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Lasing and condensation in plasmonic lattices

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ABSTRACT

I review our recent findings on lasing / condensation in plasmonic nanoparticle lattices¹⁻⁵. The system properties can be tailored with high precision, including the lasing / condensation energies, linewidths, as well as the dimensionality of the feedback. For a 2-dimensional (2-D) square lattice, we identify lasing in the bright and the dark mode of the system¹. By reducing the dimensionality to 1-D we observe the dark mode lasing². In broken symmetry 2-dimensional rectangular lattices, we observe multimode lasing³. In honeycomb lattices with hexagonal symmetry, we observe 6 beams with specific off-normal angles and polarization properties corresponding to six-fold symmetry of such a lattice⁴. Finally, I review our recent studies in plasmonic Bose-Einstein condensation in plasmonic lattices⁵.

Keywords: plasmon, lasing, Bose-Einstein condensation, nano-optics

1. INTRODUCTION

Nanoscale lasing enables strong light-matter interactions in extremely small mode volumes. Strong ohmic and radiative losses have limited the applicability of plasmonic systems, in particular at visible frequencies. One approach to overcome these losses is to hybridize the lossy plasmonic particle resonance with a low loss diffracted order, resulting in a so-called surface lattice resonance (SLR). In periodic nanoparticle lattices, the SLRs originate from the *radiative* (and therefore polarization specific) coupling of individual nanoparticles. They exhibit high tunability of their resonance energies, linewidths, as well as the dimensionality of the radiative coupling present in the system. We show how, by tuning the lattice parameters, 1-dimensional lasing, 2-dimensional multimode lasing, as well as 2-dimensional phase correlated single wavelength lasing can be realized¹⁻⁴. Finally, I review our recent demonstration the plasmonic Bose-Einstein condensate⁵.

1.1 Square lattice with bright and dark mode lasing

The origins of the plasmonic lattice lasing lie in the *radiative coupling of the particles*¹. If the particle spacing equals the wavelength of radiation in the medium, then, at each particle location the radiation fields from all the other particles are interfering constructively. In an infinite lattice, with y-polarized radiation, the modes can be considered to be the result two counter propagating plane waves traveling to $-x$ and $+x$ directions. For the bright mode, the *antinode* of the resulting standing wave is located at the center of each unit cell (i.e., at the center of each particle). For the dark mode, the *node* is located in the center of the unit cell. In a finite lattice, however, the two counter propagating plane waves have equal magnitudes only in the center of the array. For the bright mode, this results in a maximal dipole moment in the center of the lattice. At the left (right) edge of the array, only left (right) propagating component is present, resulting to a reduced dipole moment and radiation. For the dark mode, the node at the center of the array results in quadrupolar excitation and negligible radiation. However, a gradual buildup of the dipole moment towards the edges of the lattice takes place, as one of the counter propagating plane waves becomes gradually weaker. Note, that in this case we considered y-polarized nanoparticles, who mainly radiate in x-direction, so the above reasoning should hold equally to a *1-dimensional (1-D) case*. We verified this by fabricating chains of particles, i.e., a 1-dimensional periodic lattice.

Figure 1. In a finite sized lattice, we observe 2 modes that lase at visible frequencies. One of the modes is bright (a), and another is dark (b). The dipolar component of the bright mode maximizes in the center of the lattice, which is manifested as a bright area in the center of the real space image of the sample (c). The dark mode, however, is composed of quadrupole resonances in each particle. In the center of the lattice, where the two counter propagating plane waves have equal magnitude, the quadrupole mode is “perfect”, and consequently the center of the lattice radiates very little light. At the left and right edge of the lattice, however, only one of the components of the two waves are present due to the absence of the particles. This induces a large dipolar component, which is seen as intense laser emission at both edges.

1.2 1-D lattice

For 1-dimensional case, we observe very similar behavior as for 2-dimensional case above². Namely, the dark mode emission intensity increases towards the edges of the lattice (a chain in this case). This naturally raises the question, whether the other direction of a 2-D lattice plays any role. To study this, we fabricated rectangular arrays with broken symmetry such that periodicity along x-direction is different from y-direction.

Figure 2. In a finite sized 1-dimensional lattice, we observe a dark mode that lases at visible frequencies. The schematic of the pump geometry is shown in a). Below the threshold, we observe spontaneous emission mainly from the area of the pump beam, see b). The dark mode, whose origin is in quadrupole resonances in each particle, radiates very little light from the center of the chain, even with pump fluence well above the threshold, see c). At the top and bottom end of the lattice, however, only one of the components of the two counter propagating waves are present due to the absence of the particles, which manifests itself as gradually increasing laser emission towards the edges d).

1.3 2-D lattice

The fabricated rectangular arrays with broken symmetry were designed in such a way that while periodicity along x-direction is different from y-direction, we can nevertheless make both of them lase³. This was ensured by having a sufficient overlap of the periodicity dependent resonances with the gain profile of the used emitter. In this case, we observe 2 lasing wavelengths, corresponding to the 2 periodicities of the lattice. While the system is 2-D, we expect the 2 lasing directions be independent from each other in Fig. 3 (a, b), as $p_x \neq p_y$. In the case of Fig. 3 c), however, as the wavelengths are the same, it is nontrivial to say whether the 2 lasing modes are independent. In principle, system could either exhibit lasing in 2 directions which are independent of each other, or correlated lasing, where the 2 directions are phase locked. To study this in detail, we fabricated honeycomb lattices, which in essence enable radiative coupling in 2 dimensions.

Figure 3. In a broken symmetry 2-dimensional lattice, we find 2 modes that lase at visible frequencies. The x-periodicity was kept constant (380 nm), while the y-periodicity was varied. In a), we observe 2 modes lasing, with approximately 15 nm difference in lasing wavelength. In b) the difference 7.5 nm, and in c), the modes have the same lasing wavelength. This is expected as the lasing wavelength λ should depend on the periodicity p and refractive index ($n = 1.5$) as $\lambda = np$. The theoretical band structures agree well with the observed ones (d-f).

1.4 Phase locked 2-D lattice

The fabricated honeycomb lattices enable efficient radiative 2-D coupling due to hexagonal symmetry: A radiating dipole has significant intensity to angles that are 60 degrees away from the dipole orientation (in contrast to rectangular array, where the radiative coupling along the direction parallel to the dipole is negligible)⁴.

Figure 4. In a honeycomb lattice, a radiative coupling in 2 dimensions takes place. The observed lasing pattern consists of 6 beams corresponding to the 6 principal lattice directions, see a). A Fourier image and polarization analysis reveals each of the beams having a linear polarization, see b). The unpolarized realspace imaging c), as well as the polarized ones, d), e), are in good agreement with the calculated ones f-h).

We note that similar lattices can be utilized to create a band gap by the design of the nanoparticle shape. Recently, we demonstrated a plasmonic Bose-Einstein Condensate, exhibiting ultrafast, sub-picosecond dynamics for the thermalization and condensation⁵.

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