SUPPLEMENTARY INFORMATION

Computational Sensing Using Low-Cost and Mobile Plasmonic Readers Designed by Machine Learning

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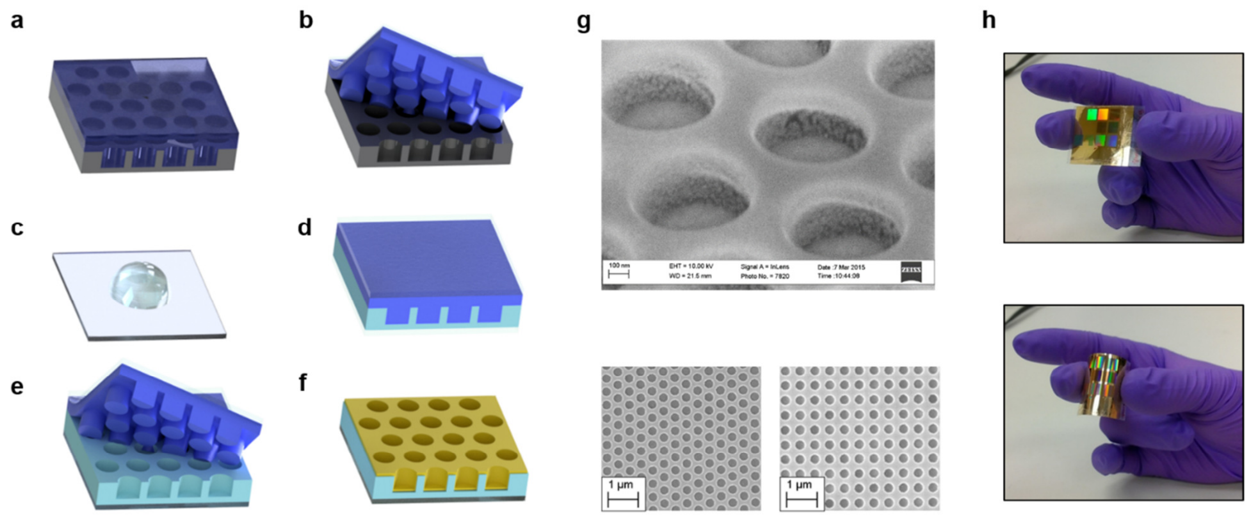
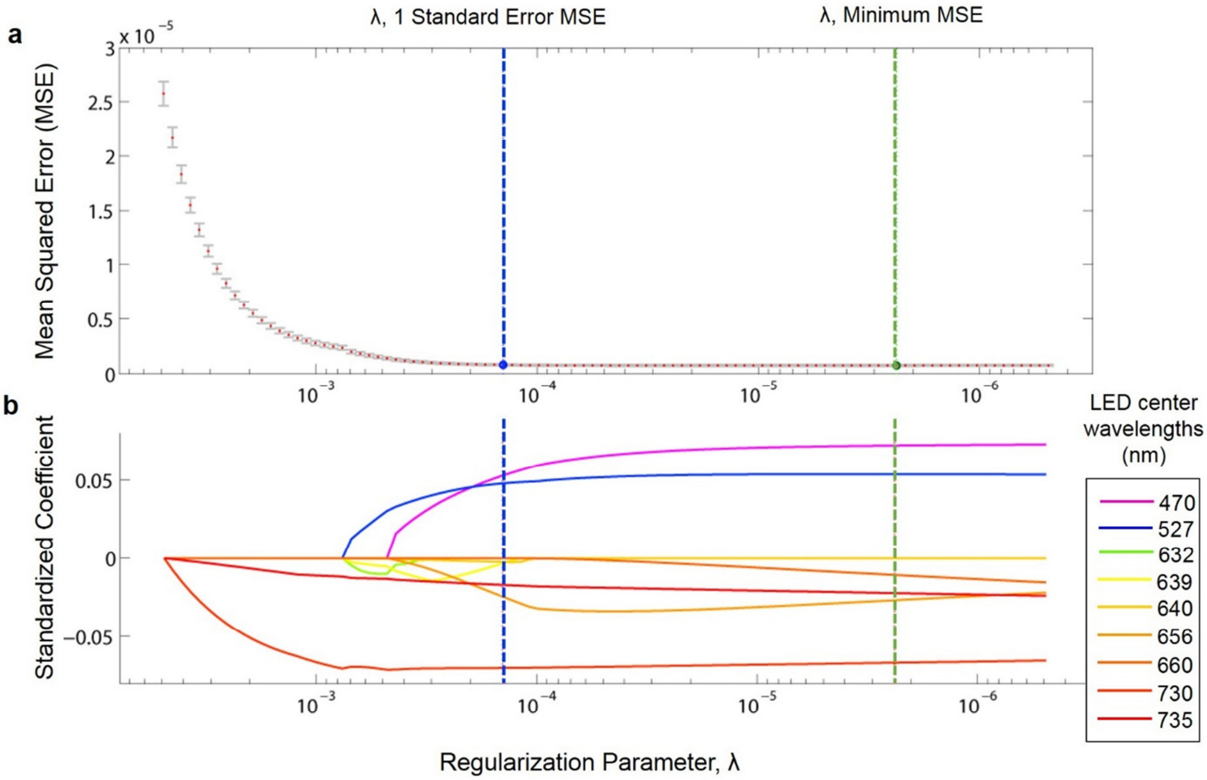
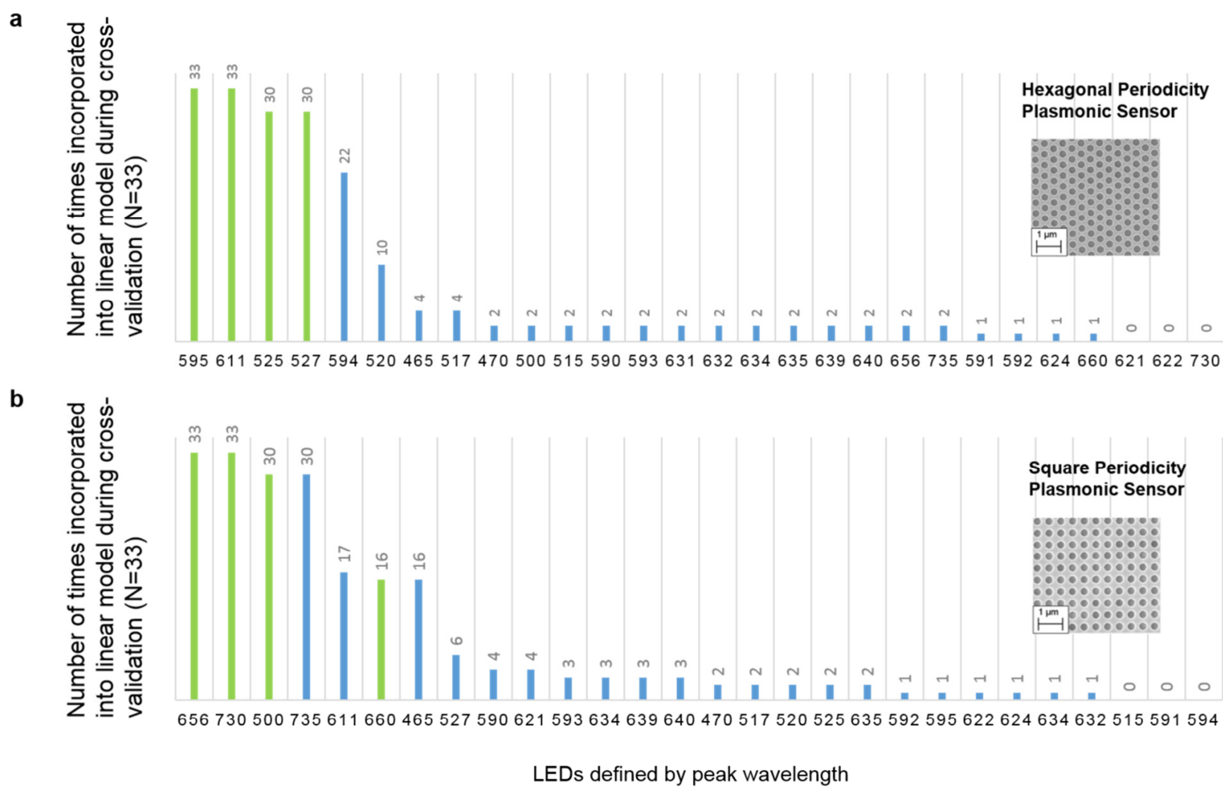
Supplementary Figures



Supplementary Figure 1. The imprint mold fabrication procedure (a)-(f) starting from the one-time produced silicon master which is molded with a polyurethane acrylate (a) and (b). This initial mold is then peeled from master and used in a secondary imprint process (c)-(e) with a UV curable polymer (NOA 81) before the final gold deposition procedure (f). SEM images (g) show the hexagonal and square periodicity nano-hole array structures with the side-wall profile characteristic of the line-of-sight Electron Beam Evaporator deposition. The final plasmonic sensors (h) fabricated on a flexible mylar backing layer.



Supplementary Figure 2. a) The mean squared error (MSE) and b) the standardized coefficients as a function of the regularization parameter for a representative set of 32 training samples within the cross validation LED selection procedure. The regularization parameter for the minimum MSE is found by analyzing the error from a nested LOOCV performed on the 32 training samples. This regularization parameter is then selected for the corresponding linear model.



Supplementary Figure 3. Ranking of LEDs in terms of the number of times they are used in linear models during the LASSO cross validation step for (a) the hexagonal and (b) square periodicity nano-hole array plasmonic sensors.

Supplementary Tables and Discussion

Supplementary Table 1. Hexagonal periodicity plasmonic sensor testing error (RIU).

LEDs used in model	LASSO	Tikhonov Regularization	Least Squares Solution
525	0.0039954	0.0039763	0.0039743
527	0.0041323	0.0041336	0.0041363
595	0.0027801	0.0027736	0.0027566
611	0.00074648	0.00074205	0.00075583
525 +527	N/A	0.0074443	0.010312
525 + 595	0.0023981	0.0024091	0.0023862
525 + 611	0.00068442	0.00068453	0.00068494
527 + 595	0.0023967	0.0023953	0.0024005
527 + 611	0.00070253	0.00070486	0.00070221
595 + 611	N/A	0.0007807	0.00078449
525 + 527 +595	N/A	0.0038764	0.0055691
525 + 527 + 611	N/A	0.00099724	0.0014449
525 + 595 + 611	N/A	0.00067642	0.00067551
527 + 595 +611	N/A	0.00069053	0.00069022
525 + 527 + 595 +611	N/A	0.00096541	0.0014056

Supplementary Table 2. Square periodicity plasmonic sensor testing error (RIU).

LEDs used in model	LASSO	Tikhonov Regularization	Least Squares Solution
500	0.0034	0.003354	0.003369
656	0.003869	0.003871	0.003864
660	0.003903	0.003878	0.003875
730	0.001392	0.001389	0.001407
500 +656	0.002202	0.002189	0.00219
500+660	0.002697	0.002714	0.00272
500+730	0.000972	0.000976	0.000939
656+660	0.003795	0.003784	0.003789
656+730	0.001512	0.00149	0.001484
660+730	0.0014	0.001411	0.00142
500 +656 + 660	0.001812	0.001809	0.001803
500+656+730	0.000971	0.000948	0.00096
500+660+730	0.000781	0.000779	0.000787
656+660+730	N/A	0.001495	0.001481
500+656+660+730	0.000748	0.000749	0.000753

Discussion:

The Tikhonov regularization is similar to the LASSO regularization (Eq. 1 of main text), differing only in the L-2 norm or the Euclidian norm, $\|\cdot\|_{\ell 2}$, *i.e.*,

Eq. S1 $$t^*_{mobile} = \underset{t_{mobile} \epsilon \mathbb{R}^{n+1}}{argmin} \left\| X_{OptLED} t_{mobile} - y \right\|^2_{\ell_2} + \|\Gamma t_{mobile}\|_{\ell_2},$$

which, similar to Eq. 3 of main text, can be used for computational sensing of the bulk refractive index:

Eq. S2 $$RIU_{prediction} = X_{test} t^*_{mobile},$$

The Tikhonov matrix, Γ, in Eq. S1 is generally employed as the regularization term and can be chosen in some cases to be a multiple of the identity matrix (I), *i.e.,* $\Gamma = \lambda I$, where λ is a scalar term referred to as the regularization parameter. The use of such regularization terms in both the Tikhonov and LASSO solutions allows for a computational sensing model which is tolerant to outliers while also accounting for the statistical variance (*e.g.*, due to fabrication) and average of the inputted features. The LASSO, however, is a very powerful optimization tool, because it forces many of the coefficients in the linear model to *zero*, due to the L-1 norm term. This process is often referred to as 'feature selection,' and in the context of this work discriminates the optimal subset of LEDs from the larger LED library. This feature selection property is precisely why many of the entries in the LASSO column of Supplementary Tables 1 and 2 are empty. When the LED features are inputted into the LASSO model, it sometimes forces the LED weights to zero, yielding a linear model that does not include all the inputted features (and is therefore not applicable for that specific entry of the comparison table).