Model-based Optimization of Laser-Reduced Graphene using in-situ Raman Analysis

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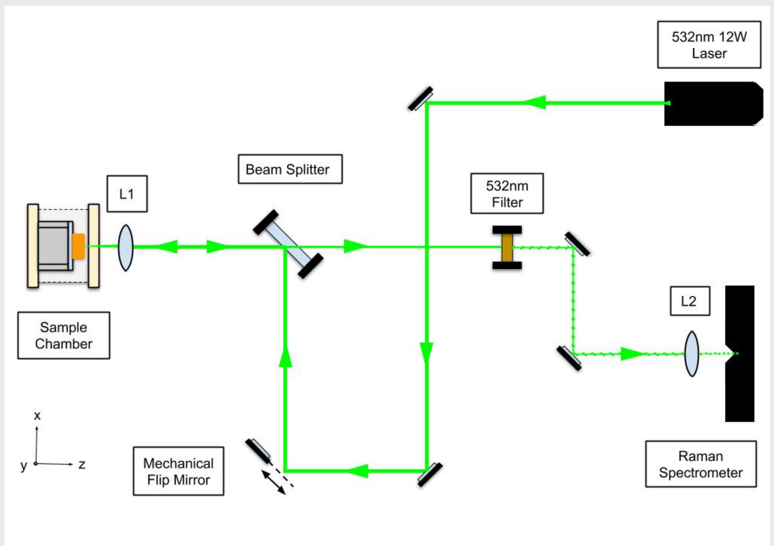
Motivation

The control of physical, chemical and electronic properties of laser-induced graphene (LIG) is crucial in the fabrication of flexible electronic devices for applications e.g. micro-supercapacitors [1] and fuel cell technology [2]. However, manual exploration of the parameter space is tedious and costly. Here, we demonstrate Bayesian model-based optimization (MBO) to advance strategies for property engineering of LIG. As a result, high-quality LIG patterns exhibiting high Raman G/D ratios, a factor of at least two compared to the literature [1, 3], was developed on average 15 predicted iterations. Human-interpretable conclusions may be drawn to distinguish substrate-dependent LIG. Our Bayesian optimization search method allows for an efficient experimental design that is independent of the experience and skills of individual researchers, while reducing experimental time and cost and accelerate materials research.

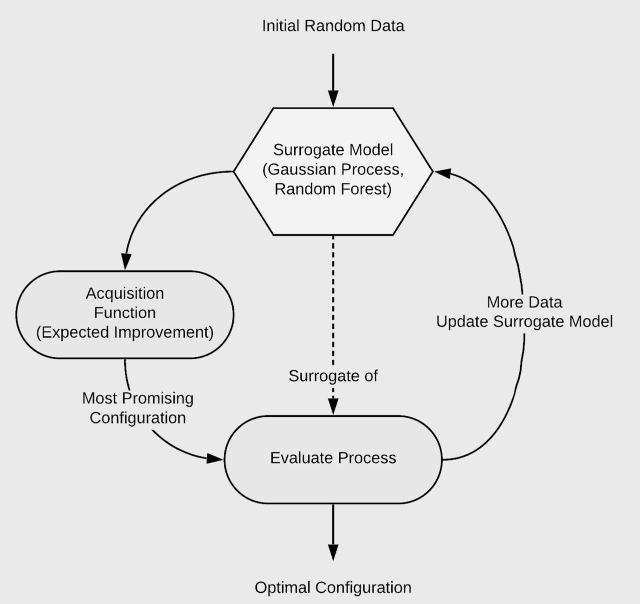
Methods

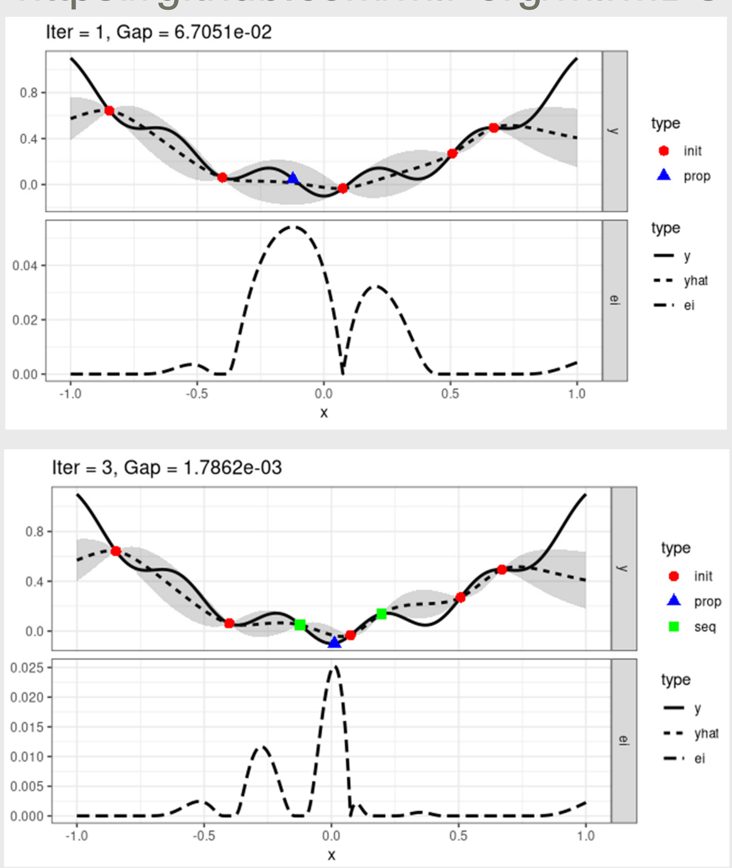


mlrMBO package

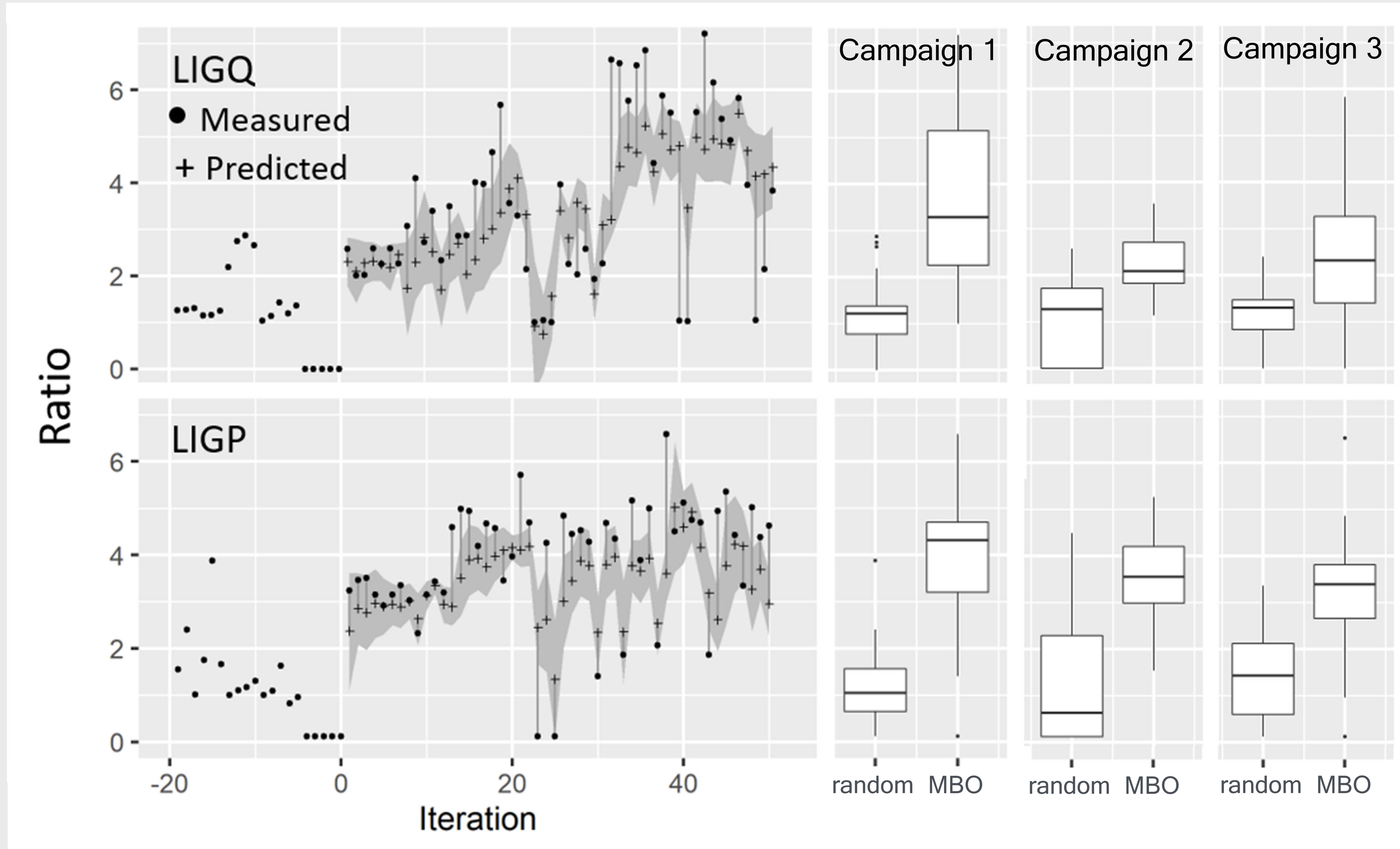
https://github.com/mlr-org/mlrMBO

Parameter space	Number of possible values	Size of parameter space
Laser power	554	
Irradiation time	19500	$3.2 \cdot 10^9$
Gas pressure	100	
Gas type	3	

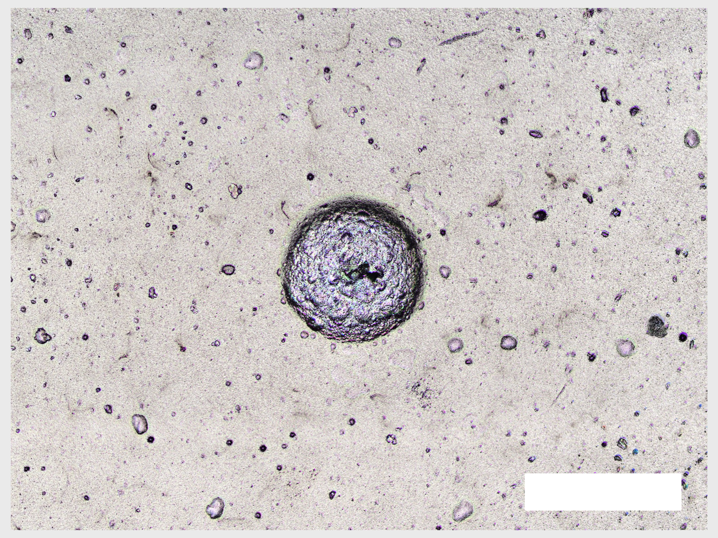
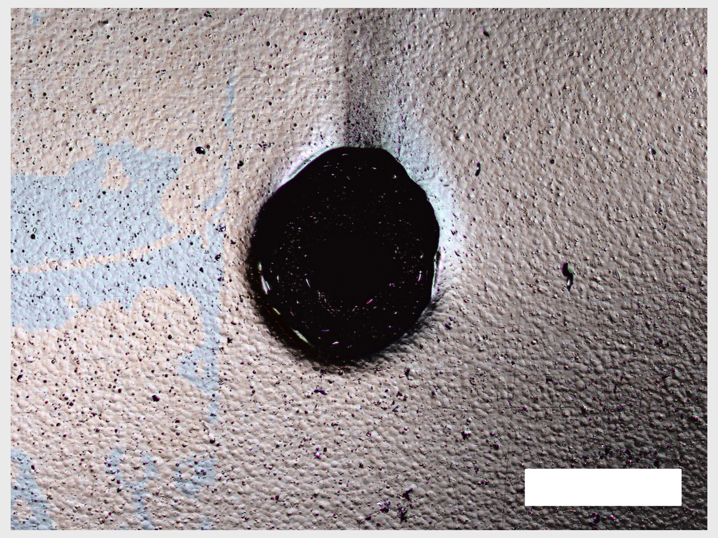




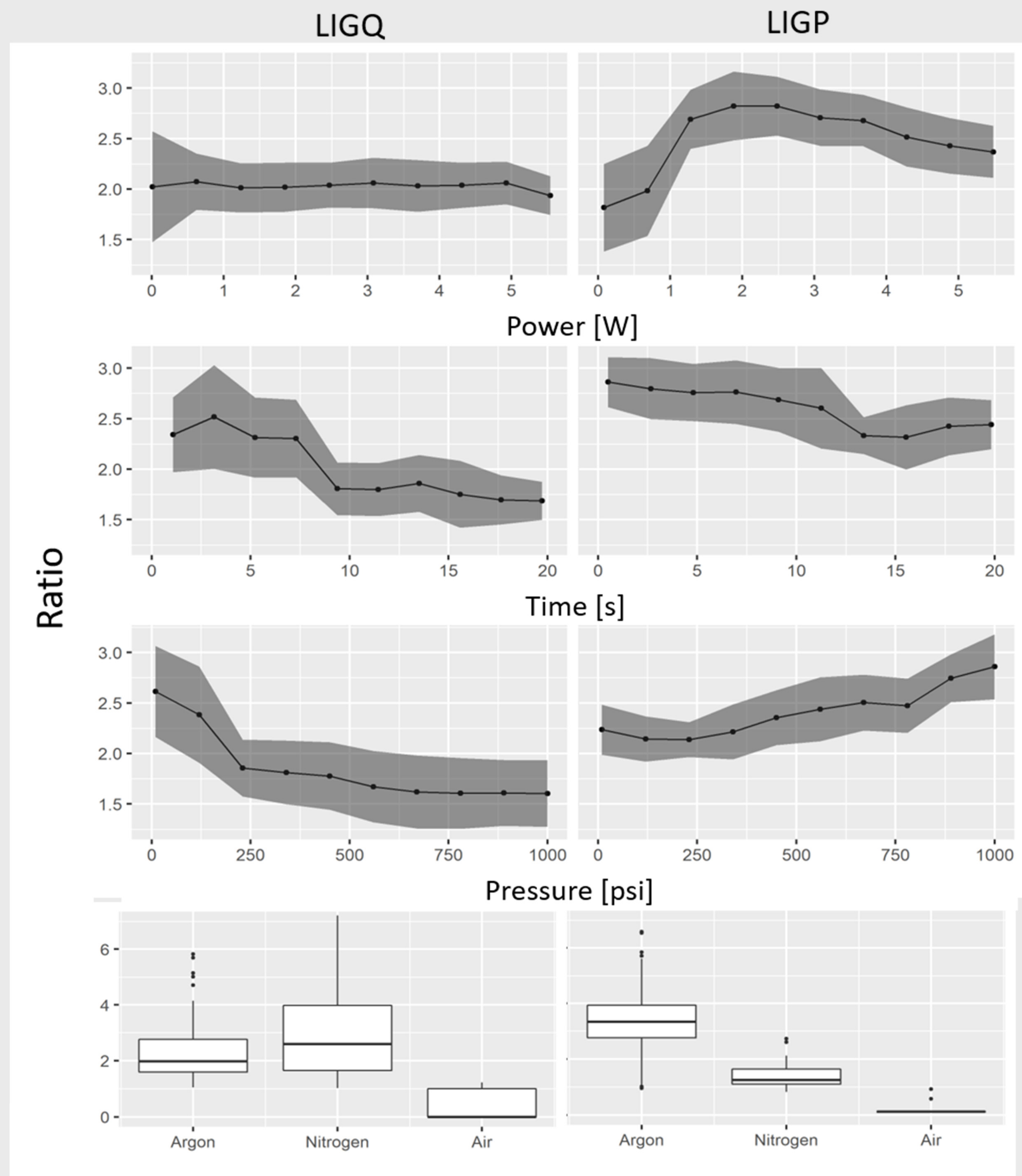
Results



Progress of G/D ratio optimization for LIG on quartz (LIGQ) and on polyimide (LIGP). Box plots corresponds to the statistic of randomly sampled (iteration <1) and MBO-assisted data (iteration >= 1). The optimization is statistically significant (t-tests $p < 0.05$). Campaigns repeated three times to account for random seed data.

Optical profilometer micrographs of MBO-assisted (left) LIGQ and (right) LIGP patterns. Bar is 20 μm.



Comparison of partial plot dependence G/D ratio to different parameters for LIG on quartz (LIGQ) and on polyimides (LIGP), as derived from the surrogate model predictions. The grey area denotes how the values vary with all possible values for other parameters. For both samples, optimized G/D is found at lower exposure times; >1W for LIGP; and lower and higher pressures for LIGQ and LIGP, respectively. The results may give insight into the underlying process of LIG formation.

Conclusions

Automated parameter tuning utilizing Bayesian model-based optimization improved quality of LIG patterns based on Raman G/D ratios by at least a factor of two in the literature on average in 15 predictions.

In spite of high G/D ratios, thermal damage may still occur. Future studies include multi-objective and multi-level optimizations.

Model was trained on random datasets without prior knowledge on the best configuration, providing efficient experimental design that is independent on skills of human researcher.

[1] In JB et al, Carbon 2015;83:144-151
[2] Ye R et al, ACS Nano 2015;9(9):9244-9251
[3] Tao Y et al, Applied Physics A 2012;106(3):523-531

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