Supporting Information

Patterned plasmonic gradient for high-precision biosensing using smartphone reader

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1. Theoretical considerations

1.1 Sensitivity of SPPG sensor

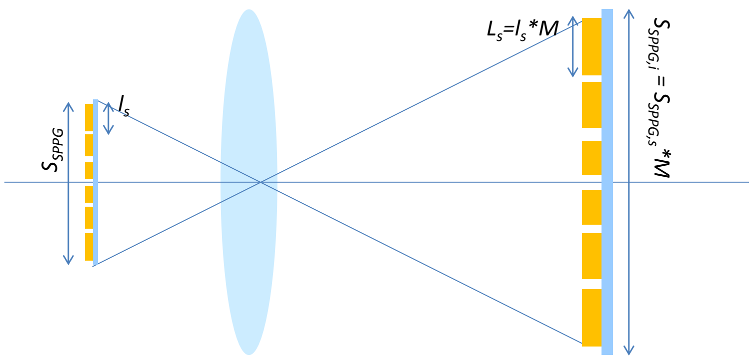

Figure S1. Schematic drawing of the SPPG sensor system.

In this work, the refractive index is determined by measuring the size of the resonance ring, and the sensitivity can therefore be defined as

$$\eta = \Delta S_{res,i} / \Delta n \qquad \text{(S1)}$$

Here, $S_{res,i}$ is the size of the resonance ring at the image plane.

It has been reported that the resonance wavelengths λ_{res} of plasmonic nanorod have an explicit relation with their geometrical parameters [3]

$$\lambda_{res} = \lambda_p \sqrt{\varepsilon_b + aR^2 n^2} \qquad \text{(S2)}$$

Here $R = l/w$ is the ratio between the length *l* and width *w* of the rod, and in the SPPG structure, the rod length is position-dependent. Since the pitch size of the nanorod array is smaller than half of the resonance wavelength, the length of the antenna *l*(*x*, *y*) can be treated as a continuous function:

$$l_{res}(x,y) = l_0 + \frac{2(l_{\max} - l_0)}{S_{SPPG}} \max(|x|,|y|) = l_0 + \frac{2\Delta l}{S_{SPPG}} \max(|x|,|y|) \quad \text{(S3)}$$

Here, l_0 is the rod length at the center point of SPPG, l_{max} is the length of the longest rod at the edge of SPPG, $\Delta l = l_{max} - l_0$, and S_{SPPG} is the size of SPPG sensor.

For most cases, ε_b is small, and after inserting eq. (3) into eq. (2), we have

$$\max(|x|,|y|) = \frac{\lambda_{res} w S_{SPPG}}{2\lambda_p \sqrt{a}\,\Delta l n} - \frac{l_0 S_{SPPG}}{2\Delta l} \quad \text{(S4)}$$

The size of the resonance rings in the object plane:

$$S_{res,s} = 2\max(|x|,|y|) = \frac{\lambda_{res} w S_{SPPG}}{\lambda_p \sqrt{a}\,\Delta l n} - \frac{l_0 S_{SPPG}}{\Delta l} \quad \text{(S5)}$$

After the imaging system, the size of the resonance ring is magnified by *M* folds (Figure S1),

$$S_{res,i} = S_{res,s} M$$

We have

$$S_{res,i} = 2\max(|x|,|y|) = \left(\frac{\lambda_{res} w S_{SPPG} M}{\lambda_p \sqrt{a}\,\Delta l}\right)\left(\frac{1}{n}\right) - \frac{l_0 S_{SPPG} M}{\Delta l}$$

Then the sensitivity becomes:

$$\eta = \frac{\Delta S_{res,i}}{\Delta n} = \frac{\lambda_{res} w S_{SPPG}}{\lambda_p \sqrt{a}\,\Delta l n^2} M \quad \text{(S6)}$$

In order to remove the errors induced by the optical characterization system, we use the relative size of the resonance ring S_{res}/S_{SPPG} instead of the absolute value of S_{res} in the measurement. The sensitivity can then be written as:

$$\eta = \frac{\Delta\left(S_{res,i}/S_{SPPG}\right)}{\Delta n} = \frac{\lambda_{res} w}{\lambda_p \sqrt{a} \Delta l n^2} M \qquad \text{(S7)}$$

1.2 Error analysis:

The limit of detection (LoD) is determined by how accurate the size of the resonance rings can be determined. In this work, the position of the resonance ring is determined by numerical fitting. The uncertainty of the position is

$$\delta S_{res,i} \propto \frac{FWHM_i}{\sqrt{N}} \qquad \text{(S8)}$$

Here, N is the number of counts. Considering that the fitting processes are repeated by N_{fit} times along the whole ring, the accuracy is improved by $N_{fit}^{0.5}$ times:

$$\delta S_{res,i} \propto \frac{FWHM_i}{\sqrt{N N_{fit}}} \qquad \text{(S9)}$$

In experiment, the exposure of each pixel are normally kept at a constant level, Q, a value close to its well-depth, and the total signal can be estimated using $Q_{res,i}/a_{pix}$. Here, a_{pix} is the pixel size of the camera. In addition, $N_{fit}= 4S_{res,i}/a_{pix}$

Then, eq. 9 becomes

$$\delta S_{res,i} \propto \frac{FWHM_i}{\sqrt{\frac{MS_{res,s}}{a_{pix}} Q \frac{4MS_{res,s}}{a_{pix}}}} = \frac{FWHM_i \sqrt{a_{pix}}}{MS_{res,s}\sqrt{Q}} \qquad \text{(S10)}$$

Knowing that $\eta = \frac{\Delta S_{res,i}}{\Delta n} = \frac{\lambda_{res} w S_{SPPG}}{\lambda_p \sqrt{a} \Delta ln^2} M$, we have

$$\delta n = \frac{\delta S_{res,i}}{\eta} \propto \frac{FWHM_i \sqrt{a_{pix}}}{MS_{res,s}\sqrt{Q}} \cdot \frac{\lambda_p \sqrt{a} \Delta ln^2}{M\lambda_{res} w S_{SPPG}} = \frac{FWHM_i \lambda_p \sqrt{a_{pix}} a \Delta ln^2}{M^2 S_{SPPG} S_{res,s} \lambda_{res} w \sqrt{Q}} \qquad \text{(S11)}$$

Since the $S_{res,s}$ is proportional to S_{SPPG}, eq. (11) can be rewritten as:

$$\delta n \propto \frac{\sqrt{a_{pix}}}{M^2 S_{SPPG}{}^2} \qquad \text{(S12)}$$

Therefore, the limit of detection of the SPPG structure can be improved by simply decreasing the pixel size of the sensor, using a larger magnification lens and increasing the size of the sensor.

To illustrate how the parameters influence the performance, we calculated the LoD of different systems using eq. S12 and the experimental result in this work (M = 20, S_{SPPG} = 76 µm, and LoD = 9.1E-4). Table S1 shows the results.

Table S1. LoD as function of S_{SPPG} (µm) and *M*.

S_{SPPG} \ M	5	10	20
76	1.4E-2	3.6E-3	9E-4
150	3.6E-3	9E-4	2.2E-4
300	9E-4	2.2E-4	5.5E-5

Here, it is worth mentioning that $MS_{SPPG} = S_{SPPG,\,i}$, is the size of image the SPPG sensor, and cannot be larger than the size of image sensor. Therefore, the sensitivity is ultimately limited by the size of image sensor (CMOS chip) of the system.

2. The sucrose concentration and refractive index

Table S2. Refractive index of sucrose solution [s1]

wt% [a)]	0	9.090	16.67	23.08	28.57	33.33	37.50	41.18	50.00	60.00
RI [b)]	1.3340	1.3477	1.3573	1.3691	1.3811	1.3872	1.3969	1.4025	1.4186	1.4407

[a)] Weight percentage of sucrose solution; [b)] Refractive index of sucrose solution

[s1]. W. Zhou; S. Luo; C. Wang and X. Zhang, *Journal of Xingyi Normal University for Nationalities* 2013, **5**, 119-121.