap widths as measure, suggests a factor 50), it stands to reason that sufficient feedback remains when many particles can be removed. A requirement is that one keeps the remaining particles on the lattice sites, so that the Fourier transform of the lattice remains strongly peaked. An exciting idea for future experiments is to explore **random lasing** and “designer-disorder” lasing [3][42][43][44][45]. Compared to other 2D systems, this lasing sample system is very easy to build and operate.

Fig. 11. (a) Fourier space output of a 2D Fibonacci plasmon array ($d=380$ nm pitch, lasing at 590 nm, as does the underlying square lattice). The left part of this panel shows the structure factor (absolute value of the structures Fourier transform) of the array. The color scale is oversaturated. (b) Real space image of the sample plane above threshold for a Fibonacci lattice, and a randomized laser (50% of particles randomly removed) showing speckle (20 μm field of view). Speckle is a direct evidence of spatial coherence (d,e) Autocorrelation of the speckle pattern (3 μm scale bar).

As an example highlighting this research direction, we have also studied lasing in a variety of **quasiperiodic** and **aperiodic** structures. Figure 11 shows lasing output in Fourier space for one such structure, a 2D **Fibonacci** lattice. This lattice is created from a square lattice (again, pitch 380 nm), by removing particle according to a deterministic, Fibonacci-sequence based scheme. The lasing wavelength remains as set by the 2nd order Bragg diffraction condition of the underlying lattice. The lasing output, however, now quantitatively matches the Fourier transform of the Fibonacci lattice. The interpretation is that this is exactly the pattern expected when considers diffraction of the $k_{\parallel}=0$ lasing beam by the quasiperiodic structure upon outcoupling. The practical importance of this example is that start-

ing from the periodic lattice, one can impose amplitude masks (in this case the binary choice of keeping or removing particles) or possibly also phase masks (varying particle resonance by size) directly in the plasmon structure that directly function as **beam shaper**. Further observations include the occurrence of many new lasing conditions at strong quasi-Bragg conditions, i.e., peaks in the structure factor.

Beyond the k-space output, in the field of random lasers and **correlated disorder** also real-space characteristics are important, or more concretely the spatial distribution and correlations in **speckle**. In our experiments, real space images of the laser structures invariably show a sharp transition from featureless, uncorrelated, Poisson noise for the incoherent fluorescence measured when pumped below threshold, to a distinct speckle pattern as threshold is crossed (Figure 11b). For lattices disordered by random particle removal, this speckle pattern simply follows the laws established for uncorrelated scattering. These state that intensities follow **Rayleigh statistics**, and the spatial **speckle autocorrelation** has a sinc-squared shape of diffraction limited width [46]. Once the arrangement is correlated, however, as in the case of the Fibonacci structure, the autocorrelation shows a plethora of additional features. Aside from providing an additional handle on unraveling the optics of deterministic aperiodic structures, we note that this observation could also have a use. Indeed, several forms of ‘speckle’-based microscopy are now developed [47],[48] where images are retrieved after illumination with speckle patterns, where the sharp autocorrelation of speckle essentially plays the role of point-spread function. Similar to traditional point-spread function engineering (for instance, by super-oscillatory lenses), deterministic aperiodic lattices provide control over the speckle autocorrelation.

9.7 Conclusions

This chapter has discussed experimental as well as theoretical aspects of plasmonic distributed feedback lasers. The discussion has been done to draw a direct comparison with dielectric gratings DFL, as well as between different approaches of plasmonic DFL. Plasmonic particles, particularly at their resonance are highly scattering elements thereby reducing significantly the finite array size required to establish significant 2nd order Bragg resonance. But this doesn’t come without a price to pay in the form of larger losses compared with dielectric gratings structures. Therefore, the lasing threshold obtained with Ag particles (that are relatively low loss) is only similar to that achievable with organic lasers and not much weaker as one may expect just in light of their high scattering.

The interplay between the Bragg resonance frequency and the plasmonic resonance is a unique character of plasmonic DFL, that modifies the region in the band diagram where the lasing takes place. Another interesting consequence of the strong coupling between the plasmonic particles is that even when randomizing

the array and removing 95% of its particles lasing continues to take place with a moderate change in the lasing threshold.

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Appendix A: 1D Green's function

First we define the normalized longitudinal wavenumbers $\zeta_i^X = \sqrt{\epsilon_{ri} - \xi_X^2}$, with $X = TE/TM$ and subject to the radiation condition $\text{Im}\{\zeta_i^X\} \geq 0$. Then, the 1D Green's function used in Eq. (8) is given by

$$g(\omega, z, z') = \frac{1}{2} \frac{Z_2^X}{D_X} \left(e^{ik_z^X |z-z'|} + R_1^X e^{ik_z^X (2h-(z+z'))} + R_3^X e^{ik_z^X (z+z')} + R_1^X R_3^X e^{ik_z^X (2h-|z-z'|)} \right) \quad (9)$$

where h is the SU8 layer thickness and $k_z^X = k_0 \zeta_2^X$, and

$$R_i^X = \frac{Z_i^X - Z_2^X}{Z_i^X + Z_2^X}, \quad i = 1, 3 \quad (10)$$

$$Z_i^{TM} = \eta_0 \frac{\zeta_1^{TM}}{\epsilon_{ri}}, \quad Z_i^{TE} = \frac{\eta_0}{\zeta_i^{TE}} \quad i = 1, 2, 3 \quad (11)$$

and

$$D_X = \frac{d}{d\xi} (1 - R_1^X R_3^X e^{2ik_0 \zeta_2^X h}) \Big|_{\xi_X = k_X/k_0} \quad (12)$$

Appendix B: Ewald summation

The convergence of the infinite summation in Eq. (5) can be significantly accelerated by using the Ewald summation technique. First, we write

$$C(\omega, k_x, k_y) = 2A_{TE} \left(S(k_{TE}) + \frac{S_{xx}(k_{TE})}{k_{TE}^2} \right) - 2A_{TM} \frac{S_{xx}(k_{TM})}{k_{TM}^2} \quad (13)$$

with $k_{TE} = k_0 \xi_{TE}$, and $k_{TM} = k_0 \xi_{TM}$, and

$$S(k) = \lim_{x'y' \rightarrow 0} \sum' H_0^{(1)}(kR_{mn}) e^{id(mk_x + nk_y)}, \quad (14)$$

$$S_{xx}(k) = \partial_{x'x'} S(k) \quad (15)$$

Where $R_{mn} = \sqrt{(x' - md)^2 + (y' - nd)^2}$. The primed summation sign in Eq. (14) is used to exclude the $(m, n) = (0, 0)$ term from the infinite two dimensional summation. The summation can also be written as

$$S(k) = \lim_{x'y' \rightarrow 0} \sum H_0^{(1)}(kR_{mn}) e^{id(mk_x + nk_y)} - H_0^{(1)}(k\rho'), \quad (16)$$

where $\rho' = \sqrt{x'^2 + y'^2}$. The unprimed summation is used for infinite summation $(m, n) \in (-\infty, \infty) \times (-\infty, \infty)$. Next we replace the Hankel function by one of its integral representations

$$H_0^{(1)}(kR_{mn}) = -\frac{2i}{\pi} \int_0^\infty \frac{du}{u} e^{(k^2/4u^2 - R_{mn}^2u^2)} \quad (17)$$

Note that since $R_{mn}^2 > 0$, and assuming that $k^2 > 0$, to formally guarantee convergence of the integral representation in Eq. (17), we have to require that u goes to infinity along the line $\arg u = -\pi/4$. However, once we use this representation and derive an alternative, rapidly converging representation for the summation, we may apply Cauchy theorem and calculate the required integrals along a more convenient path. The semi-infinite integration path above is decomposed into two intervals, $0 \rightarrow E$, and $E \rightarrow \infty$, where E is an arbitrarily chosen constant picked as a trade-off between fast convergence of S_1 and S_2 . We define

$$S_1 = \sum -\frac{2i}{\pi} \int_0^E \frac{du}{u} e^{(k^2/4u^2 - R_{mn}^2u^2)} e^{id(mk_x + nk_y)} \quad (18)$$

$$S_2 = \sum' -\frac{2i}{\pi} \int_E^\infty \frac{du}{u} e^{(k^2/4u^2 - R_{mn}^2u^2)} e^{id(mk_x + nk_y)} \quad (19)$$

$$C = \frac{2i}{\pi} \int_0^E \frac{du}{u} e^{(k^2/4u^2 - \rho'^2u^2)} \quad (20)$$

such that $S = S_1 + S_2 + C$. Note that as long as $E \gg k/2$, the integration in the summands of S_2 yields a Gaussian decay of the summands with respect to the

summation indexes hence the summation over this part of the integral convergence rapidly. Similarly, the integration required to calculate C converges rapidly. The only issue left is the slow convergence of S_1 which is similar to the poor convergence of the original series. In this case, however, we are able to apply Poisson summation to accelerate the convergence. We obtain

$$S_1 = \frac{4i}{d^2} \sum_{p,q} \frac{e^{k_{pq}^2/4E^2}}{k_{pq}^2} \quad (21)$$

where $\mathbf{k}_{ppq} = (k_x, k_y) - 2\pi/d(p, q)$, and $k_{pq}^2 = k^2 - \mathbf{k}_{ppq} \cdot \mathbf{k}_{ppq}$, $p, q \in (-\infty, \infty) \times (-\infty, \infty)$. The convergence of the summation S_1 in its new representation is Gaussian, therefore, practically only a few terms are required.

Finally, we have $S_{xx} = S_{1,xx} + S_{2,xx} + C_{xx}$ where

$$S_{1,xx} = -\frac{4i}{d^2} \sum_{p,q} \frac{e^{k_{pq}^2/4E^2}}{k_{pq}^2} \left(k_x - \frac{2\pi}{d} p \right)^2 \quad (22)$$

$$S_{2,xx} = \sum \frac{4i}{\pi} \int_E^\infty du (1 - 2m^2 d^2 u^2) u \times e^{(k^2/4u^2 - R_{mm}^2 u^2)} e^{id(mk_x + nk_y)} \quad (23)$$

$$C_{xx} = -\frac{4i}{\pi} \int_0^E du u e^{(k^2/4u^2 - \rho^2 u^2)} \quad (24)$$

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