Supplementary Materials for

Plasmonic bound states in the continuum to tailor light-matter coupling

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Sci. Adv. 8, eadd4816 (2022)
DOI: 10.1126/sciadv.add4816

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SUPPLEMENTARY NOTES

Note 1: Strong influence of the nanofin tip on the maximal surface field enhancement. The maximal surface field enhancement given in Fig. 2e is for a smooth spherical nanofin cap with \( d = 700\text{nm} \). Reducing the surface of the cap in any way will drastically enhance the maximal surface field enhancement. To illustrate this, we simulated PNMs with elongated caps (top sphere scaled in z-direction by a factor of 2), with \( d = 500\text{nm} \), and with conical tips, yielding maximal surface field enhancements of 1380, 1690, and 182000, respectively. Thus, our maximal surface field enhancement values as well as most values claimed by others, especially in the IR, need to be taken with a grain of salt.

Note 2: Derivation of the temporal coupled-mode theory (TCMT) model. In the following, we adapt the generally formulated TCMT of optical resonators\(^4\)\(^7\). The dynamic equations of a coupled resonator system can be written as:

\[
\frac{d\vec{a}}{dt} = (i\Omega \mathbf{I} - \Gamma)\vec{a} + \mathbf{K}^\dagger |s_+ \rangle \\
|s_- \rangle = \mathbf{C}|s_+ \rangle + \mathbf{D}\vec{a} 
\]

with \( \vec{a} \) as the amplitude vector of the modes and \( \Omega \) containing the eigenfrequencies and mode-to-mode coupling. \( \Gamma \) describes the energy decay, \( \mathbf{K} \) the port-to-mode coupling, \( \mathbf{C} \) the port-to-port coupling, and \( \mathbf{D} \) the mode-to-port coupling while \( |s_+ \rangle \) and \( |s_- \rangle \) represent incoming and outgoing waves.

For our system, we consider two modes: the plasmon (mode 1) and the vibrational absorption (mode 2) of the analyte. While mode 1 is coupled to one radiation and one intrinsic channel (bright), mode 2 is only connected to an intrinsic channel (dark). Considering intrinsic channels as ports with only outgoing energy results in

\[
\vec{a} = \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}, \Omega = \begin{pmatrix} \omega_1 & \mu \\ \mu & \omega_2 \end{pmatrix}, \Gamma = \begin{pmatrix} y_{11} & 0 \\ 0 & y_{22} \end{pmatrix}, \mathbf{D} = \begin{pmatrix} d_{11} & 0 \\ d_{21} & 0 \\ 0 & d_{32} \end{pmatrix}, \text{ and } |s_+ \rangle = \begin{pmatrix} s_{1+} \\ 0 \\ 0 \end{pmatrix}.
\]

Energy conversion and time reversal symmetry yields the following three relations\(^4\)\(^7\)

\[
\mathbf{D}^\dagger \mathbf{D} = 2\Gamma, \mathbf{K} = \mathbf{D}, \mathbf{C}\mathbf{D}^* = -\mathbf{D}, \text{ which further lead to}
\]
\[ \Gamma = \frac{1}{2} D^\dagger D = \frac{1}{2} \begin{pmatrix} d_{11}^2 + d_{21}^2 & 0 \\ 0 & d_{32}^2 \end{pmatrix}, \quad K^\dagger = \begin{pmatrix} k_{11} & 0 \\ k_{21} & 0 \\ 0 & k_{32} \end{pmatrix} = \begin{pmatrix} d_{11} & 0 \\ d_{21} & 0 \\ 0 & d_{32} \end{pmatrix}, \quad C = - \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \]

and \[ |s_-\rangle = \begin{pmatrix} s_{1+} \\ 0 \\ 0 \end{pmatrix} + D\bar{a}. \]

The energy decay for mode 1 consists of two parts: \( \frac{1}{2} d_{11}^2 \) and \( \frac{1}{2} d_{21}^2 \) representing radiation and intrinsic loss, respectively, so we define \( \gamma_{\text{rad1}} = \frac{1}{2} d_{11}^2 \) and \( \gamma_{\text{int1}} = \frac{1}{2} d_{21}^2 \) (for mode 2: \( \gamma_{22} = \gamma_{\text{int2}} = \frac{1}{2} d_{32}^2 \)), which yields the dynamic equations

\[
\begin{align*}
  i\omega a_1 &= \left( i\omega - \frac{1}{2}(d_{11}^2 + d_{21}^2) \right) a_1 + i\mu a_2 + d_{11}s_{1+} \\
  i\omega a_2 &= \left( i\omega - \frac{1}{2}d_{32}^2 \right) a_2 + i\mu a_1. 
\end{align*}
\]

Substituting \( a_2 \) in (S3) using (S4) and setting \( \omega = \omega_1 = \omega_2 \) results in

\[
0 = -\frac{1}{2}\left( d_{11}^2 + d_{21}^2 + \frac{2\mu^2}{d_{32}^2} \right) + d_{11}s_{1+} = -\left( \gamma_{\text{rad1}} + \gamma_{\text{int1}} + \frac{\mu^2}{\gamma_{\text{int2}}} \right) + d_{11}s_{1+} \text{ and}
\]

\[
|s_+\rangle = \begin{pmatrix} s_{1+} + d_{11}a_1 \\ d_{21}a_1 \\ d_{32}a_2 \end{pmatrix} = \begin{pmatrix} s_{1+} + \sqrt{2}\gamma_{\text{rad1}}a_1 \\ \sqrt{2}\gamma_{\text{int1}}a_1 \\ \sqrt{2}\gamma_{\text{int2}}a_2 \end{pmatrix}. 
\]

Now we can use equation (S1) and (S2) to find a solution for the reflectance \( R \) and thus a solution for the absorbance \( A \):

\[
A = 1 - R = 1 - \frac{|s_-|^2}{|s_+|^2} = 1 - \frac{(s_{1+} - d_{11}a_1)^2}{s_{1+}} = \frac{4\gamma_{\text{rad1}}(\gamma_{\text{int1}} + \frac{\mu^2}{\gamma_{\text{int2}}})}{(\gamma_{\text{rad1}} + \gamma_{\text{int1}} + \frac{\mu^2}{\gamma_{\text{int2}}})^2}. \tag{S5}
\]

For a single resonator without analyte, the absorbance simplifies to

\[
A = \frac{4\gamma_{\text{rad1}}\gamma_{\text{int1}}}{(\gamma_{\text{rad1}} + \gamma_{\text{int1}})^2}. \tag{S6}
\]

**Note 3: Adapting the simulation parameters to the fabricated structures.** When comparing the cylindrical shapes of the simulated PNM with the fabricated ones, two differences are apparent which need to be accounted for. The first one is the not perfect cylindrical shape, which is a result of the laser writing process where each nanofin layer...
is printed by multiple lines of equal length, which results in a slightly (in-plane) rectangular shape. The second deviation comes from an $\alpha$-dependent exposure time to the laser. Since all nanofins are printed using the same parameters for power and speed, nanofins with larger $\alpha$ receive more laser intensity (larger in-plane rectangles) and thus their diameter and top width is increased compared to those with a smaller $\alpha$. We compensate these effects by measuring the diameter $d$ and the top width in x-direction $d_x$ for $0^\circ$ and $26^\circ$ samples and interpolate the values in between with a linear function and smoothing the triangle edges with a $d_x$-dependent radius $r$.

Table S1 lists the used parameters while Fig.S5 sketches the simulated model:

**Note 4: Higher-order BICs.** It is known that the metasurface unit cell must be subwavelength to prevent higher-order diffraction effects, so the resonance wavelength $\lambda_{\text{res}}$ should satisfy: $\lambda_{\text{res}} > n_s \cdot \Lambda$, where $n_s$ and $\Lambda$ are the substrate refractive index and pitch, respectively. It turned out to be extremely challenging for conventional in-plane symmetry-broken geometries to access to higher-order BICs, as finding resonators supporting in-plane multipole moments in a subwavelength scale is difficult. To date, only lower-order BIC modes ($1^{\text{st}}$ and $2^{\text{nd}}$ orders) have been realized from high refractive index resonators\textsuperscript{48}. On the other hand, for our out-of-plane symmetry-broken PNM, the dipole moments do not lie within the metasurface plane. We show that $3^{\text{rd}}$ and $4^{\text{th}}$ order BICs can be achieved for PNMs with $\Lambda = 6\mu m$ and aspect ratios of 1.33 and 2, respectively (Fig.S6a,b). All the BIC modes feature an absorbance above 0.5 and Q-factors of up to 84. The simultaneous access to multiple BIC modes based on our PNM platform opens the possibility to develop multifunctional photonic devices including multicomponent sensors\textsuperscript{49}.

**Supplementary Note 5: Spectral resolution of a PNM sensor**

“Even though our PNM features different Q-factors across different coupling regimes, here we use an averaged Q-factor of 50. Based on the centre wavelength of 1375 cm$^{-1}$ of our used commercial mid-infrared microscope (with a bandwidth from 950 to 1800
cm\(^{-1}\), see Methods), we can obtain a full width at half maximum (FWHM) of 27.5 cm\(^{-1}\) for the resonance. To measure a molecular fingerprint, it is important to separate the neighbouring peaks of vibrational absorption lines. As such, the FWHM must be equal or smaller than the smallest peak distance. In Table S2, we listed some of the most common molecules used for sensing, including PMMA, Streptavidin, polymers (Polyacetal and foam rubber), polycarbohydrates (cellulose and starch), and biological mixtures (blood and semen). All listed molecules were evaluated based on their number of peaks as well as average and smallest peak distances. The overall smallest peak distance we found is for Polyacetal with 37 cm\(^{-1}\), while for other examples it is above 50 cm\(^{-1}\). Given the fact that our reported metasurface platform features a FWHM of 27.5 cm\(^{-1}\), it can resolve all the listed molecules and their corresponding peaks. Therefore, our PNMs linewidth is more than adequate to spectrally resolve multiple absorption lines, and the full molecular fingerprint can be well reproduced.

**Supplementary Note 6: Influence of higher diffraction orders**

Our PNM has a subwavelength pitch and even for incident angles below 30° the BIC resonance wavelength \(\lambda_{\text{res}}\) remains above the wavelength of the first diffraction order, i.e., \(\lambda_{\text{1st order}} = p (1 + |\sin \theta|) < \lambda_{\text{res}}\), with the pitch \(p\) and the incident angle \(\theta\). Thus, only 0\(^{th}\) order of reflection is possible. This is crucial for the existence of our BIC modes since it requires to limit the number of loss channels to one (0\(^{th}\) order reflection) and suppress higher-order diffraction. Otherwise, the resonance energy will be lost quickly via far-field radiation. Thus, we can directly calculate the absorbance via \(A = 1 - R\) (absorbance \(A\), reflectance \(R\)) from the measured spectra normalized to a gold mirror. In order to verify this, the simulation settings with two ports are sketched in Fig. S10a, b showing the reflectance from our PNM at different incident angles \(\theta\) for the fundamental and higher reflectance orders. It is apparent that the higher orders are negligible up to 30°. This is because the BIC mode is redshifted compared to the first order refraction up to around \(\theta = 30°\), see Fig. S10c.
Supplementary Tables

Table S1: Geometrical parameters used in Fig.3. To account for proximity effects, the size for the of diameter $d$, the top diameter $d_x$ and the radius $r$, their value was scaled with $\alpha$.

| | Value at 0° | Value at 26° | Linear interpolation |
|---|---|---|---|
| $d$ [nm] | 635 | 780 | $635 + 5.58 \cdot \alpha$ |
| $d_x$ [nm] | 410 | 600 | $410 + 7.31 \cdot \alpha$ |
| $r$ [nm] | $\frac{1}{2} d_x - 20 = 185$ | $\frac{1}{2} d_x - 20 = 280$ | $185 + 3.65 \cdot \alpha$ |

Table S2: RI sensitivies of comparable systems. Comparison of the refractive index sensing capabilities of commonly used metallic and dielectric nanoparticles (ref. 50 and 51) and metasurfaces (ref. 48, 52, 53, 54, and 55). Values were either calculated from or directly taken from the references.

| Resonator | Material type | Resonance type | nm/RIU | nm/RIU $/\lambda_{res}$ | Q-factor | FOM | Ref. |
|---|---|---|---|---|---|---|---|
| Nanosphere | Metallic | LSPR | 44 | 0.08 | 7.2 | 0.6 | Ref. 50 |
| Nanorod | Metallic | LSPR | 540 | 0.49 | 9.1 | 4.5 | Ref. 50 |
| Nanopyramid | Metallic | LSPR | 195 | 0.29 | 8.7 | 2.6 | Ref. 50 |
| Double layer grating | Metallic | Wood’s anomaly and LSPR | 525 | 0.81 | 47.1 | 38 | Ref. 52 |
| Structure                  | Material | Interaction Type                  | λ(min) | δ | Δθ | Ref. |
|----------------------------|----------|-----------------------------------|--------|---|----|------|
| Mushroom-pillars           | Metallic | Wood’s anomaly and LSPR           | 1026   | 0.80 | 135 | 108  | Ref. 53 |
| Nanodiscs                  | Dielectric | Mie resonance                     | 227    | 0.27 | 20  | 5.4  | Ref. 51 |
| Dielectric metasurface     | Dielectric | Electromagnetically induced transparency | 289    | 0.21 | 480 | 103  | Ref. 54 |
| Randomised nanodiscs       | Dielectric | Mie resonance                     | 86     | <0.10 | <20 | <1.9 | Ref. 55 |
| Crescent metasurface       | Dielectric | SP-BIC                            | 326    | 0.48 | 120 | 58   | Ref. 48 |
| Nanofin                    | Metallic | SP-BIC                            | 6400   | 0.98 | 71  | 70   |
Table S3: Comparison of average and smallest peak distance of vibrational absorption lines. Peaks between 950 and 1800 cm$^{-1}$ of widely used molecules or mixtures were taken into account.

| Molecule     | Number of peaks (cm$^{-1}$) | Average peak distance (cm$^{-1}$) | Smallest peak distance (cm$^{-1}$) | Source   |
|--------------|-----------------------------|-----------------------------------|------------------------------------|----------|
| PMMA         | 4                           | 245                               | 201                                | Ref. 56  |
| Streptavidin | 2                           | 110                               | 100                                | Ref. 57  |
| Ethanol      | 6                           | 100                               | 50                                 | Ref. 58  |
| Polyacetal   | 6                           | 136                               | 37                                 | Ref. 58  |
| Foam rubber  | 6                           | 131                               | 69                                 | Ref. 58  |
| Cellulose    | 6                           | 106                               | 52                                 | Ref. 58  |
| Starch       | 9                           | 81                                | 56                                 | Ref. 58  |
| Blood        | 8                           | 79                                | 62                                 | Ref. 59  |
| Semen        | 5                           | 144                               | 94                                 | Ref. 59  |
**Supplementary Figures**

**Fig. S1: PNM unit cell sketch.** Sketch of a single PNM unit cell, defined by the pitch $\Lambda$, the height $h$, the diameter $d$, the triangle angle $\alpha$, and the gold film thickness $t$.

**Fig. S2: Influence of the film thickness on the BIC resonance.** (a) Reflectance and (b) transmittance spectrum around the BIC mode ($p = h = 3.5\mu m$, $d = 0.7\mu m$, $\alpha = 12^\circ$) for different gold coating thicknesses in 2D color plots. The transmittance quickly drops to zero for thicknesses above $10\text{nm}$ while the BIC lineshape, visible in the reflectance spectrum, becomes narrower and shifts for increasing thicknesses up to $30\text{nm}$. For larger values the mode remains constant so we can assume an optically dense layer of gold for thicknesses larger than $30\text{nm}$.
**Fig.S3:** Dependence of the SLR on the asymmetry. Close-up of the SLR absorbance color plot of Fig.2a for different asymmetries in parameter space featuring a BIC around $\alpha = 18^\circ$.

**Fig.S4:** Dependence of BIC2 on the asymmetry. (a) Maximal absorbance and (b) Q-factors of BIC2 for $\alpha = 0 - 25^\circ$ corresponding to Fig.2a with $p = h = 3.5\mu m$ and $d = 0.7\mu m$. 
Fig. S5: PNM unit cell used in Fig. 3b. Sketch of the PNM unit cell used for simulations in Fig. 3b with the adaptations according to the fabricated structures. (a) Side view (y-z-plane) and (b) top view (x-y-plane) with the widths $d$ and $d_x$ and the smoothing radius $r$. 
**Fig.S6: Higher order BICs.** Absorption spectra for PNMs with $\Lambda = 6\mu\text{m}$ and aspect ratios $AR$ between height $h$ and pitch $\Lambda$ of (a) 1.33 and (b) 2, revealing BIC modes up to the 3$^{rd}$ and 4$^{th}$ order, respectively.
**Fig.S7: RI sensing.** PNM immersed in refractive index liquids with $n = 1.3 - 1.39$ in four steps utilizing the 2nd order BIC resonance, yielding 6400nm/RIU and a FOM of 70. This value is competitive with current sensor designs$^{40,60}$. 
**Fig.S8: Experimental SLR.** SLR of a fabricated PNM with \( p = 6\mu m, h = 7\mu m, d = 0.9\mu m, \) and \( \alpha = 0^\circ \). This mode only appeared this pronounced in a handful of structures, and for most PNMs the SLR was damped almost completely.

**Fig.S9: Amplitude modulation normalized to the Absorbance.** Absolute \((A_{2\text{res}} - A_{1\text{res}})\) and normalized \((\frac{A_{2\text{res}} - A_{1\text{res}}}{A_{1\text{res}}})\) change in absorbance of Fig.4b with \( A_{1\text{res}} \) as the absorbance of the one-resonator system and \( A_{2\text{res}} \) of the two-resonator system.
Fig. S10: Influence of higher refraction orders. (a) Sketch of simulation settings with two ports and excitation form port 1 under the angle $\theta$. (b) Simulated reflectance for incident angles from $0^\circ$ to $40^\circ$ in $5^\circ$ steps for a PNM with parameters adapted from Fig. 4d using $\lambda = h = 3.5 \, \mu m$, $d = 0.7 \, \mu m$, and $\alpha = 12^\circ$. The reflected light of the $0^{th}$ order is plotted with solid lines, and the contributions of higher orders are plotted as dashed lines. For angles up to $30^\circ$ higher orders are negligible and only $0^{th}$ order refraction is taking place. (c) Resonance frequency $\lambda_{res}$ for the reflection parameters of (b) and the first order reflection calculated by $\lambda_{1^{st\, order}} = \rho (1 + |\sin \theta|)$ for an incident angle $\theta$ between $0^\circ$ and $45^\circ$. 
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