Symmetry-breaking induced magnetic Fano resonances in densely packed arrays of symmetric nanotrimers

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Due to unique properties and great design flexibilities, Fano resonances represent one of the most promising optical features mediated by metallic nanostructures, while the excitation of some Fano modes is impossible due to symmetry reasons. The aim of this work is to show that dense lattice arrangements can have a profound impact on the optical properties of nanostructures and, in particular, can enable the excitation of otherwise dark modes. Here, we demonstrate this concept using the example of rectangular arrays of symmetric trimers packed so densely that the coupling between neighbouring unit cells imposes a symmetry break, enabling the excitation of magnetic Fano resonances. We found that in experiments as well as in simulations, electric and magnetic Fano resonances can be simultaneously formed in cases where the inter-trimer distances are sufficiently small. By analysing the transition from an isolated trimer mode into a regime of strong near-field coupling, we show that by modifying the rectangular unit cell lengths due to the symmetry mismatch between lattice and trimer, two types of Fano resonances can be found, especially magnetic Fano resonances with loop-type magnetic field distributions within the centre of each trimer, which can be either enhanced or suppressed. In addition, the influence of the refractive index environment was measured, showing sensitivity values of approximately 300 nm/RIU. Our work provides fundamental insights into the interaction of the lattice and nanostructure response and paves the way towards the observation of novel optical excitations.

In recent years, nano-optics has attracted substantial attention, with a large number of studies¹–³ carried out to understand the interaction between light and matter on the nanoscale. By using well-designed plasmonic nanoantennae⁴,⁵, sophisticated photonic functionalities have been realized on ultrasmall geometric footprints on the basis of, e.g., deep sub-wavelength metasurfaces⁶,⁷, hybrid plasmonic nanosensors⁸, and monolithic fibre nanoprobes⁹.

One particular striking effect mediated by nanostructures is the Fano resonance, which arises from interference between a broadband bright mode and a narrowband dark mode¹⁰,¹¹. This modal interaction dramatically changes the lineshape of scattered light by introducing a narrow dip into the extinction spectrum. Due to the low damping of the dark mode, the electromagnetic near-field at the wavelength of the Fano resonance is greatly enhanced, which has the consequence that the scattered light is enhanced accordingly¹². To achieve such a unique effect, numerous types of nanostructures such as single split nanodots¹²,¹³, nanocavities¹⁴, nanosphere dimers¹⁵, dolmen-like nanostructures¹⁶,¹⁷, plasmonic clusters¹⁸ and oligomers¹⁹,²⁰ are currently being employed. Among these, nanotrimers consisting of three close-packed nanoparticles represent a promising design for realizing the Fano resonance (details of the hybridization are discussed in the Supplementary Information sec. 6). As first experimentally demonstrated in 2010 for the example of self-assembled metal-dielectric spheres²¹, trimers can...
exhibit a strong magnetic resonance. Later, several studies tried to obtain this magnetic enhancement in trimers by symmetry breaking within the trimer and have successfully shown excitation of magnetic-based Fano resonances with the magnetic resonance being the dark mode. In contrast to an internal symmetry break, the aim of this study is to show that excitation of magnetic Fano resonances in symmetric trimers can be achieved by placing them into densely packed arrays, with the symmetry breaking induced by the lattice arrangement. Here, the trimers are arranged in a rectangular lattice (two-fold symmetry) breaking the three-fold symmetry of the otherwise symmetric trimer unit.

For a majority of the Fano resonances mentioned above, arrays of trimer arrays are used where inter-trimer distances are chosen such to significantly exceed the individual trimer resonance wavelength. In a denser configuration, the compact geometrical arrangement leads to strong near-field unit cell interactions, with the diffractive coupling of localized plasmonic resonances further modifying the resonance lineshape. For instance, Chen et al. have demonstrated that the optical response of arrays of nanodolmens can be significantly altered by the near-field coupling to adjacent unit cells. By varying the periodicities, the far-field response of the plasmonic system can be evolved into a tight-binding regime or a hybridization regime showing a combined response of the individual trimer response and of the diffraction of the array. However, a detailed investigation for this type of hybrid response between nanostructures and inter-unit cell diffractive coupling has not been intensively carried out to date.

In this report, we reveal the optical properties of densely packed arrays of nanotrimers and their dependence on the geometrical parameters, such as intra-trimer gaps and array pitch. We conducted a set of experiments as well as numerical calculations with a focus on the trimer modes and found that magnetic Fano resonances can be excited for the symmetric trimer geometry when the trimers are arranged in densely packed arrays, with the symmetry breaking induced by the lattice arrangement and not by the individual trimer unit. The report is arranged as follows: First, we analyse the extinction spectrum of the trimer array and find a magnetic Fano resonance mode, identified by its experimental spectral features and simulated near-field distributions. Second, to further explore the impact of the near-field coupling effect on Fano resonances, extinction spectra with varying trimer-to-trimer distances were modelled and analysed. Finally, the trimer arrays were optically characterized under various refractive index (RI) environments. The results indicate that an asymmetric refractive index arrangement between substrate and superstrate weakens the formation of magnetic Fano resonances.

Design and Fabrication
The trimer array is fabricated by lift-off technology using electron beam lithography and dry etching (a sketch of one sample is shown in Fig. 1). The fabrication details can be found in the Supplementary Information (SI) in Sec. 3. The array of trimers has a size of $110 \times 110 \mu m^2$ covering the beam spot of the $20 \times$ objectives used in the optical characterization experiments. Each trimer unit is composed of three identical planar gold nanodisks (diameter 425 nm) with a G1 (intra-trimer gap, sketch in Fig. 1(b)) of 75 nm. The choice of a gold film thickness of 40 nm was mostly based on preliminary fabrication experiments (i.e., this thickness gave the best nanostructures) and has no substantial influence on the mode formation discussed in the remaining part of the manuscript. For the specific sample used in the characterization and RI sensing experiments, the value for G2 (inter-trimer gap, sketch in Fig. 1(b)) was chosen to be 175 nm, corresponding to an array pitch $\Lambda$ of 1100 nm. The scanning electron micrograph (SEM) image shown in Fig. 1(b) clearly demonstrates the compactness of the trimer array. Note that in all the extinction data, the polarization of the incident light is along the x-axis.
Discussions

Magnetic Fano resonance in closely packed trimer arrays. As a first step, we characterize the optical response of the trimer array (D: 425 nm H: 40 nm G1: 75 nm) for a fixed pitch (1100 nm, G2: 175 nm). In Fig. 2, we show the measured and simulated trimer extinction spectra (from 1600 nm to 2100 nm) together with the simulated near-field patterns at selected spectral positions. Within this specific spectral domain, each individual nanoparticle of one trimer behaves like a dipole, meaning that a more complex (multipolar) response for the individual nanodot does not need to be considered. According to the spectra shown in Figs S1 and S2 (Supplementary Information section 1 and 2), the spectral response of closely packed trimer arrays exhibits a highly asymmetrical lineshape and a strong polarization dependence, which contrasts with the isolated trimers discussed in refs 31–34. Hence, we carried out a set of studies with a focus on these densely packed trimers. In addition, only x-polarized incident light is applied since the polarization influence is beyond the scope of this work.

To obtain a homogeneous RI environment along the vertical direction (Fig. 2(a)), the trimer samples are immersed in a liquid that matches the RI of the substrate (n = 1.44). For spectral measurements, a commercial broadband supercontinuum source (NKT Photonics SuperK Compact) was employed for illumination, with the transmitted light recorded by a spectrometer (Instrument Systems SP320-124). Two transmitted spectra were recorded with one beam directed to the respective trimer array and the other passing through an unstructured part of the silica substrate. The experimental transmission T is then obtained by the power ratio of the two spectra. Please note that in this report, we present extinction spectra obtained using (1 – T).

Corresponding numerical calculations (Fig. 2(b)) based on the finite-element-method (FEM) are also conducted in the index-matched configuration by applying periodic boundary conditions. The simulated electric field (Eₕ, i.e., the component perpendicular to the sample surface - see Fig. 2(c) upper row) plus polarization arrows indicate the charge distributions for the trimer. The electric field data are taken from a plane that is located 1 nm above the disks (z = 41 nm), with the polarization distribution calculated within the middle plane (z = 20 nm) of the nanodisks. In addition, the spatial distribution of the magnetic field (|H| at z = 50 nm) is presented to help one to understand the formed trimer modes in this lattice arrangement. The experimental transmission T is then obtained by the power ratio of the two spectra. Please note that in this report, we present extinction spectra obtained using (1 – T).

Figure 2. (a) Measured extinction spectrum and (b) corresponding numerical simulation for a trimer array with selected near-field patterns shown in (c) (array pitch Λ: 1100 nm, SEM in Fig. 1(b)). Polarization directions (arrows) and distribution of the out-of-plane electric field component Eₕ (colour indicates the charge distribution) of one trimer unit cell at a fixed point of time are shown in the upper row of (c) together with the normalized magnetic field distributions in the lower row. The number labels correspond to the wavelengths highlighted in (b). Each image shown in (c) corresponds to an area of 1 x 1 μm².
At the sides of the investigated spectral domain (points 1 and 4 in Fig. 2(b)), the related field patterns (Fig. 2(c)) exhibit dipolar excitations of the upper particle (monomer excitation) and the two lower particles (dimer excitation), showing a transition from a monomer-like mode to a dimer-like mode (schematically illustrated in Fig. 2(b)).

Between these two cases, two extinction minima (points 2 and 3 in Fig. 2) are found corresponding to different Fano resonance modes. At a wavelength of 1860 nm (point 2), a dipolar excitation of the upper and quadrupolar excitations of the two lower dots are observed, resembling a dolmen-like electric Fano pattern14,16.

It is of great interest to note the loop-like polarization distribution (named as magnetic Fano resonance in Fig. 2(b,c)) situated at the extinction dip close to 1970 nm. In particular, the magnetic field (case 3 in Fig. 2(c)) at this wavelength is highly confined within the inner area of the trimer unit, which cannot be found at any of the other three wavelengths. This pattern has been intensively discussed in the literature and has been identified as a magnetic-based coil-type Fano resonance23–25. Here, the bright mode, formed by the three individual dipole modes being in electric resonance, couples to the magnetic resonance resulting in the circle-like polarization distribution. Therefore, destructive interference of the bright electric mode and the dark magnetic mode results in an extinction dip as confirmed in simulations and experiments. Several reports reveal that this magnetic Fano resonance requires additional symmetry breaking (modifying inter-particle gaps22 or geometry of trimer dots23, etc.) to be excited. This mode is assumed to be forbidden for an individual isolated symmetric trimer or for a sparse array of trimers31,32 since its total electric dipole moment is zero. In addition, its magnetic dipole moment is perpendicular to the sample surface. Therefore, this mode cannot be excited by a plane wave at normal incidence. However, in this closely packed trimer array where the coupling to neighbouring unit cells cannot be neglected, we experimentally observe spectral features for the magnetic Fano resonance modes. In the following sections, we will explore how the coupling between different trimer units can influence the overall optical response of the investigated arrays.

**Trimer plasmon modes in a square-shaped unit cell.** The following section discusses the impact of the intra-trimer gap ($G_1$) and inter-trimer gap ($G_2$) on the spectral behaviour with the trimer located in the centre of a square-shaped unit cell. Here, the diameter (D) and height (H) of the dots is fixed to 425 nm and 40 nm, respectively. For convenience, we address our study to the periodic constant $(\Lambda)$ instead of the inter-trimer gap $G_2$ ($\Lambda = 2 \times D + G_1 + G_3$). In detail, the spectral distribution of the extinction for various values of pitches (1000 nm $< \Lambda < 1700$ nm; steps of 50 nm; $G_3$: 75 nm to 775 nm; $G_1$ is fixed at 75 nm) is presented in Fig. 3(a). Additionally, Fig. 3(b) shows the simulated extinction spectra obtained by modifying the intra-trimer gap $G_1$ from 12.5 to 132.5 nm in steps of 12.5 nm. Both calculations are carried out in the index-matched configuration (incident polarization is along the x-polarized direction) with the white dashed line (Fig. 3(a)) indicating the $<1, 0>$ diffraction order in this configuration.

Overall, the extinction spectra can be dramatically modified by varying the inter- and intra-trimer gaps. In Fig. 3(a), electric and magnetic Fano resonance are observed at pitch values between 1000 and 1400 nm, which are indicated by short-dashed and long-dashed black lines. Within this range of pitches, the trimer-to-trimer
coupling distance $G_2$ is gradually varied while the dot-to-dot interaction remains the same ($G_1$ is fixed). Noticeably, both patterns shift towards longer wavelengths, which indicates that the modal interactions can be modified by the lattice-induced coupling. Once the pitch exceeds 1400 nm, the extinctions only exhibit step-like lineshapes along the onset of the first diffraction order ($\lambda = n$ $\times$ $\Lambda$) in the medium ($n = 1.44$), whereas the distributions referring to electric and magnetic Fano resonance can no longer be observed. More sophisticated lattice modes in the left corner are greyed out since they are beyond the discussion of this article.

Now we consider a complex coupling configuration where the intra-trimer gap $G_1$ and inter-trimer gap $G_2$ are both modified. Figure 3(b) displays simulated extinction spectra that are obtained by varying the intra-trimer gaps $G_1$ (from 12.5 nm to 137.5 nm) for a fixed pitch ($\Lambda = 1100$ nm). In this case, the gap size $G_2$ is correspondingly changed from 237.5 to 112.5 nm. Surprisingly, the central spectral position of electric Fano resonance remains at approximately 1850 nm, while magnetic Fano resonance reduces in wavelength from approximately 2100 to 1970 nm. This blue-shift of the magnetic Fano resonance with increasing gap separation agrees well with the discussion in ref.21.

We need to point out that the emergence of the magnetic Fano resonance mode strongly relies on the near-field coupling of neighbouring unit cells. In Fig. 3(a), the spectral feature of the Fano resonance is most evident at a pitch of 1000 nm, where the inter-trimer gap distance reaches the minimum value investigated here ($G_2 = 75$ nm, $G_1$ is 75 nm). This amplitude enhancement for the Fano resonance mode is also visible in Fig. 3(b) when decreasing $G_2$. In particular, at the minimum value of $G_2$ (Fig. 3(b)), the array cannot be divided into individual trimer unit cells since the inter-trimer coupling of the lower two dots is stronger than the intra-trimer interactions, i.e., $G_2$ is smaller than $G_1$ ($G_2$ 112.5 nm and $G_1$ 137.5 nm).

**Trimer behaviour for the case when the coupling is varied along the horizontal or vertical coupling direction.** In this section, we study the impact of the inter-unit cell coupling on the overall extinction spectra by altering the unit cell along the horizontal (x-axis) and vertical (y-axis) directions. For the following
numerical simulations, arrays of trimers with a dot diameter of $D = 425$ nm, height of $H = 40$ nm and intra-trimer gap of $G_1 = 75$ nm are numerically investigated assuming a homogenous RI environment. Using a periodic boundary condition, the corresponding simulation results are presented in Figs 4 and 5. Here, the horizontal and vertical edges of the rectangular unit cell are named in the following as $\Lambda_x$ and $\Lambda_y$, respectively.

We first examine the trimer spectra and mode profiles by merely extending $\Lambda_x$ from 1000 to 1600 nm ($G_2$: 75 to 675 nm) while keeping the vertical pitch constant ($\Lambda_y = 1100$ nm). The simulations are presented in Fig. 4 with a sketch used to show how the unit cell is modified (left upper corner in Fig. 4 (a)). Two distinct modes dominate the overall extinction response, namely, the monomer-like mode (extinction minima, blue-dashed line) and the dimer-like mode (extinction maxima, orange-dashed line), which both roughly remain at the same spectral position when $\Lambda_x > 1200$ nm (monomer mode: at approximately 1900 nm; dimer mode: close to 1950 nm). For such large horizontal pitches, the inter-trimer gaps $G_2$ are larger than 275 nm, being nearly four times the size of the intra-trimer gap $G_1$ (75 nm), with the result that the inter-trimer coupling is smaller than the inter-dot coupling. Hence, the trimers behave like individual elements and the interplay between the monomer mode and dimer mode is less pronounced.

As highlighted by the blue dashed line in Fig. 4(a), a transition from a monomer-like mode (cases 7 to 3) to an electric Fano mode (case 2 and 1) is clearly observable. In particular, for $\Lambda_x = 1000$ nm, the inter-trimer coupling along the x-axis is maximal, with both electric and magnetic Fano modes clearly visible in the corresponding spectrum. Here, the magnetic Fano resonance mode becomes most prominent, i.e., the respective extinction minimum is most distinct among all the spectra presented.

The next step in the analysis targets the understanding of the impact of the vertical coupling on the optical response. Here, the extinction spectra for the trimer arrays in a rectangle cell with variation along the y-direction are calculated ($G_1$ and $G_2$ are fixed to 75 nm and 175 nm, respectively, with $\Lambda_y = 1100$ nm). Through varying $\Lambda_y$, we obtain a set of extinction spectra that are shown in Fig. 5(a), which are accompanied by corresponding near-field distributions (Fig. 5(b)). Here, the monomer response becomes more complicated and highly asymmetric due to enhanced coupling between the trimers at the onset of the first diffraction order. Nevertheless, a dimer-like pattern (orange dashed line) can be observed for all six cases, while the corresponding extinction maximum shifts...
towards longer wavelengths when increasing \( \Lambda_y \). For cases 2 to 5 (pitch increases from 1100 to 1500 nm), the magnetic Fano resonance is formed, which is evidenced by the respective extinction minimum (red dashed curve in Fig. 5(a)). Inspection of the corresponding near field patterns (red frame) shows that for the case when \( \Lambda_y \) is increased, the amplitude of the excitation of the lower two dots is reduced while the top dot shows a gradually stronger response. Once \( \Lambda_y > 1500 \text{ nm} \), the magnetic Fano resonance cannot be observed anymore.

At this stage, we would like to conclude the discussion of the impact of the near-field coupling on the formation of the magnetic Fano resonance obtained from the simulated results presented in Figs 3, 4 and 5. First, the spatial distribution of the trimers within the array matters. We clearly observe that in the experiments and in simulations, a magnetic Fano resonance mode is formed in densely packed arrays of symmetrical trimers, which should be forbidden for an isolated symmetric trimer based on group theory\(^22,32,34\). Here, we attribute the formation of the magnetic Fano resonance to the symmetry breaking imposed by the square lattice, i.e., to the fact that the trimers and the array have different symmetries. Specifically, the square-shaped lattice breaks the three-fold symmetry of individual trimers, enabling the magnetic and electric modes of the trimer to interact. Second, symmetry breaking-mediated formation of the magnetic Fano resonance only works under particular conditions, i.e., only for certain combinations of lattice parameters, wavelength and incident light polarization. For instance, according to the square-shaped unit cell studied in Fig. 3(a), the pitch should not exceed 1400 nm to trigger the formation of the magnetic Fano resonance. For this configuration, the impact of diffraction can be suppressed below the actual operation wavelength while the inter-unit cell coupling remains effective. Finally, we would like to emphasize again that the trimer-to-trimer coupling plays a more important role for the formation of the magnetic Fano resonance compared to the dot-to-dot interaction within a trimer unit. This phenomenon can be found in Figs 3(b) and 4(a), where a complete loop-like polarization is gradually formed when decreasing the inter-trimer gap \( G_2 \).

**Trimer plasmon modes in various RI environment.** Detecting small changes of the refractive index in nanoscale environments is of key importance for numerous fields particularly within the fields of bioanalytics and molecular sensing, representing a topic that is intensively addressed by the nanophotonics community\(^35–39\). To understand the impact of the RI environment on the optical response of the trimer array and to determine the related RI sensitivity, we immersed the samples into different RI liquids (\( n = 1.31 \))...
to 1.73) and measured the extinction under normal incidence (Fig. 6). In general, the resonance dip, being located near 1650 nm (electric Fano resonance in Fig. 2) in the situation of an air superstrate, shifts towards longer wavelengths as the RI of the liquids increases (this spectral red-shift is highlighted in Fig. 6(a) by the dashed magenta line). The magnetic Fano resonance mode is only found in three of the five extinction spectra (n = 1.31, 1.44 and 1.55). The corresponding wavelengths for the resonances are plotted in Fig. 6(b) and are fitted by linear functions to obtain the RI sensitivity. This procedure yields an RI sensitivity of 312.8 nm/RIU (coefficient of determination $R^2 = 96.9\%$) for the electric Fano-like mode and 330.5 nm/RIU (coefficient of determination $R^2 = 98.8\%$) for the magnetic Fano resonance mode. Compared to sensitivities obtained in other types of plasmonic structures (nanodisks (~180 nm/RIU in VIS$^{40}$), lattices of trimers (~170 nm/RIU in VIS$^{41}$, ~370 nm/RIU in IR$^{45}$), our densely packed trimmer arrays offer sensitivities of the same order, which are comparable to other Fano resonance sensors (approximately 300 nm/RIU in nanohole quadrumers$^{42}$ and heptamer cluster$^{45}$). One advantage of our structure is that the area covered by the array can be minimized to several $\mu$m$^2$ due to the dense configuration. As reported in ref.$^{33}$, we designed a trimer-based fibre sensor, where the diameter of the sensing area is approximately 100 $\mu$m enabling one to perform sensing with the light from the fibre core only, which would otherwise be difficult if the array was very sparse.

It is important to note that the magnetic Fano resonance mode is more pronounced for the case when the particle array is immersed in the liquid with an RI equal to that of the substrate. In this case, a unique relationship between the scattered field and the pitch is obtained because the wavenumber of the scattered wave is the same as that in the substrate and in the superstrate. For the RI-mismatched situation (the bottom and top case in Fig. 6(a)), the two different RIs impose two scattered waves at the same vacuum wavelength (schematically illustrated in Fig. 6(c)). Hence, the distribution of the scattered energy into two waves with different wavenumbers leads to weaker spectral features rather than the strong feature realized in the case of a homogeneous environment.

**Conclusion**

Artificial nanostructures have appreciably promoted the development of nanophotonics research in the last decade, with a multitude of novel optical effects discovered with one of the most important examples being the Fano resonance. In this report, we have studied the impact of the lattice geometry on the optical response of nanotrimmers arranged in densely packed square arrays at infrared wavelengths. We found that in experiments as well as in simulations, electric and magnetic Fano resonance modes can be formed in cases when the inter-trimmer distances are sufficiently small, i.e., arise solely due to the array arrangement. In particular, the observation that magnetic Fano resonances can be excited in a system that consists of a symmetric nanostructure unit supporting a magnetic Fano resonance as a dark mode only clearly shows that the lattice arrangement imposes a symmetry breaking, making the magnetic Fano resonance visible in the spectral response of the array. By varying the array parameters, we clearly reveal a transition from an isolated trimer mode into a regime of strong near-field coupling, showing magnetic Fano resonances with loop-type magnetic field distributions. This otherwise hard-to-observe excitation only emerges due to the presence of the neighbouring trimer units and the symmetry mismatch between the square lattice and three-fold symmetry of the nanotrimmers. Finally, we reveal the impact of different refractive index environments on the optical response of the trimer arrays, with the sensitivity measured to be approximately 300 nm/RIU.

By transferring the concept of lattice-induced symmetry breaking to other nanostructures, our findings provide a pathway towards observing novel kinds of optical excitations and yield a new degree of freedom to design artificial nanocomponents and -devices. Promising applications for the trimer lattice can be envisioned within fields such as nonlinear optics to generate frequencies at desired wavelengths or bioanalytics via high precision sensing$^{44}$. Both examples exploit the unique features of the strongly enhanced electric and magnetic fields generated within the centre of the structure.

**References**

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Author Contributions

N.W. and M.A.S. conceived the idea. N.W. conducted the experimental measurements. N.W. and M.Z. performed the numerical simulations. N.W., M.Z., V.G. and M.A.S. analyzed the data and discussed the results. U.H. fabricated the trimer sample. M.A.S supervised the research. The manuscript was written by N.W., M.Z. and M.A.S. through contributions of all authors. All authors have given approval to the final version of the manuscript.

Additional Information

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