Supplementary Information for

Inverse designed plasmonic metasurface with parts per billion optical hydrogen detection

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1. Ultrasensitive Hydrogen Sensors State-of-the-Art

In Table S1 we summarize the available literature on hydrogen sensors with different transduction principles (categorized following the definition given in Ref. 1) that report sensitivity down to the ppb level at room temperature. From the Table, excluding our results here, it is clear that until now, ppb sensitivity is only achieved by electrical (and one thermal conductivity) sensors.

Table S1. Ultrasensitive hydrogen sensors operating at room temperature from the literature.

| Active Materials | Transducer Platform | LoD  | Test Environment | Ref. |
|------------------|---------------------|------|-----------------|------|
| Pd@Ni foam | Electrical | 0.007 | N₂ | ² |
| MWCNT | Electrical | 0.06 | Vacuum | ³ |
| Pd NP on Y₂O₃/CNT | Electrical | 0.09 | N₂ | ⁴ |
| Pd NP@CPPy | Electrical | 0.1 | N₂ | ⁵ |
| Pd nanoflower/graphene | Electrical | 0.1 | N₂ | ⁶ |
| PdCo nanohole | Electrical | 0.18 | Vacuum | ⁷ |
| Pt@PtOₓ nanostructures | Electrical | 0.2 | N.A. | ⁸ |
| Pd NP@PMMA | Optical | 0.25 | Ar and Air | This work |
| Pd-Pt NP on ITPES | Electrical | 0.4 | Ar | ⁹ |
| Chalcogenide NW | Thermal conductivity | 0.4 | Air | ¹⁰ |
| Ru@CPPy | Electrical | 0.5 | N₂ | ¹¹ |
| MoO₃ NR/graphene | Electrical | 0.5 | Air | ¹² |
| MoO₃ NR | Electrical | 0.5 | Air | ¹³ |
| Pd@CNT film | Electrical | 0.89 | N₂ | ¹⁴ |
| Pd hemispherical NP | Optical | 2.5 | N₂ | ¹⁵ |
| PdAuCu NP | Optical | 7 | Vacuum | ¹⁶ |
| PdAu NP@PTFE/PMMA | Optical | 7 | Vacuum | ¹⁷ |
| Pd nanostrip | Optical | 10 | N₂ | ¹⁸ |
| Pd-Al NR | Optical | 40 | N₂ | ¹⁹ |

CNT = carbon nanotubes, CPPy = 3-carboxylated polypyrrole, ITPES = imidazolium-functionalized triptycene polyether sulfone, MWCNT = multi-walled carbon nanotubes, NP = nanoparticles, NR = nanorods, NW = nanowires, PMMA = poly(methyl methacrylate). bThe category of the transduction mechanism follow the ones defined in Ref. 1. cLoD = limit of detection. Only the works that explicitly measured such response at the reported concentration (i.e. not extrapolation) are included here. dN.A. = Information is not available.
2. Scanning Electron Microscopy Images of Pd Square Arrays with Different Diameters and Pitch Arrays

Supplementary Fig. 1. A collage of scanning electron microscopy images of 42 Pd arrays with different diameters and pitch arrays, but with constant height of 45 nm. Note that for imaging purpose, the overlayer PMMA thin film is etched away. Each panel is 24×24 µm².
3. Peak Position and FWHM in Pd Square Array as Function of Pitch Distance and Diameter

Supplementary Fig. 2. (a) Excerpt of simulated extinction spectra of a Pd square array taken from Fig. 1 with different numbers of “peaks” (left: \(a = 300\) nm, middle: \(a = 450\) nm, right: \(a = 450\) nm, all with similar \(d = 180\) nm, \(h = 45\) nm and \(t_{\text{PMMA}} = 200\) nm). In each case, \(\lambda_1\) is consistently assigned to the peak at the longest wavelength, followed by \(\lambda_2\) and \(\lambda_3\) (the shortest wavelength). (b) Peak position of \(\lambda_1\) (left), \(\lambda_2\) (middle) and \(\lambda_3\) (right) as a function of array pitch distance \((h = 45\) nm, \(t_{\text{PMMA}} = 200\) nm). Clear dependence of the peak position on the pitch distance in all peaks is observed. (c) Peak position of \(\lambda_1\) (left), \(\lambda_2\) (middle) and \(\lambda_3\) (right) as a function of nanodisk diameter. In contrast to (b), here only \(\lambda_1\) has dependency on the diameter, whereas \(\lambda_2\) and \(\lambda_3\) are rather constant. This behavior corroborates the dominating contribution of the LSPR in \(\lambda_1\), and of the RA in \(\lambda_2\) and \(\lambda_3\), respectively.
Supplementary Fig. 3. (a) FWHM of $\lambda_1$ (left), $\lambda_2$ (middle) and $\lambda_3$ (right) as a function of array pitch distance ($h = 45$ nm, $t_{\text{PMMA}} = 200$ nm). Clear dependence of the FWHM on pitch distance in $\lambda_1$ is observed, while $\lambda_2$ and $\lambda_3$ exhibit weak dependency. (b) FWHM of $\lambda_1$ (left), $\lambda_2$ (middle) and $\lambda_3$ (right) as a function of nanodisk diameter. Similar to the case in (a), only $\lambda_1$ has dependency on the diameter, whereas $\lambda_2$ and $\lambda_3$ are rather constant. This behavior, along with the dependency of the peak position shown in Supplementary Fig. 2, corroborates the dominating contribution of the LSPR in $\lambda_1$, and of the RA in $\lambda_2$ and $\lambda_3$, respectively.
4. Calculation of Rayleigh Anomalies in Periodic Nanoparticle Array

When light is incident on a 2D square array of subwavelength nanoparticle with pitch distance, $a$, photons can gain additional momentum in integer multiples of

$$|G_x| = |G_y| = 2\pi/a.$$  \hfill (S1)

Rayleigh anomaly (RA) is associated with light diffracted parallel to the lattice surface, and it occurs when

$$k_{out} = |k_0\sin\theta + iG_x + jG_y|.$$  \hfill (S2)

The wavelength of the RA occurs at the onset of the $(i,j)$ diffraction order, above which free-space light diffraction is forbidden in the order. Here, $\theta$ is the incident angle.

In more detail, the different orders of RAs are given by equation:

$$k_{out} = \sqrt{(k_0\sin\theta)^2 + (i^2 + j^2)(2\pi/a)^2 + 2ik_0\sin\theta(2\pi/a)}.$$  \hfill (S3)

Therefore, when the incident light has polarization of $x$ direction, the wavelengths of $(1,0)$ and $(-1,0)$ orders of RAs are calculated by

$$\lambda_{\pm(1,0)} = \pm a(n - \sin\theta),$$  \hfill (S4)

where $n$ is the refractive index of material at diffracted side and assuming the light is coming from air. Similarly, the wavelength of $(0,\pm1)$ can be expressed as

$$\lambda_{(0,\pm1)} = \sqrt{a^2(n^2 - \sin^2\theta)}.$$  \hfill (S5)

For simplicity, the wavelength or wavevector of higher orders of RAs also can be calculated from Eq. S3.
5. Additional Data on the Angle-Dispersion Extinction Spectra of Pd Array

Supplementary Fig. 4. (a) Extinction spectra of a array sample with \( d = 180 \) nm, \( h = 45 \) nm, \( a = 300 \) nm and \( t_{\text{PMMA}} = 200 \) nm. (b) 2D maps of the normalized total field amplitude \( |E|^2 \) of the array at wavelength as marked in panel (a) at the mid-height of the nanoparticles. Dashed lines outline the nanodisks. (c) Experimental and simulated wavelength-resolved angle dispersion extinction spectra of the array with \( d = 180 \) nm, \( h = 45 \) nm, \( t_{\text{PMMA}} = 200 \) nm and \( a = 300 \) nm, showing the different RA orders (dashed lines) and the LSPR position of the corresponding single-particle counterpart.

Supplementary Fig. 5. (a) Extinction spectra of a array sample with \( d = 180 \) nm, \( h = 45 \) nm, \( a = 400 \) nm and \( t_{\text{PMMA}} = 200 \) nm. (b) 2D maps of the normalized total field amplitude \( |E|^2 \) of the array at different excitation wavelengths, as marked in panel (a) at the mid-height of the nanoparticles. Dashed lines outline the nanodisks. (c) Experimental and simulated wavelength-resolved angle dispersion extinction spectra of the array with \( d = 180 \) nm, \( h = 45 \) nm, \( t_{\text{PMMA}} = 200 \) nm and \( a = 400 \) nm, showing the different RA orders (dashed lines) and the LSPR position of the corresponding single-particle counterpart.
6. Sensor Array Parameters Obtained by Particle Swarm Optimization

Table S2. Figure-of-Merit and Sensor Array Parameters Obtained by Particle Swarm Optimization after the 15th Generation.

| Population | FoM | Δλ_{peak} [nm] | FWHM [nm] | d [nm] | h [nm] | a [nm] | t_{pssic} [nm] |
|------------|-----|----------------|-----------|-------|-------|------|---------------|
| 1          | 0.091 | 63.75          | 700       | 300   | 25    | 500  | 161           |
| 2          | 0.079 | 24.5           | 310.75    | 150   | 20    | 400  | 293           |
| 3          | 0.087 | 19.75          | 227.25    | 132   | 22    | 438  | 300           |
| 4          | 0.096 | 21.75          | 226.25    | 120   | 20    | 431  | 300           |
| 5          | 0.082 | 17.25          | 211.5     | 138   | 20    | 443  | 290           |
| 6          | 0.102 | 28             | 274.5     | 121   | 21    | 395  | 297           |
| 7          | 0.095 | 21.5           | 227.25    | 104   | 23    | 396  | 299           |
| 8          | 0.081 | 20.75          | 255.5     | 146   | 20    | 429  | 298           |
| 9          | 0.105 | 31             | 296.75    | 124   | 20    | 376  | 300           |
| 10         | 0.103 | 30.25          | 294.5     | 120   | 20    | 376  | 293           |

Supplementary Fig. 6. Simulated extinction spectra of the calculated Pd and Pd hydride array obtained by PSO after the 15th generation.
7. Angle Dispersion Extinction and Field Distribution of the Optimized Sensor Array

Supplementary Fig. 7. (a) Simulated wavelength-resolved angle dispersion extinction spectra of the optimized sensor array, showing the different RA orders (dashed lines) and the LSPR position of the corresponding single-particle counterpart (Supplementary Fig. 8). (b) Field distribution surrounding the nanoparticle (left) and at the close vicinity and inside of the nanoparticle (right) at three different excitation wavelengths corresponding to the extinction peaks ($\lambda_1-\lambda_3$). From the maps it is clear that relative field amplitude inside the nanoparticle excited at $\lambda_1$ and $\lambda_2$ are lower than the one at $\lambda_3$. This again corroborates the nature of the peak, in which $\lambda_3$ is dominated by the LSPR and thus is sensitive to the change from Pd to Pd hydride. Dashed lines outline the interfaces between glass/nanodisks/PMMA/air.

8. Single Particle Counterpart of the Optimized Sensor Array

Supplementary Fig. 8. Calculated Pd and PdH$_{0.12}$ extinction spectra of a single Pd nanodisk with same geometrical parameters to the optimized array sensor obtained by PSO ($d = 124$ nm, $h = 20$ nm, $a = 376$ nm, $t_{PMMA} = 300$ nm). Comparable peak shift as for the array sensor is found upon hydrogenation but with very broad FWHM. This combination results in a FoM of 0.07.
9. Extended Particle Swarm Optimization Method

If we assume the position of the Pd/PdH₀.₁₂ in the parameter space as a function of four variables, $x_i(n) = f(d(n), h(n), a(n), t_{PMMA}(n))$, where $d(n)$, $h(n)$, $a(n)$ and $t_{PMMA}(n)$ are the particle diameter, height and the pitch distance of the array, $t_{PMMA}$ is the thickness of PMMA, and $n$ is the generation counter. During the optimizing process, the particle is subjected to three forces as it moves through the parameter spaces: (i) a frictional force that is proportional to the velocity, $\alpha v_i(n)$, where $\alpha$ is the inertial weight; (ii) a spring force towards the individual best value of this particle, $c_1 r_1 [\hat{x}_i(n) - x_i(n)]$, where $c_1$ is the cognitive factor and $r_1$ is a random number between 0 and 1; and (iii) a spring force towards the global best value of all the particles, $c_2 r_2 [g(n) - x_i(n)]$, where $c_2$ is the social factor and $r_2$ is a random number between 0 and 1.

The velocity in the next generation can be obtained from the sum of these three forces, i.e., $v_i(n + 1) = \alpha v_i(n) + c_1 r_1 [\hat{x}_i(n) - x_i(n)] + c_2 r_2 [g(n) - x_i(n)]$. In the Lumerical solver, we used the default values of $c_1 = c_2 = 1.49$ and linearly spaced values of $\alpha$ between minimum 0.4 and maximum 0.9 for PSO simulations that have been verified to converge well in many test optimization for photonic design problems. The position of the particle in the next generation is then given by $x_i(n + 1) = x_i(n) + v_i(n + 1)$.

Supplementary Fig. 9. PSO algorithm for one nanoparticle array in the PSO terminology updating during one generation.
Supplementary Fig. 10. Simulated extinction spectra of Pd and PdH$_{0.12}$ for Population 2 after 18$^{th}$ generation of optimization. Clearly the PSO reaches a configuration where the two SLR peaks are close to each other and thus indistinguishable. Due to this condition, when the spectra change from Pd to Pd hydride, the program assigns a different peak, which then results in seemingly large peak shift (and thus falsely large FoM). To avoid this problem, we stopped our PSO simulation at the 15$^{th}$ generation.
10. Optimized Array Sensor: Structural and Optical Characterization

Supplementary Fig. 11. (a) SEM image of the sensor array. (b) Diameter distribution of the particles forming the sensor, showing an average of 130 nm, which is slightly larger than the targeted diameter of 124 nm. (c) Pitch distance distribution of the sensor array with average of 378 nm, which is very close to the targeted 376 nm. (d) Experimental extinction spectra of the fabricated array sensor.

11. Determination of Experimental $\lambda_{\text{peak}}$ and Its Noise

Supplementary Fig. 12. (a) Lorentzian function fitting (red dashed line) to the experimental optical spectra to extract $\lambda_{\text{peak}}$. In our analysis, the fit is only applied within $\pm60$ nm from the peak maximum (grey shaded area, following the method established in Ref. 17), where the peak is symmetric. (b) Zoomed-in version of (a) within the fit range. Clearly, the Lorentzian represents well the data and thus enables a good fit with $R^2 > 0.99$. (c) Lorentzian-fitted $\Delta \lambda_{\text{peak}}$ response of the best sensor (cf. Fig. 4a) in the first 30 min of operation used to derive the peak-to-peak readout noise, $\sigma$, of 0.01 nm. The dashed lines and gray-shaded areas denote the mean of the signal and $\pm \sigma$ from the mean, respectively.
12. Other Performance Aspects: Reproducibility, Speed, and Poisoning Resistance

**Supplementary Fig. 13.** (a) $\Delta \lambda_{\text{peak}}$ response to stepwise random H$_2$ concentration (250 to 0.25 ppm) in Ar carrier gas at room temperature. Inset: zoomed-in version of the sensor response to 250 ppb H$_2$. (b) Measured $\Delta \lambda_{\text{peak}}$ as a function of H$_2$ concentration derived from (a). The transparent symbols and gray dashed line are reproduced from Fig. 4b. The sensor’s responses to these random H$_2$ exposure are consistent with the descending one, and thus exemplifying the reproducibility of the sensor.

**Supplementary Fig. 14.** (a) $\Delta \lambda_{\text{peak}}$ response to three consecutive cycles of 250 ppb H$_2$ (grey areas). A reversible and reproducible sensor response to such low concentration of H$_2$ is observed. (b) Average sensor signal to the three cycles of 250 ppb H$_2$ exposure. An uncertainty of $\sim$0.01 nm is recorded, which is in the same order of the sensor’s signal noise.
To deduce the response and recovery times of the sensors we use the commonly used $t_{90}$ and $t_{10}$, respectively (Supplementary Fig. 15a-b). As plotted in Supplementary Fig. 15c-d, the response and recovery times of the sensor increase with the lowering H$_2$ concentration. Such observation is inherent to Pd nanostructures as previously shown.$^{15,17,20}$ As a result, at the lowest H$_2$ concentration of 250 ppb, the sensor’s response time is in the order of 40 min. Interestingly, the response and recovery times of the control random array sensor are practically similar to the optimized one (Supplementary Fig. 15c-d). Such finding reveals that our method of increasing the sensor’s sensitivity via periodic arrangement does not affect its sensing speed as it is mainly defined by the materials design. Thus, our sensitivity improvement method can be combined with other methods aimed to directly enhance the sensor’s speed, for example by employing nanoparticles with reported faster kinetics than Pd (e.g. PdAu$^{17,21}$ PdCo$^{15}$ and PdTa$^{22}$ alloys, with speed twice as fast compared to pure Pd) or by utilizing polymer coatings with higher kinetics-enhancements such as PTFE (twice as high as PMMA).$^{17}$

Supplementary Fig. 15. The definition of (a) response time as $t_{90}$ and (b) recovery times as $t_{10}$, which correspond to the time it takes to reach 90% and 10% of the normalized signal (with respect to signal during the exposure and in the absence of H$_2$), respectively. (c) Response times and (d) recovery times of the optimized periodic array sensor and control random array sensor as function of H$_2$ concentration. Data is extracted from Fig. 4a and c, respectively. The recovery and response times of both sensors are comparable and can practically be described with a single trend (the dashed lines), as established in ref. $^{17}$. 


Supplementary Fig. 16. (a) Time-resolved $\Delta \lambda_{\text{peak}}$ response of sensor 1 pulse of 1000 ppm $\text{H}_2$ followed by 5 pulses of 1000 ppm $\text{H}_2 + 500$ ppm $\text{CO}$, and 1000 ppm $\text{H}_2 + 50$ ppm $\text{NO}_2$ in Ar. (b) Normalized sensor signal to the one obtained in 1000 ppm $\text{H}_2$. The error bars denote the standard deviation from 5 cycles. The shaded area indicates the $\pm 20\%$ deviation limit from the normalized $\Delta \lambda_{\text{peak}}$ in 1000 ppm $\text{H}_2$.

13. Optical Properties of Tandem Sensors

Supplementary Fig. 17. (a) Experimental refractive indices of PMMA and PVOH. The data of PMMA is reproduced from $^{23}$. (b) Unit cell schematic the tandem sensor ($d = 124$ nm, $h = 20$ nm, $a = 376$ nm), with $t_{\text{PMMA}} = 295$ nm and $t_{\text{PVOH}} = 5$ nm. (b) FDTD-calculated extinction spectra of tandem sensor (see the schematic in Fig. 5b) for Pd (light gray) and PdH$_{0.12}$ (dark grey) nanodisk arrays. The spectra are basically identical to the ones of the sensor coated with 300 nm PMMA (cf. Fig. 3d).
14. Quasi-Random Array Control Sample: Structural, Optical and Noise Characterization

Supplementary Fig. 18. (a) SEM image of the control sensor array with quasi-random particle distribution. (b) Diameter distribution of the particles forming the sensor, showing an average of 122 nm, which is slightly smaller than the targeted diameter of 124 nm. (c) Radial distribution function (RDF) of the control sensor. The primary peak in the RDF (i.e., \( \sim 2.5 \bar{\varnothing} \)) indicates the average center-to-center distance between neighboring nanostructures. (d) Experimental extinction spectra of the quasi-random array.

Supplementary Fig. 19. (a) Lorentzian function fitting (red dashed line) within \( \pm 60 \) nm from the peak maximum (grey shaded area) to extract \( \lambda_{\text{peak}} \). (b) Zoomed-in version of (a) within the fit range. The Lorentzian represents the data well in the peak-maximum region and thus enables a good fit with \( R^2 > 0.97 \). (c) Lorentzian-fitted \( \Delta \lambda_{\text{peak}} \) response of the quasi-random array control sensor (cf. Fig. 4c). The derived peak-to-peak noise, \( \sigma_{\text{control}} \), is 0.08 nm, much higher than that of the optimized regular array sensor. The dashed lines and gray-shaded areas denote the mean of the signal and \( \pm \sigma \) from the mean, respectively.
15. Experimental Details on the Gas Flow Used

Table S3. Set Flow of Hydrogen and Argon Gas to Achieve the Targeted Hydrogen Concentrations.

| Targeted H₂ Concentration [ppm] | 1000 ppm H₂ Flow [mL/min] | Ar Flow [mL/min] |
|----------------------------------|--------------------------|------------------|
| 1000                            | 1                        | 0                |
| 250                             | 5                        | 15               |
| 100                             | 2                        | 18               |
| 46                              | 1                        | 21               |
| 22                              | 1                        | 44               |
| 10                              | 1                        | 99               |
| 5                               | 0.5                      | 199              |
| 2                               | 0.25                     | 125              |
| 1                               | 0.1                      | 100              |
| 0.75                            | 0.1                      | 133              |
| 0.5                             | 0.1                      | 200              |
| 0.25                            | 0.05                     | 200              |
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