

Autonomous Optimization of an Organic Solar Cell in a 4-dimensional Parameter Space

Supporting Information

A Materials

PM6: Poly[(2,6-(4,8-bis(5-(2-ethylhexyl-3-fluoro)thiophen-2-yl)-benzo[1,2-b:4,5-b'] dithiophene))-alt-(5,5-(1',3'-di-2-thienyl-5',7'-bis(2-ethylhexyl)benzo[1',2'-c:4',5'-c']dithiophene-4,8-dione)]

Y12: 2,2'-((2Z,2'Z)-((12,13-bis(2-butyloctyl)-3,9-diundecyl-12,13-dihydro-[1,2,5]thiadiazolo[3,4-e]thieno[2'',3'':4',5'']thieno[2',3':4,5]pyrrolo[3,2-g]thieno[2',3':4,5]thieno[3,2-b]indole-2,10-diyl)bis(methanylylidene))bis(5,6-difluoro-3-oxo-2,3-dihydro-1H-indene-2,1-diylidene))dimalononitrile

PC70BM: [6,6]-Phenyl-C71-butyric acid methyl ester

PM6 was purchased from Solarmer Materials Inc. (Beijing, China).

Y12 was purchased from Brilliant Matters Inc. (Quebec City, Canada).

PC70BM was purchased from Solenne BV (Groningen, The Netherlands)

ZnO (N10) nanoparticles were received from Avantama AG (Stäfa, Switzerland)

B Model Evaluation Bayesian Optimizer

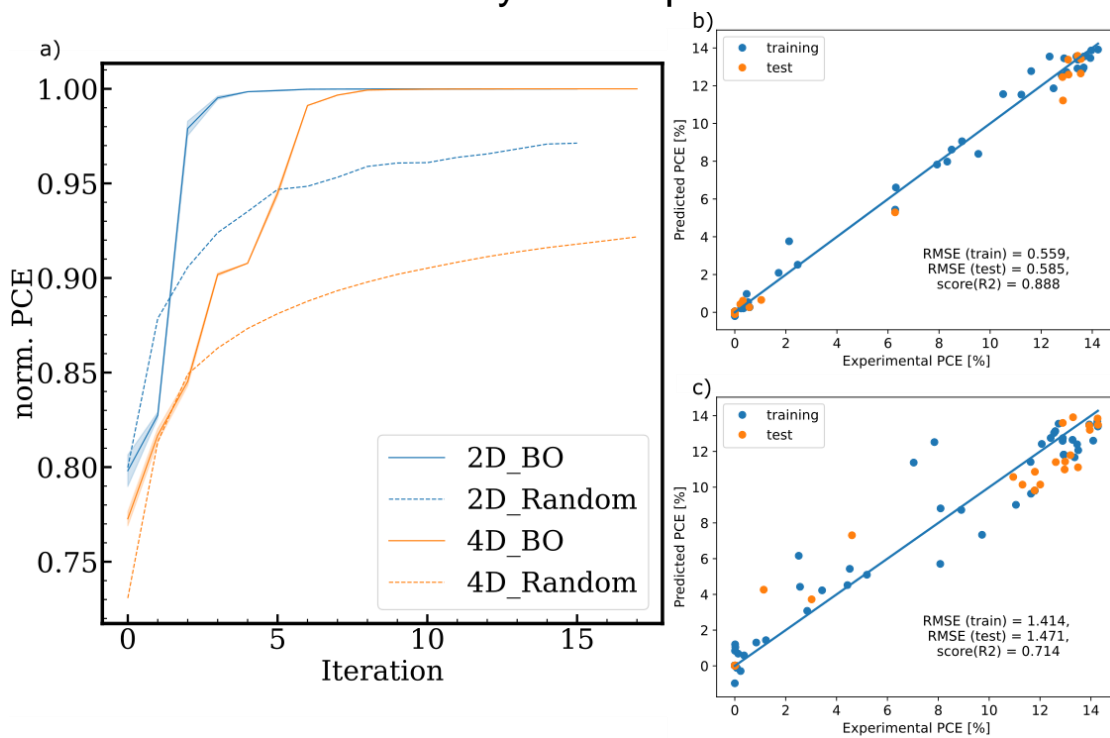


Figure S1: Model Evaluation. (a) The Bayesian Optimizer was run 50 times on the obtained objective function from the two optimizations. The solid lines (blue: 2D, orange: 4D) show the mean of the 50 runs of all devices' cumulatively highest normalized efficiency until the respective iteration, and the interquartile range is the shaded area. As a baseline, the dotted lines show the mean of 50 random sampling runs. The BO takes around five iterations to find the optimum. Note that we obtained the initial data sets of 7 and 21 samples by LHS sampling for the 2D and 4D, respectively. The experimental vs. the predicted PCE of the model of the (b) 2D and (c) 4D optimization using a 70/30 training test split 50 times.

C LineOne Reproducibility of the Reference Cells

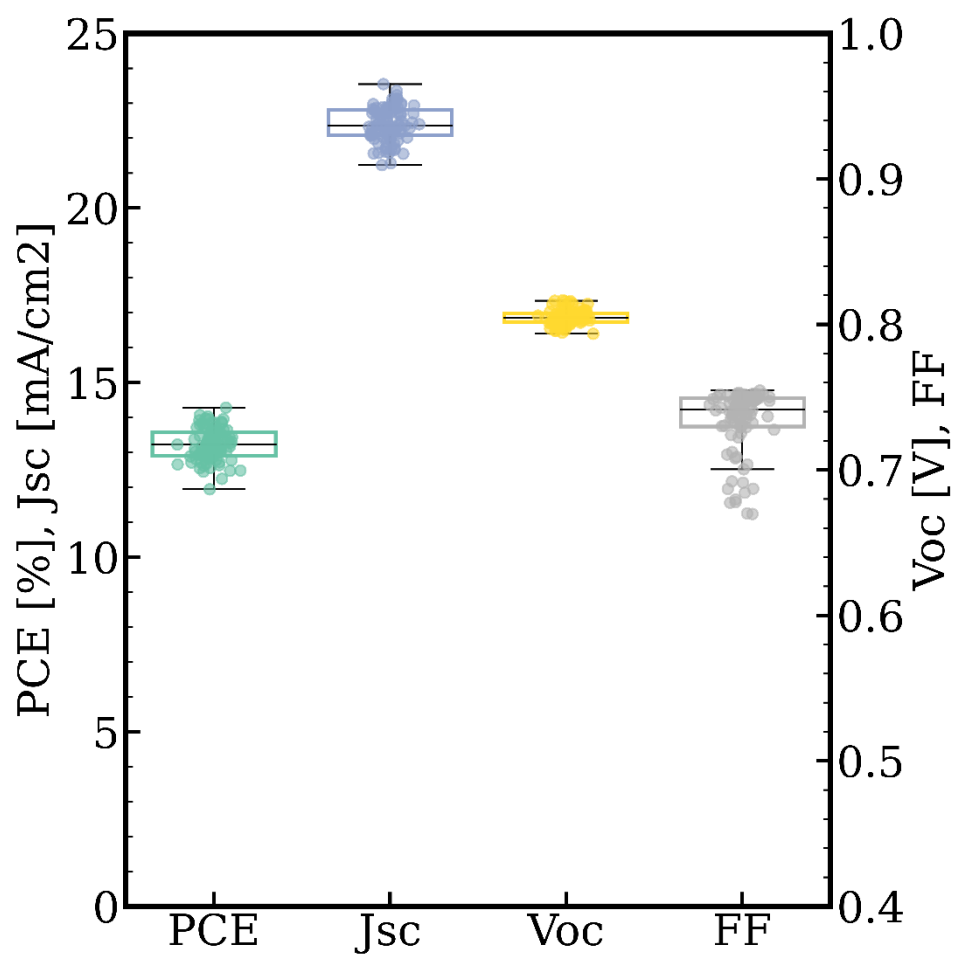


Figure S2: LineOne Reproducibility. Results of jV -measurements of the reference samples. In total, 18 cells were fabricated with the same parameters (PM6:Y12:PC70BM (1:1.2:0.2), 20 mg/mL, 1200 rpm) during the experiments. Each point represents one cell (6 cells per sample).

D Model Performance of the GPR to predict the cell efficiency

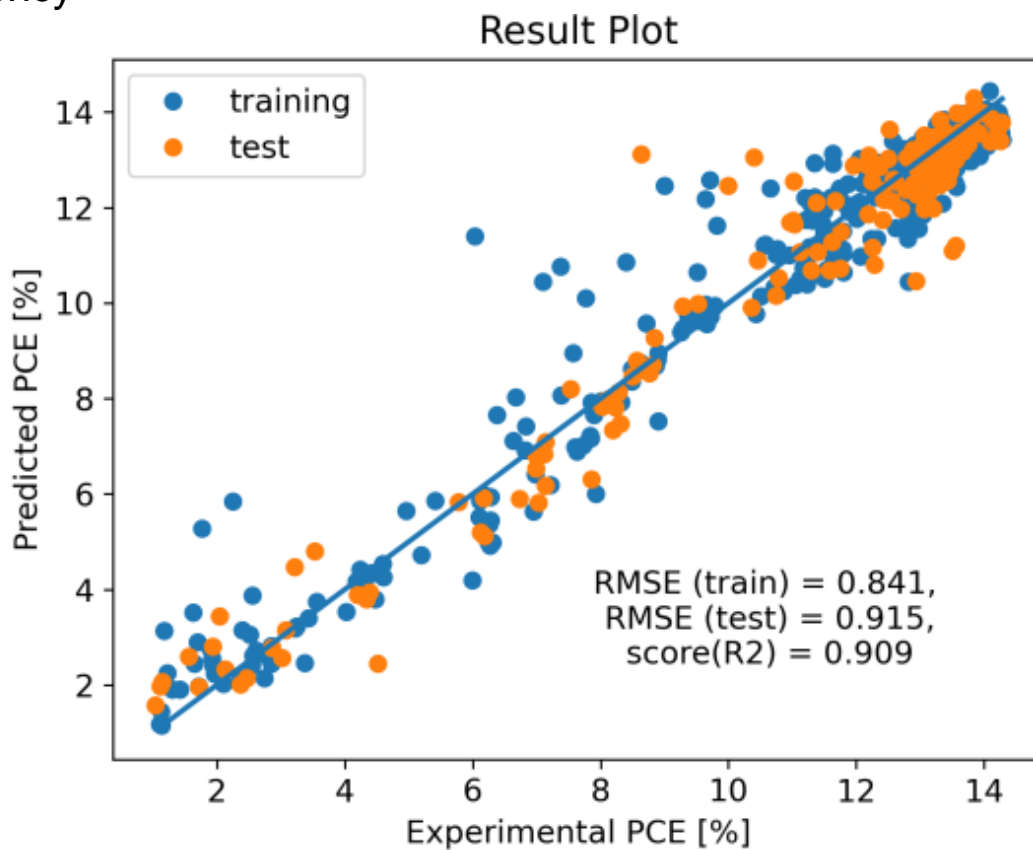


Figure S3: Model Performance of the PCE prediction using the UV-Vis data. The experimental vs. the predicted efficiency of the GPR model using a 70/30 test-train split 50 times. Each point represents one cell of the samples fabricated during the experiments. Cells with PCE < 1 % were excluded.

E Optimal spectral Features

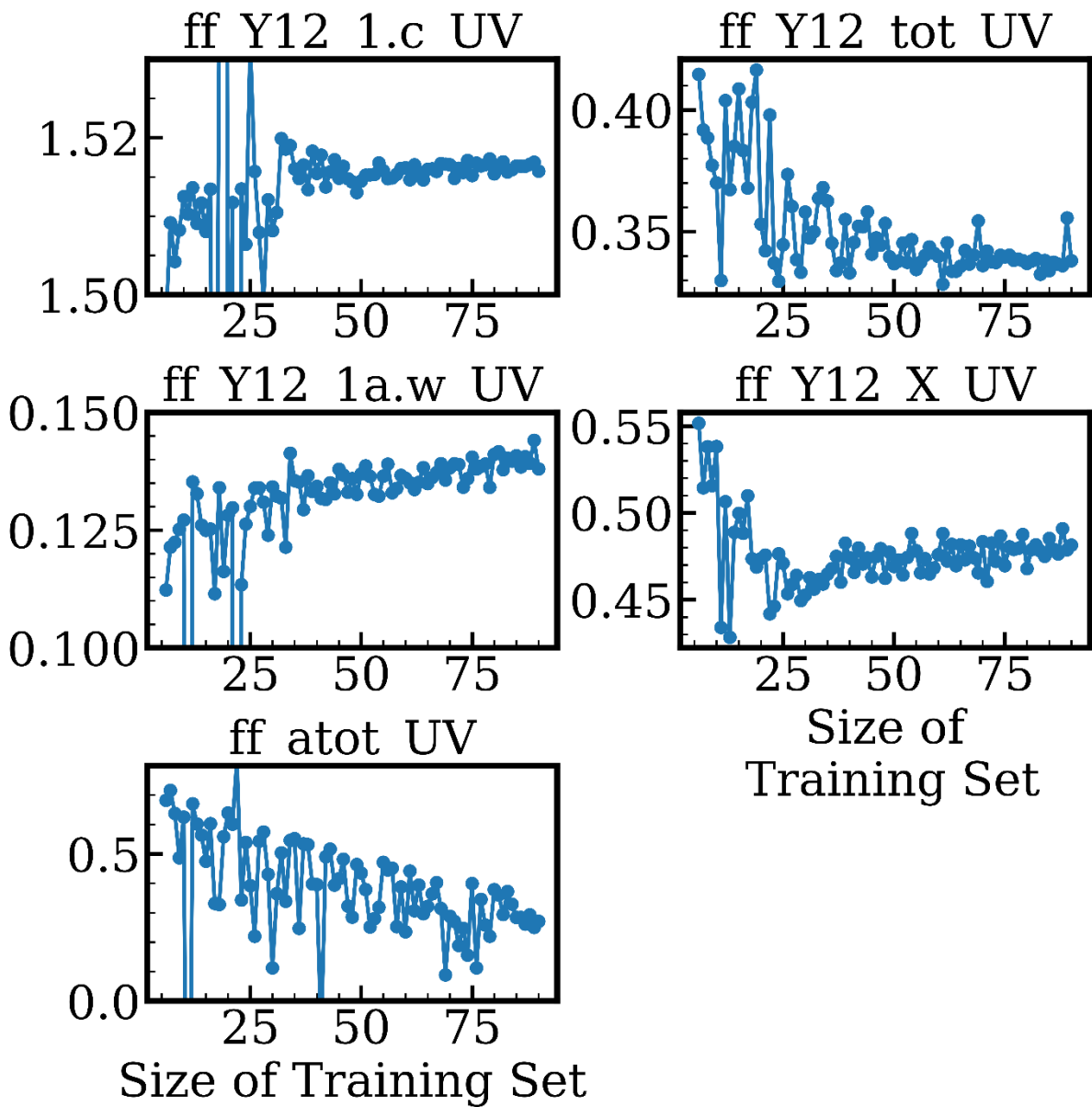


Figure S4: Predicted Optimum of each optical feature depending on the size of the training set.

F Distribution of the optical features in the dataset

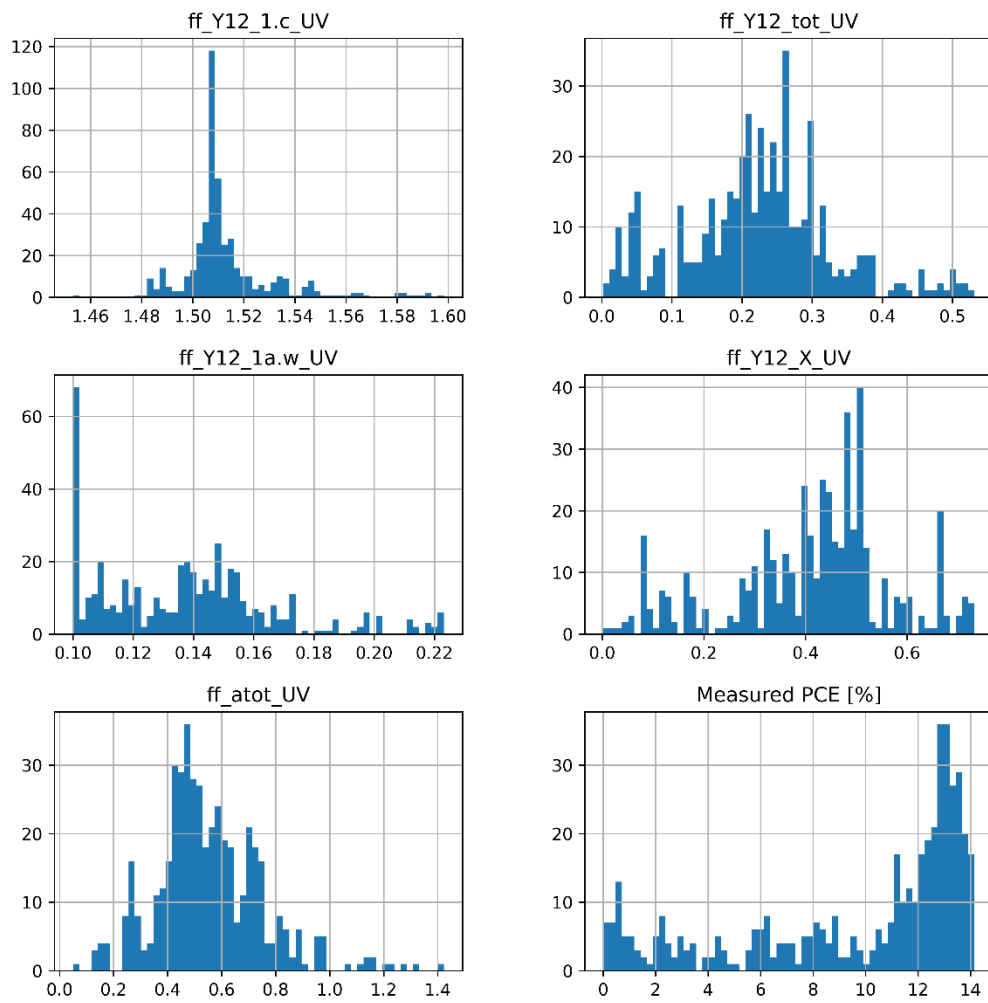


Figure S5: Distribution of the optical features used to predict the efficiencies of the cells.

G Uncertainty of the Bayesian Optimizer in the 4D experiment

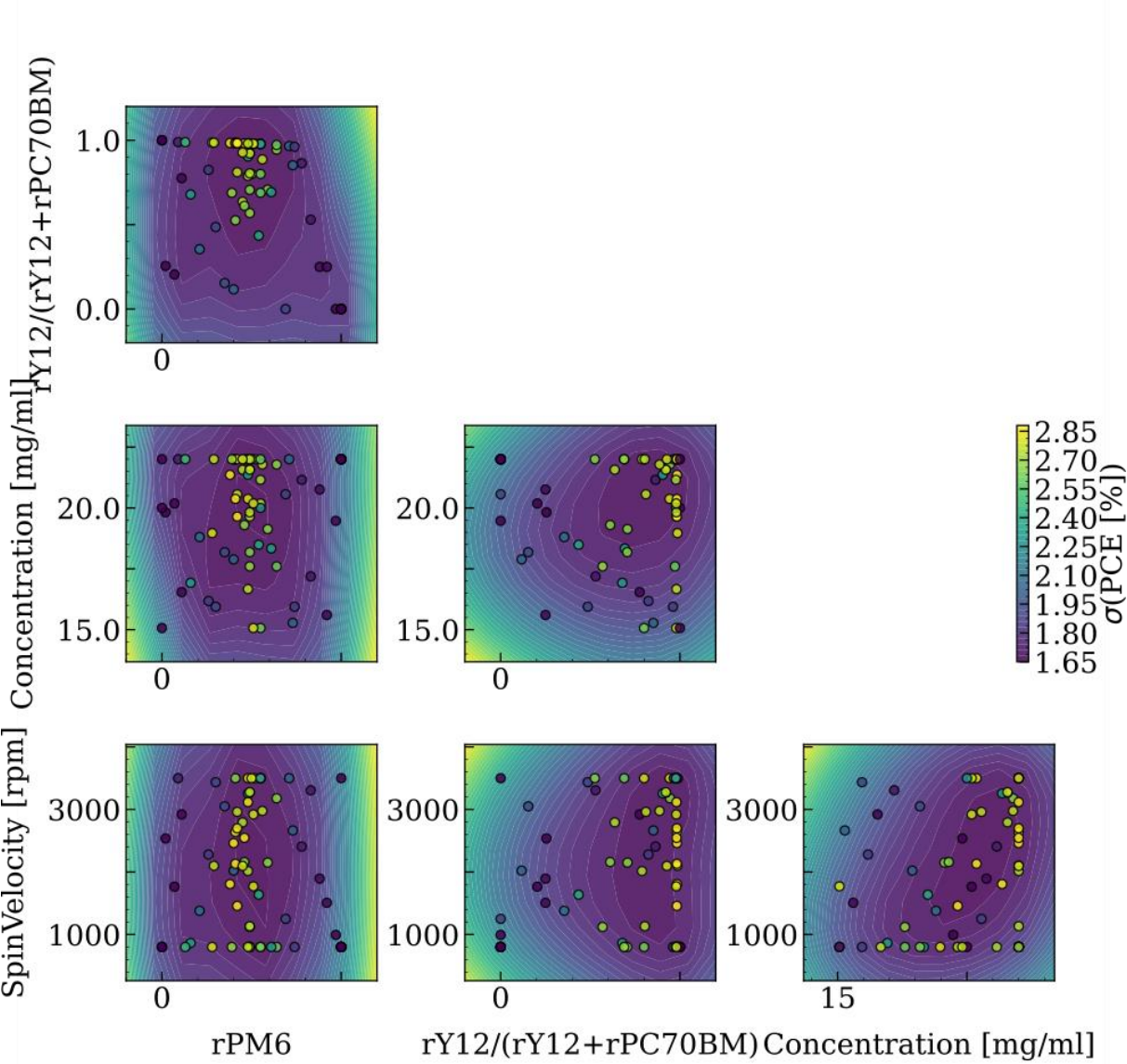


Figure S6: Uncertainty of the Bayesian Optimizer during the 4D Experiment.