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Communication

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Symmetry Breaking in Oligomer Surface Plasmon Lattice Resonances

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Abstract

We describe a novel plasmonic mode engineering, enabled by the structural symmetry of a plasmonic crystal with a metallic oligomer as unit cell. We show how the oligomer symmetry can tailor the scattering directions to spatially overlap with the diffractive orders directions of a plasmonic array. Applied to the color generation field, the presented approach enables the challenging achievement of a broad spectrum of angle-dependent colours since smooth and continuous generation of transmitted vibrant colors, covering both, the cyan-magenta-yellow and the red-green-blue color spaces, is demonstrated, by scattering angle- and polarization-dependent optical response. The addition of a symmetry driven level of control multiplies the possibility of optical information storage, being of potential interest for secured optical information encoding but also for nanophotonic applications, from demultiplexers or signal processing devices to on-chip optical nanocircuitry.

Keywords: aluminum plasmonics, symmetry breaking, lattice resonances, plasmonic color generation, diffractive coupling, scattering

Plasmonic nanostructures offer uncountable opportunities for optical properties engineering towards applications in nano-optics, nano-photronics and biosensing. Shape, size, composition and local environment have been known since long time as tools for the tuning of optical resonances, named Localized Surface Plasmon Resonances (LSPRs). More recently, arranging single particles in periodical arrays has allowed to introduce new intriguing effects arising from diffractively coupled resonances known as surface plasmon lattice resonances (SPLRs)1,2. In these systems, the light scattered by each nanoparticle into the array plane is in phase with the plasmon resonance of the neighboring structure, leading to remarkable improvements of resonance line-shape and quality visible in reflection, transmission, extinction and absorption. An important application field of these concepts is the plasmonic color generation.3–8 Plasmonic patterns can work in additive (reflection) or subtractive (transmission) colour filter mode depending on the engineered
scattering and absorption properties, usually operating in the red-green-blue (RGB) and cyan-magenta-yellow (CMY) colour space, respectively. This technology opened the way towards great potential applications such as high-density data encryption storage,\textsuperscript{9,10} anti-counterfeiting devices and security tags,\textsuperscript{11–13} colour printing and high resolution displays\textsuperscript{6,9,14–19} and plasmonic colour sensors.\textsuperscript{20}

Spectrally resonant coupling between LSPR and diffraction order (DO) modes of periodic lattices, i.e., SPLRs, allowed to reach high colour vibrancy in both transmission\textsuperscript{21–23} and reflection/scattering\textsuperscript{24–26}. Full colour palettes have been achieved\textsuperscript{15,21–23,25–28} by properly combining the fundamental geometrical features of plasmonic nanopixel, i.e., nanostructures size (to tailor the LSPR) and lattice period (to set the DO dispersion).

On the other hand, if the periodic arrays consist of complex multiparticle unit cells, a broader optical properties scenario can be revealed, as determined by the system architecture and by mutual interparticle positions in the unit cell.\textsuperscript{29}

Lattices with narrow-gap and non-trivial geometries have shown interesting properties, including the polarization control of the modes quality factor (Q) and effective mode volume\textsuperscript{30,31}.

In particular, when exploring complex nanoparticles assemblies the specific symmetry of the unit cell can represent an additional degree of freedom which has been not fully explored so far, and which can affect the near and far field response of plasmonic lattices, finding novel applications in the field of nanophotonics and plasmonic color generation.\textsuperscript{3–8}

Here, we present and discuss a novel mechanism for engineering collective plasmon resonances in a plasmonic crystal, by exploiting the symmetry breaking of its multiparticle unit cell in the form of an aluminium tetramer.\textsuperscript{22,32–35}

In particular, we will show that symmetry breaking allows to drive the tetramer resonant scattering directions, and, in turn, to enable their spatial overlap with the DO directions. This design can be used for the generation of plasmonic colors characterized by an additional degree of freedom for the color tone, depending not only on the single particle geometry and lattice size, but also on the mutual multiparticle position within the unit cell. In particular, the discussed novel mode engineering allows a smooth and continuous generation of transmitted vibrant colours, covering the CMY colour space, while, by exploiting the in-plane rotation of the nanocluster symmetry axis, even RGB colours can be revealed at off-axis angles. The reported scattering angle- and polarization-dependent optical response of the system can add advanced functionalities to the colour generation, allowing the challenging achievement of broad spectrum of angle-dependent colours and multiplying the capability of stored optical information density.

The fundamental unit of our system consists of a cluster of four aluminium nanodisks.\textsuperscript{36–39} Aluminium has been chosen because of its plasmonic response in the visible spectral range, without the instability problems\textsuperscript{40} of silver (i.e., uncontrolled oxidation or sulfidation). The nanodisks are arranged in a rhombic configuration (figure 1a) with $C_{2v}$ group symmetry, on a silica glass substrate, with the nanodisk diameter $D$ and height $H$ fixed at 100 nm and 40 nm,
respectively. The core dimer has a nanodisk spacing $g$ of 20 nm. This architecture exhibits two orthogonally polarized plasmonic modes, as numerically calculated in figure 1b: a double and broad LSPR, polarized along Y direction, originated from the overlap between the core dimer and single nanodisk peaks (close to 700 nm and 500 nm, respectively as shown in figure S1) and a broad LSPR polarized along the X-axis. At a first level, if the proposed unit cell is integrated within a periodic array, with lattice periods $P_x$ and $P_y$ to create a rectangular nanopixel, the resonant overlap between the LSPR and the DO modes can be obtained either varying $D$ while keeping the lattice period constant, or, similarly, by keeping $D$ constant and changing $P_x$. Numerical calculations reported in supporting information section (figure S2 and S3), as well as experimental measurements that will be discussed in the following, show how the achievement of resonant coupling between the LSPR mode and the lattice diffractive orders affects the functionalities of the proposed structure towards the generation of a wide range of colour tones, with engineered properties in terms of transmission bandwidth and wavelength. Moreover, given the structure symmetry, the proposed pixels behave as polarization dependent dual color filters, similar to the ones observed in cross-shaped nanoapertures in agreement with recent literature results\textsuperscript{22,27}

**Figure 1** a) Sketch of the tetramer unit with nanodisk diameter $D$, height $h$ and inter-particle gap $g$. b) FDTD simulation of polarization dependent transmission spectra (expressed in arbitrary units as a function of the extinction cross section $\sigma_{ext}$) of the single symmetric oligomer. c) Scanning electron microscopy image of the rectangular array configuration with designed in plane lattice periods $P_x$ (450 nm) and $P_y$ (340 nm) that define the fundamental nanopixel, and d) corresponding normal incidence transmission spectra measured with light polarized along the two orthogonal oligomer axes and normalized to the substrate\textsuperscript{11} is the
light intensity through the array and \( I_0 \) is the light intensity through the unpatterned substrate). The coloured arrows evidence the BTW, GTW and RTW. As expected, it is evident the improvement of plasmonic features with respect to resonance strength and linewidth when passing from a single structure to an array configuration.

In particular, we experimentally realized a 50 \( \mu m \times 80 \mu m \) periodic array, shown in figure 1c. As design criteria for the nanopixel, we considered the DOs momentum dispersion in a homogeneous medium with refractive index \( n \):

\[
\lambda_{DO} = \frac{2\pi n}{\sqrt{(k_x + N_x P_x)^2 + (k_y + N_y P_y)^2}}
\]

where \( k_x \) and \( k_y \) are the k-vector components of the incident light, and \( N_x, N_y \) identify the diffraction orders in the xy basis.\(^{31}\) By the proper choice of the two lattice periods \( P_x \) and \( P_y \), it is thus possible to tune the DOs dispersions in the visible spectral range, as described in figure S2. In particular, with \( P_x=450 \) nm and \( P_y=340 \) nm, we are engineering, for normal incidence \( (k_x=k_y=0) \), three transmission windows as highlighted in figure 1d: a Red Transmission Window (RTW) by setting the \((\pm1, 0)\) DO wavelength at 680 nm, a Green Transmission Window (GTW) by setting the \((0, \pm1)\) DO at 515 nm, and a Blue Transmission Window (BTW) by setting the \((\pm1, \pm1)\) DO wavelength at 415 nm. Note that these diffractive modes are characterized by a precise polarization state, i.e., aligned along the Y-axis for the \((0, \pm1)\) mode and along the X-axis for the \((\pm1, 0)\) mode. The \((\pm1, \pm1)\) mode is oriented along different in-plane angle (\( \alpha=37^\circ \), figure S4).

However, what mainly distinguishes our structure from previous works is the multiparticle unit cell providing, as shown below, the additional possibility for a mode plasmon-photon hybridization tuning driven by the spatial overlap between the radiating LSPR and the DO of the lattice. Indeed, as shown below, by precisely shifting the satellite nanodisks along the y direction, introducing a displacement \( S \) from the central position (figure 2a), one can modify the structure symmetry and control the scattering angular direction of the LSPR.\(^{41,42}\)
Figure 2. a) Scheme of the structural change induced by the displacement parameter (S) applied to the satellite disk along the Y-axis: the axis of the large gap dimer is progressively rotated with respect to X-axis and the structure symmetry passes from a $C_{2v}$ to a $C_2$ symmetry group. b) SEM images of experimentally realized oligomers as a function of S and related transmission curves, FDTD simulations and measurements for X-polarization (left side) and Y-polarization (right side). Transmission curves are vertically stacked for clarity, whereas measured and simulated data are reported for completeness in the supporting information section (Figure S6). Magnified colour pictures obtained with the microscope in transmission mode for each array are also shown.

15 oligomer arrays with S varying from 0 to 140 nm, with 10 nm steps, have been fabricated on a glass substrate and optically characterized in an homogeneous environment (see details in the Methods section). The X-polarization transmission at $S=0$, shown in Figure 2b, is characterized by a normal mode splitting originating from the hybridization between the oligomer LSPR and the lattice $(0, \pm 1)$ DO. Such coupling, also discussed in the supporting information section (figure S2 and S3), results in the formation of the upper and lower SLPR branches, positioned around the GTW, and labelled as SLPR-1 and SLPR-2 on the spectra in Figure 2b. Similarly to what observed in close gap dimer arrays $^{31}$, the coupling strength between the LSPR and the $(0, \pm 1)$ DO can be assumed as directly proportional to the energy...
gap of these two hybrid modes (i.e., the related transmission valleys in the spectra). Indeed, as the satellite displacement $S$ increases (Figure 2b, left panel), the coupling strength between LSPR and $(0, \pm 1)$ DO progressively decreases and the mode splitting decreases accordingly. For $S=80$ nm, the two hybrid modes converge into a single resonance valley close to the energy of $(0, \pm 1)$ DO. For $S>80$ nm, two modes re-originate with different mixing fractions with respect to the initial configuration\textsuperscript{31}, as confirmed by the modes relative linewidth. In particular, for $S=140$ nm the lower energy SLPR centred at 560 nm exhibits a significant spectral narrowing with respect to the broad mode at the same energy for $S=0$, indicating a non-trivial and symmetry-driven change in the optical characteristics of the SLPR mode excited in the system.\textsuperscript{2} The maps shown in the supporting section (figure S5) clearly highlight this evolution. Later we will discuss in details how the origin of this effect can be ascribed to the variation of the LSPR scattering pattern with respect to the directions of the pixel DOs, thus resulting in a variable plasmon-photon nature of the hybridization in the lattice.

The spectra clearly evidence how effective can be the breaking of oligomer symmetry in tuning the chromatic response. For $X$-polarized light, the pixel colour undergoes a smooth and continuous transition in the Cyan-Magenta-Yellow colour space as the disk satellites are moved far from the central position. In the $S=0$ condition, the main asymmetric resonance, generated by the LSPR/DO overlap, provides a vibrant violet colour tone. The appearance of the other spectral modulations as a function of $S$ contributes to transmitted colour evolution including other components from the red side of the visible spectral range, ending up to a yellow colour tone for the maximum $S$.

Switching to $Y$-polarization, at $S=0$, the $(\pm 1, 0)$ DO, centred at 680 nm (RTW) is excited. Here, by increasing $S$, the transmission spectrum remains almost unchanged. Moreover, also the $(\pm 1, \pm 1)$ DO component along the $Y$-axis doesn’t introduce spectral variation with $S$ increment. Consequently, an extremely weak colour change around a yellow tone is recorded as a function of $S$.

Thus, this structure operates as a dual state plasmonic nanopixel, showing a fine and continuous colour gradient control with $X$-polarized light, spacing in the Cyan-Magenta-Yellow space, and an almost constant $Y$-polarization response, firmly set on a specific colour tone (yellow).

The proposed multidisk structure can be particularly advantageous for color generation with respect to other efficient diffractively coupled plasmonic filters, such as recently reported nanopatches\textsuperscript{23}, nanodisks\textsuperscript{21}, and nanocrosses\textsuperscript{5,22}. In fact, the far field spectrum of the array takes advantage of the diffractive coupling, which can lead to large quality factor values, However, the far field scattering can be positively influenced also by the presence, in the unit cell, of multiple close-gap nanodisks, giving rise to localized electromagnetic field hot spots within the gaps\textsuperscript{30,43,44}. This combined effect leads to the narrow and well defined transmission peak in the VIS, present in experimental spectra, and even more apparent in numerical simulations (figure 2b and, in supplementary section, figure S6), where fabrication defects and tolerances are not taken into account.
Since geometry influences the scattering behaviour of plasmonic systems \(^{41,42,45}\), we performed numerical calculations by using the T-Matrix method\(^{46}\) (see the Methods section) for the scattering directivity of the isolated oligomer varying from the rhombic configuration \(C_{2v}\), with \(S=0\), to \(C_2\) for \(S\neq0\). Mode hybridization (between the cluster elements)\(^{36,37}\) and symmetry breaking are able to effectively affect the near-field scattering behaviour of the single oligomer, resulting in asymmetric radiation patterns, as shown in figure 3 for different regimes.\(^{47,48}\) Analysing this effect in the far field at the three transmission windows, we note that the different symmetry tunes the overlap between the radiating directions of the plasmon mode and of the diffractive orders. In fact, for GTW (X-pol, figure 3a), the far field scattering pattern of the symmetric (\(S=0\)) oligomer is aligned to the (0, ±1) DO direction. As \(S\) is increased, the main scattering direction rotates, thus deviating from the DO propagation direction, and secondary, lower intensity scattering directions appear. Similarly, for RTW (Y-pol, figure 3b), the scattering is initially directed along the X-axis, and then rotates by increasing the lateral displacement.

For BTW (X-pol, figure 3c), the scattering radiation at the reference configuration (null displacement) is still oriented along the Y-axis. By increasing \(S\), the plasmons mainly scatter along the (±1, ±1) DO mode direction, while additional out of axes directions arise. Conversely, under Y-polarized excitation, at this wavelength, the oligomer shows limited scattering efficiency (above all for \(S=0\)) and negligible scattering directional changes with \(S\) (Figure S7).

In particular, figure 3d summarizes the evolution of scattering far field intensity by the oligomer, calculated as a function of \(S\). Here, we observe a decrease in the far field intensity of the GTW mode in the (0, ±1) DO direction when increasing \(S\), that is quite coherent with the decrease in coupling strength between this mode and the LSPR mode, as observed in transmission spectra (figure 2b, X-polarization). The BTW mode in the (±1, ±1) DO direction experiences an opposite trend with \(S\). In particular, for \(S=70\) nm, BTW and GTW curves cross with each other and approach RTW value (in the (±1, 0) DO direction), which, in turn, is constantly low, given the weak interaction, for any \(S\), with the tetramer LSPR (Figure 2b). This demonstrates also that, at \(S=70\) nm, the LSPR scattering direction lays in between the two (0, ±1) and (±1, ±1) DO directions, thus reducing its spatial overlap with any DOs. Further \(S\) increment up to 140 nm fully aligns the LSPR scattering along the ((±1, ±1) DO direction.

Therefore, one can conclude that the ability of individual oligomer to scatter light in the different DO directions, as a function of \(S\), can determine a different coupling strength between the LSPR mode of individual tetramer and the photonic modes, associated to the in-plane diffraction of the periodic lattice. As a consequence, when integrating these structures in ordered arrays, the displacement parameter \(S\) becomes an additional degree of freedom for the control of the overall spectral response, including colour generation, through the tuning of the in-plane scattering directions and the related LSPR/DO hybridization efficiency. Thus, the symmetry drives the spectral and spatial overlap of resonances with different character (localized and plasmonic on one side, and photonic and disperse on the other side).
Figure 3. Near and far field ($E$) simulations of single clusters excited at (a) the GTW with a X-polarization, (b) the RTW with a Y-polarization and (c) the GWT with a X-polarization. In each case the far field scattered light magnitude (top left) is plotted as a function of the in-plane angle and $S$, and the corresponding polar plots (bottom left) are also displayed at three significant $S$ values (0, 70 nm, 140 nm) for which the near-field magnitude distributions (right) are also given (using a logarithmic scale). d) Complete calculated evolution of scattering far field magnitude of the oligomer as a function of $S$, along the three different DOs directions.

As a further step, we analysed the in-plane coupling between plasmonic and photonic resonances in the array configurations. To this purpose, the electric field magnitude $|E|^2$ in the X-Y plane was calculated at the SLPR-2 excitation as a function of structure symmetry (Figure 4 a-c). For $S=0$, the resonant condition between LSPR and $(0, \pm 1)$ DO, discussed in the previous figures, leads to the electric field localized both, at the edge of the nanostructures and
between the tetramers, with a standing wave pattern directed along the Y-axis (figure 4a). By increasing S, transmission spectra show the convergence of the two SLPRs into a broad plasmonic resonance, centred at the GTW: accordingly, the electric field mainly concentrates in the nanostructure hot-spots (figure 4b), with an additional in plane diffuse interference pattern, rotated by the modified structure symmetry. Indeed, the simulations in figure 3a show a reduction of oligomer scattering intensity along the (0, +1) direction with S, thus leading to a consequent lower overlap between plasmonic and photonic resonance. At the maximum S, the electric field distribution assumes a more pronounced photonic component with respect to previous case (figure 4c).

Given the dispersive character of the SLPRs, typically resulting in strongly angular dependent transmission colours, a further indication of the displacement S effect on the spatial overlap between LSPR and DOs, as well as on their coupling strength, is experimentally provided by imaging the transmission of the lattices in the Fourier space, as also shown in figure 4, lower panels. Here we have filtered white transmitted light with bandpass optical filters, around the hybrid modes energy (see Methods section for details).

In particular, in Figure 4d-f the Fourier space image of transmitted light is shown for the lattice with S=0, 70 nm and 140 nm, respectively. A well-defined angular dispersion of the hybrid mode appears at S=0, and is well fitted with the dispersion of the corresponding (0, ±1) DO (shown as white lines in figure 4d). This is a clear signature of LSPR-DO coupling since the plasmon mode does not exhibit angular dispersion. As S is increased, the tetramer scattering rotation results in a more blurred transmission pattern, since the spatial overlap of the plasmon scattering radiation with the (0, ±1) DO is reduced, while its interaction with the (±1, ±1) DO is progressively strengthened (figure 4e and f).

For Y-polarization, at excitation corresponding to the (±1, 0) DO, the electric field distribution is only photonic, independently on S. Correspondingly, no variation occur in the Fourier space. This effect can be attributed to the scattering intensity in the (±1, 0) DO direction being quasi-independent from the satellite displacement (figure S8 and S9).
Figure 4 Simulations of the electric field magnitude (in logarithmic scale) distributions in (x,y) plane (a, b, c) and Fourier space transmission maps (d, e, f), with overlapped the fitting of the DO dispersions at the SLPR-2 for S=0(a,d), S=80 nm(b,e), S=140 nm(c, f), respectively. For S=0, the electric field is localized at the nanoparticle edges and in the interparticle space exhibiting a standing wave pattern. When S increases, the electric field is mainly concentrated at the nanodisks and the pattern is rotated due to the symmetry change. Similarly, the Fourier images show the evolution of the symmetry driven LSPR/DOs coupling. At S=0, the SLPR-2 exhibits the dispersive character of the \((0, \pm 1)\) DO while, increasing S, the plasmon scattering rotation triggers a coupling condition with higher DO orders.

Finally, in order to effectively demonstrate all the makings of the proposed structure in terms of colour generation, we fabricated arrays with different nanodisk diameter (from 95 nm to 150 nm) and different displacement, while keeping the same lattice periods. A wide range of polarization sensitive colours can be observed in the transmission images (figure 5), covering the full visible range. In particular, for X-polarization we still note the smooth colour gradient enabled by the particle displacement, while for Y-polarization, the colour is again almost independent on the structure arrangement and driven only by the nanodisk diameter.

Hence, as an ultimate limit, the colour information can be encoded not only in nanocluster dimensional parameters, considering the structural variation of diameter and lattice period (two dimensions), but also in the particle displacement-dependent structural symmetry (third dimension), building a more complex 3D colour table with ultrafine tonal gradient, towards a wide colour gamut. Given the angular dependence of scattering in the presented oligomer, it is
possible to extend the colour palette to the RGB space by working in crossed polarization configuration (figure 5b-c).

Here, the incident X-polarized light experiences a wavelength dependent polarization rotation and/or ellipticity formation by interacting with the cluster arrays. This process, enabled by the structure asymmetry, can be more formally described as a transmission amplitude (linear dichroism) and/or phase (birefringence) difference between two orthogonal components of the incident electric field oriented at -45 and +45 degrees relative to the X-axis. As a result, the tetramer lattices at the symmetric condition S=0, highlighted in figure 5b, clearly appear dark. Increasing S, i.e., changing the cluster symmetry, the pixels are coloured as a result of polarization change of incident light. Most notably, RGB colours have been also obtained in such experimental configuration (figure 5c), even if working as transmission (i.e., subtractive) colour filters.

Overall, the intuitive design rules, applied to the proposed Al tetramer geometry, have allowed to generate a colorimetric mechanism of information encoding, based on the association between the complete set of nanopixel parameters, and the corresponding colour, spectrum and angular dispersion. In this sense, the structural displacement (S) represents an additional security level (or dimension) since it can be uniquely retrieved by multi-level optical decoding. These plasmon rulers could be employed to build a lookup table to document the colour spectra associated to each structural arrangement. By this way, complex covert optical signatures for anti-counterfeiting applications are enabled with respect to the polarization dependent security pattern, and concealed messages can be revealed under angle-sensitive optical conditions and by comparison with stored colour spectra.

Furthermore, the symmetry-breaking paradigm applied to plasmonic oligomers and to their scattering properties can open a novel route toward a large number of nanophotonic applications, ranging from demultiplexers or signal processing devices to on-chip optical nanocircuitry.
Figure 5 a) Bright-field microscope images of the polarization dependent dual state color palette. The nanopixel geometry has been changed to generate a color range (CMY color space) spanning the visible spectrum. Moving from left to right, the nanodisk diameter increases, while from bottom to top the satellite displacement is increased. In particular, in the X-polarization case, for each column (same diameter) a continuous and ultrafine color variation is shown as a function of the structural symmetry variation, while in the Y-polarization the color tone is quasi unchanged. b) Crossed polarizers color palette image as a function of the nanodisk diameter and S variation. The white box highlights the color pixels at the S=0 state that clearly appear dark since the plasmon scattering direction is oriented along the two orthogonal axes, while increasing the S value, a gradient of vivid colors come out, as a consequence of the symmetry driven scattering direction, spanning the RGB space as shown in c) the CIE maps.


**Methods**

**Sample fabrication.** Two-dimensional arrays of aluminum tetramers were realized by electron beam lithography on a glass substrate with 40 nm height, 20 nm gap, nanodisk diameter ranging from 95 to 150 nm and lattice period $P_y=450$ nm and $P_x=340$ nm. The substrate was cleaned in acetone and 2-propanol and then a 250 nm poly(methyl methacrylate) (PMMA) layer was spin-coated at 6000 rpm and soft-baked at 180 °C for 3 min. A 2 nm thick chrome layer was thermally evaporated onto the PMMA to prevent charge effects. After electron exposure, the Cr layer was removed and the resist was developed. Finally, after thermal evaporation of 40 nm of aluminum, a lift-off process was performed. The array size was 50 $\mu$m x 80 $\mu$m, consisting of about 27000 elements, can be considered large enough to observe the effect of surface lattice resonances in the transmission spectra $^{50,51}$

**Modelling.** Electromagnetic simulations have been performed assuming infinite array size (given the large number of fabricated elements) using a commercial finite difference time-domain-based software (Lumerical FDTD Solutions) and the T-matrix method$^{46}$. The nanoparticles are considered here as having an oblate geometry, with semi-axes equal to 50 nm (particle radius D/2) and 20 nm (half-height H/2), and embedded in a homogeneous medium having a refractive index of 1.515. The used aluminum refractive index is from Ref $^{52}$. In the text, the near-field magnitude refers to the electric field magnitude $|E|^2$ around the particles. In turn, the far-field magnitude (or equivalently far-field pattern, or scattering amplitude) corresponds to the electric field magnitude $E_\infty$ at infinity ($r\rightarrow\infty$) in a given direction, omitting the $\exp(ikr)/r$ term.

**Optical characterization.** The transmission measurements have been carried out on an homemade confocal setup based on an optical microscope (Zeiss AxioScope A1) coupled to an imaging spectrometer. Light from a tungsten lamp was focused on the sample by an adjustable numerical aperture condenser (NA from less than 0.1 to 0.95). Transmitted light was collected with a 40×0.95 NA objective lens and a three lenses system was employed to confocally collect light and focus the real image of the sample on the spectrometer entrance slits. Transmission spectra are obtained by normalizing the intensity transmitted by the arrays to the intensity transmitted by the un-patterned substrate. Both the real and the Fourier space imaging can be obtained on the setup, the former by using all the three lenses, the latter by removing the intermediate lens and selecting the region of interest in the intermediate real image plane. The Fourier space images have been obtained by filtering white transmitted light with bandpass optical filters centered at 470 nm and 560 nm. Color images are taken in transmission mode with the same microscope (20x, NA=0.65 objective).

Supporting Information Available: Calculated extinction cross section of single nanodisk and dimer; calculated response of the 2D colour palette driven by size and periodicity with the tetramer in symmetrical configuration; Calculated transmission maps of arrayed oligomers as a function of diameter and lattice period; discussion on the nanopixel engineering criteria; Measured transmission maps of fabricated array samples as a function of displacement parameter (S, from 0 to 140 nm)) and for the two incident polarizations (a, X-polarization, and b, Y-polarization); Far field simulations of scattering intensity distribution and near field behaviour of the cluster excited at the BTW (Y-polarization), as a function of S; electric field distribution maps calculated at excitation wavelengths corresponding to different lattice regimes; Measured Fourier images of the arrays as a function of displacement S and at excitation wavelength of SLPR-1 for X-polarization and of RTW for Y-polarization.
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