Electronic Supplementary Material (ESI) for Nanoscale.

Highly sensitive biosensors based on all-dielectric nanoresonators

Electronic Supplementary Information

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Transmittance Measurement

We measure the linear-optical transmittance spectra of nine identical arrays (fabricated during the same EBL-exposure session with all the same parameters) in three cases, the "bare", "biotin-coated", and "streptavidin-bound" cases. Two sets of example spectra for sample B2 are shown in Figure S1a and b for vertical and horizontal polarization incident

light respectively.

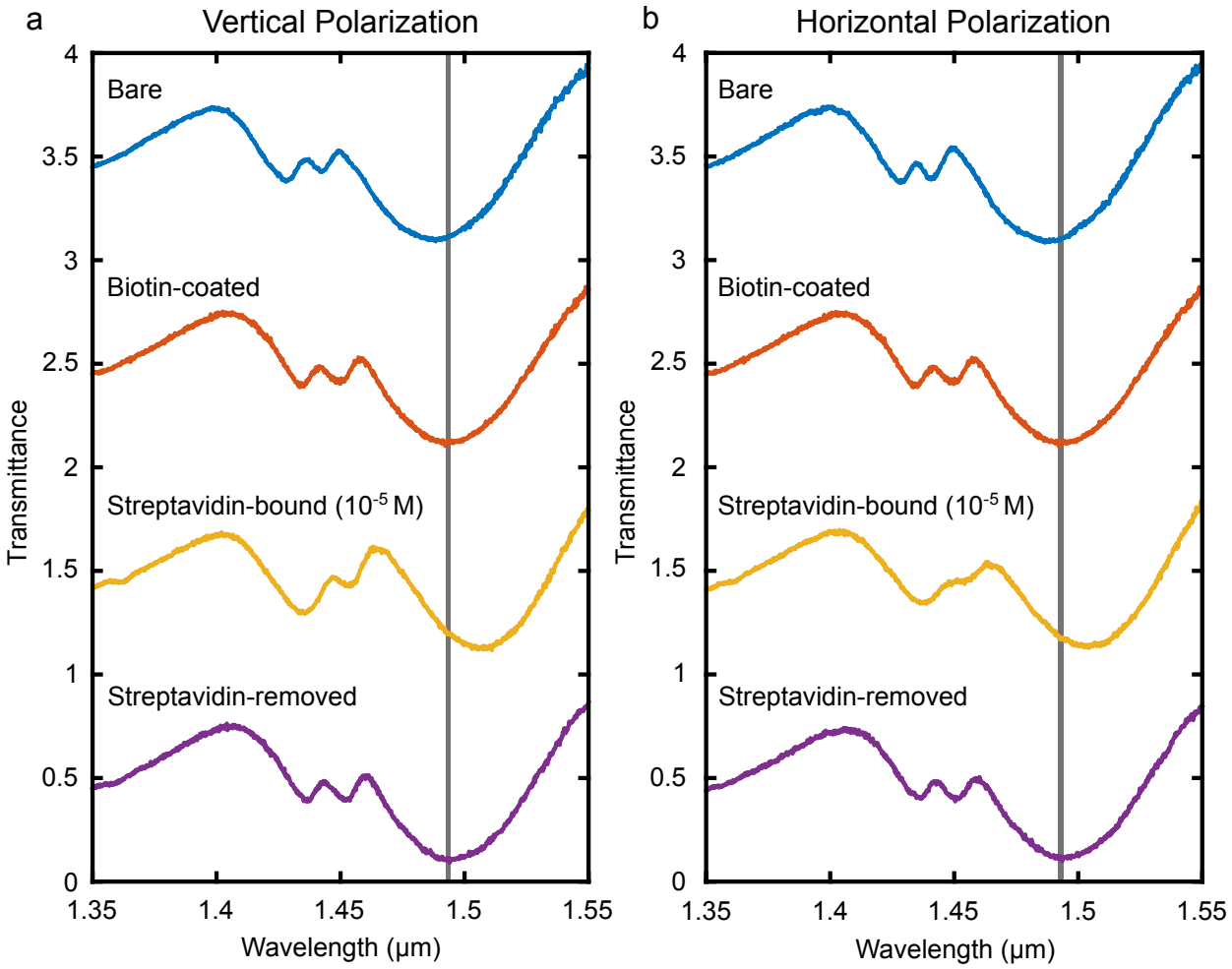

Figure S1: Linear-optical transmittance spectra with (a) vertically and (b) horizontally polarized light for the bare (blue), biotin-coated (orange) and streptavidin-bound (yellow) nanodisk array red shift progressively. After the removal of the streptavidin (purple), the resonance returns to the position prior to the application of streptavidin (the biotin-coated case) as highlighted by the gray line.

In case one ("bare", blue curves), the nanodisks have no linker molecules or streptavidin bound to them.

In case two ("biotin-coated", orange curves), the arrays are measured after functionalization of the Silane-PEG-Biotin molecules as described in Methods of the main text. The spectra obtained here serve as the baseline-measurement for the spectral shift of the streptavidin-bound measurement.

In case three ("streptavidin-bound"), six different concentrations of streptavidin solution

are used to bind with the biotin on all the nine arrays. The transmittance is measured after the streptavidin binding process described in Methods of the main text. The yellow curves shown in Figure S1 depict the spectra for the 10^{-10} M case.

For all three cases, the measurement procedures are as follows:

The incident light is polarized linearly in either vertical or horizontal direction. Furthermore, we measure the transmittance for an unstructured etched region on the wafer as a reference, allowing us to retrieve the transmitted power through the combined system of the silicon disk array and the silica layer.

We note that the effect of the silica layer cannot be fully eliminated by the referencing procedure, as the presence of the nanodisk arrays changes the transmittance properties of the silica layer itself. This leads to an overestimation of the transmittance due to Fabry-Perot oscillations in the silica box layer, hence the presented results have been corrected using the calculated Fabry-Perot spectra using a box thickness of 1990 nm.

At the end of each concentration measurement, the bound-streptavidin is removed and a spectrum is taken again (puple curve) for verification. The resonance returns to the position of the biotin-coated case, indicating the complete removal of the streptavdin. This also reaffirms that our streptavidin removal process is reliable, our experiment is repeatable and results reproducible.

Polarization Sensitivity

From Figure S1 and Figure 4b in the main text, identical resonance dip positions for the "bare" and "biotin-coated" cases are observed between the two polarizations, suggesting the polarization insensitivity of the system, which can be explained by the symmetry along the optical axis of the nanodisks. However, polarization dependence is observed when streptavidin is applied (Figure 5b in the main texts). This is potentially due to a preferred binding orientation of the streptavidin, or a photonic nearfield effect where an asymmetry in the nearfield is only translated to the farfield upon the coupling with the analytes.

Such a photonic nearfield effect could be introduced by fabrication inaccuracies of the nanodisk sample which create slight differences in the nearfield profiles for orthogonal orientations of the excitation polarization. This is consistent with a polarization independent response of the bare structure if the respective modes are accidentally degenerate. However, the binding of the streptavidin may lift this degeneracy if the overlap of the streptavidin layer with the excited nearfields differs for different excitation polarizations, leading to the polarization dependence observed.

Statistical Analysis

To determine the spectral position of the transmission dip for each spectrum, a parabolic function is fitted using MATlab to the spectrum ±10 nm around the position of the dip.

The minima values for all nine arrays are then collected; the mean and standard deviation of each concentration of streptavidin, and the biotin-coated nanodisk are then computed. The spectral shift results for each concentration of streptavidin in Figure 3b is the mean of the nine arrays referenced to the mean of the biotin data. The error bars show the standard deviation of the shift calculated using the computed data, taking into account propagation of error.

Dose-response curve fitting

The dose-response curve is fitted using the Hills equation of the form of Equation S1.

$$y = B + \frac{A - B}{1 + \left(\frac{x}{C}\right)^D} \tag{S1}$$

Here, A represents the maximum asymptote, B the minimum asymptote, C the half maximal effective concentration (EC50), D the Hill coefficient, x the concentration and y the spectral shift value.

The fitting parameters for the two curves are shown below in Table S1.

Table S1: Fitting parameters for dose-response curve in Figure 5b

Polarization	**A**	**B**	**C**	**D**
Horizontal	11	0	1×10^{-9}	0.45
Vertical	13	0	1×10^{-9}	0.43

BSA reference test

Similar to the binding of streptavidin, we place a 10 μl droplet of bovine serum albumin (BSA) (in 10× diluted PBS solution at pH 7.2) with a concentration of 10^{-5} M on the sample after the linker functionalization. After the complete evaporation of the droplet we washed the sample with de-ionized water at room temperature to remove any unbound BSA. Transmittance measurement is taken after the sample is blow-dried with compressed nitrogen gas. Spectral shifts of 0.1 nm and 1 nm are observed for the vertical and horizontal polarization respectively, which can be regarded as no shift within experimental error. Figure S2a and b show the spectra before application of BSA (blue curve) and after (orange curve) for vertical and horizontal polarization respectively.

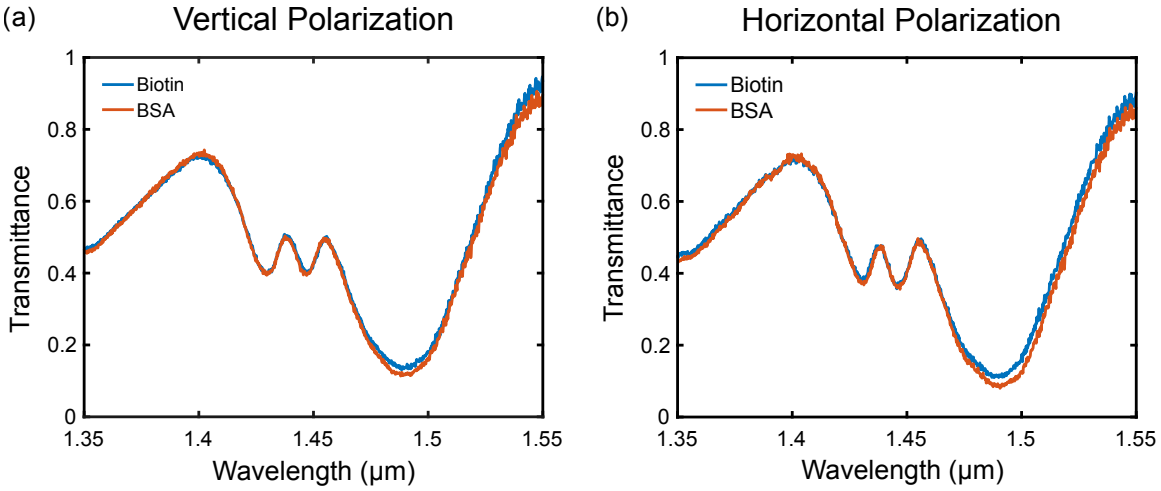

Figure S2: The spectra of the biotin-coated nanodisk arrays before (blue) and after (orange) the application of bovine serum albumin are basically identical for (a) vertical and (b) horizontal polarization, confirming the specificity our experiment.