
Applying Bayesian Optimization to Understand Tradeoffs for Antireflective Optical Designs

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Abstract

Many modern optoelectronic applications, from photovoltaic panels to smartphone displays, require antireflection coating to perform. Recent research has shown the promise of *random* subwavelength structure at reducing reflection; however, experimentation on additive manufacturing techniques for building nanostructures is time consuming. Such circumstances may benefit from thorough numerical simulations prior to actual manufacturing. In this paper, we propose adapting a machine learning strategy, Bayesian optimization, to efficiently search the design space of nanostructures that can minimize reflection at different angles. We use this sample-efficient strategy to identify nanostructure designs which minimize both normal and unpolarized reflection, so that we can fabricate only the most promising configurations.

1 Introduction

Optoelectronic devices such as solar cells require the protective surface layer to have low light reflection in order to increase light absorption, thus achieving higher energy conversion efficiency [19]. Modern high performance displays in TVs, smartphones, and computers also desire low reflectance for wider viewing angles. In recent years, research efforts have been focused on constructing bioinspired nanostructured antireflective materials [18, 5, 14]. Extending on the analysis of glasswing butterfly wings [20, 8], this research focuses on studying glass with random subwavelength structures. These nanostructures can generate a gradual change of refractive index between air and materials, thus reducing reflectance while improving light transmission.

Many design decisions, including those regarding the shape and the density of these nanostructures, can impact the performance of these devices. The fabrication process for these devices is time-consuming; using nanosphere lithography for additive manufacturing, a single glass panel of 1 cm^2 surface area may require over 24 hours of fabrication time to print millions nanostructures on the surface. In practice, numerical simulations are often used first to search for the optimal design of the nanostructure cells to guide the physical fabrication.

We turn to active learning, specifically Bayesian optimization [2], to efficiently search for the optimal designs. In particular, we are interested in finding the set of Pareto optimal design choices, devices that can simultaneously minimize reflection at multiple angles of incidence. Our goal is to *densely* sample the Pareto efficient frontier to thoroughly expose the tradeoffs between different design decisions.

2 Background

Recent research has demonstrated nanowires and nanocones to be among the best subwavelength structures for antireflection [20, 6, 7]. We are particularly interested in studying the reflection behavior of *randomly* nanostructured glass; by increasing the number of design parameters, we wish to create devices with superior antireflective performance to those with *uniform* nanostructures. We use the Lumerical commercial implementation of the finite-difference time-domain (FDTD) method to model and simulate the optical reflection of the nanowire and nanocone structures on glass substrate [15]. Such numerical simulations can be run prior to the physical fabrication. We measure and optimize the reflectance of each device at two different angles of incidence: 1) normal reflection at 90° incident angle (R_n), and 2) unpolarized reflection at 65° (R_u).

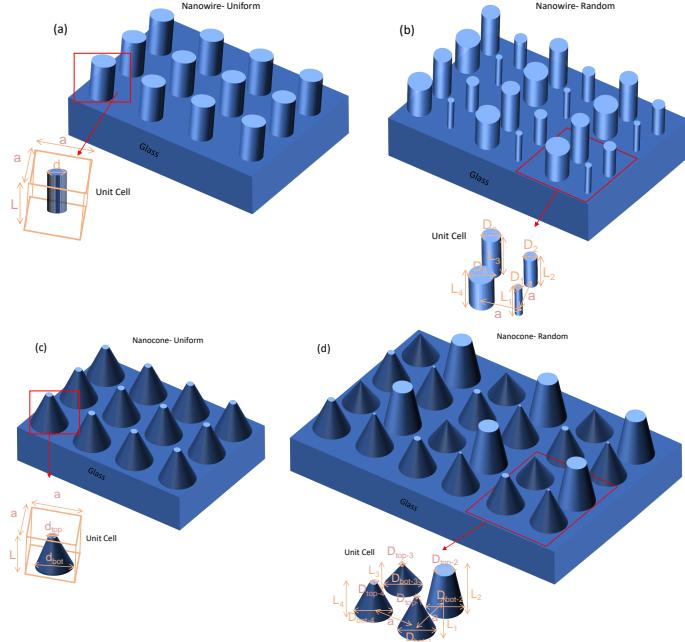


Figure 1: Schematic of simulated structures (a) and (b) show uniform and random nanowire structure, (c) and (d) show uniform and random nanocone structure on glass substrate, respectively. The unit cell and different design parameters of each structures are shown as well.

In the *uniform nanowire* setting, we define three design parameters: the pitch size or box size in which each nanostructure exists (a), the length (L) and diameter (D) of nanowire. For *random nanowire* structures, we consider four nanowires per unit cell (in 2×2 layout). Each cell has nine design parameters, including the pitch size (a), the length of each nanowire ($L_i, i \in \{1, 2, 3, 4\}$), and the diameters of each nanowire in the unit cell ($D_i, i \in \{1, 2, 3, 4\}$). Note that the pitch size is the same for all four nanowires in the unit cell. There are four parameters used for the *uniform nanocone* device: pitch size (a), length (L), top diameter D_{top} , and bottom diameter D_{bot} of the nanocones. *Random nanocone* structures also have four nanocones per unit cell. Each unit has 13 parameters, including the pitch size (a), the length of each nanocone ($L_i, i \in \{1, 2, 3, 4\}$), the top diameters ($D_{top,i}, i \in \{1, 2, 3, 4\}$), and bottom diameters ($D_{bot,i}, i \in \{1, 2, 3, 4\}$) of each of the four nanocones in the cell. The schematic of each device is illustrated in Figure 1.

Additionally, we need to impose physical constraints on the design space. In the nanowire settings, we limit the diameter of each nanowire to be $D_i \leq a$. In the nanocone settings, we also limit the bottom diameter of each cone to be $D_{bot,i} \leq a$. Furthermore, we restrict the top diameter to be $D_{top,i} \leq D_{bot,i}$, which preserves an upright, cone-shaped structure. Note that the design space of the nanocone structure is a subspace of that of nanowire structure.

We frame the search for the Pareto optimal designs as a multi-objective optimization problem that jointly minimizes the normal reflectance and unpolarized reflectance,

$$\min_{\mathbf{x} \in \mathcal{X}} R_n(\mathbf{x}), R_u(\mathbf{x}), \quad (1)$$

where \mathcal{X} is the space of all viable choices of the design parameters (those that satisfy the constraints stated above). We limit $5 \leq a \leq 400$ and $5 \leq L_i \leq 800$, both in nanometers.

This optimization setting poses several problems. We have to optimize (1) using only evaluations from the numerical simulator, without any additional information such as gradients. With a larger design parameter space (as in the case of random nanowires setting), we need to efficiently search for the desirable designs of nanostructures. At the same time, we also wish to densely sample the Pareto efficient frontier. These requirements lead us to investigating Bayesian optimization approach.

3 Bayesian Optimization

Bayesian optimization is a sample efficient method for optimizing black-box objective functions. Standard Bayesian optimization consists of two components: a *probabilistic model*, to model the objective function f , and an *acquisition function*, to determine which parameters \mathbf{x} to sample next.

In a standard Bayesian optimization setting, the objective function f is assumed to be a realization of a Gaussian process (GP) with mean function μ and a positive definite covariance kernel K , i.e., $f \sim \mathcal{GP}(\mu, K)$ [17, 1]. The mean and covariance functions are defined to have hyperparameters, such as length scales, which are learned through strategies such as maximum likelihood estimation. In all of our modeling, we assume our GPs to have $\mu \equiv 0$ and a C^4 Matérn kernel K with independent length scales in each dimension. A Tikhonov regularization (noise variance) parameter is fixed to be 10^{-3} , mainly to avoid ill-conditioning.

An acquisition function is a utility function that measures the value of sampling at different points within \mathcal{X} , given the observed data. Acquisition functions balance the tradeoff between *exploitation*, suggesting parameters near where we have observed the best results so far, and *exploration*, suggesting parameters in regions where we have not sampled. After n different input parameters have been tested, the n th probabilistic model can be created. The maximal argument of the acquisition function determines the next design parameters \mathbf{x} .

3.1 Multiobjective Bayesian Optimization

We describe our adaptation of Bayesian optimization to efficiently search for the Pareto optimal set of design parameters. The strategy is derived from the ϵ -constraint method [9]. We reformulate the multiobjective optimization problem (1) as two constrained scalar optimization problems:

$$\min_{x \in \mathcal{X}} R_n(x), \text{ s.t. } R_u(x) \geq \hat{R}_u, \quad (2a)$$

$$\min_{x \in \mathcal{X}} R_u(x), \text{ s.t. } R_n(x) \geq \hat{R}_n, \quad (2b)$$

where \hat{R}_n and \hat{R}_u are thresholds. By changing \hat{R}_n and \hat{R}_u throughout the optimization we can discover different sections of the Pareto efficient frontier.

In particular, we adapt techniques from the constrained Bayesian optimization literature [4, 3]. After k simulation runs have been observed, two independent GP models $s_{n,k}$ and $s_{u,k}$ are created for both normal reflectance and unpolarized reflectance. Using these models, an acquisition function is defined for each component of (2). This acquisition function is modified from the *expected improvement* [10] to account for the desire for viability. Considering, at first, only the solution to (2a), imposing the viability requires us to consider not only the distribution of $r_n \sim s_{n,k}(\mathbf{x})$ (a Gaussian distribution), but the joint distribution $r_n, r_u \sim s_{n,k}(\mathbf{x}), s_{u,k}(\mathbf{x})$. The acquisition function would be defined as

$$a_{R_n,k}(\mathbf{x}) = \mathbb{E}_{r_n, r_u \sim s_{n,k}(\mathbf{x}), s_{u,k}(\mathbf{x})}[(\tilde{r}_{n,k} - r_n)_+ I_{r_u \geq \hat{R}_u}], \quad (3)$$

where $\tilde{r}_{n,k}$ is the lowest R_n value observed thus far, $(\xi)_+$ denotes $\max(\xi, 0)$, and I_ν is the indicator function. In other words, we are maximizing the expected improvement for viable points; points which do not satisfy the threshold contribute zero improvement.

Since our design parameter space \mathcal{X} has linear constraints, we employ a multistart quasi-Newton optimizer that can take in these constraints into consideration when optimizing the acquisition function, namely Sequential Least SQuare Programming (SLSQP) [11, 13]. This ensures that all suggestions made by our algorithm are feasible. We also implement a hit-and-run sampling method for efficiently generating points within the constrained space for initializing the optimizer [21].

In the following section, we benchmark our multiobjective Bayesian optimization method against a popular multiobjective genetic algorithm NSGA-II (as natively implemented in Matlab) [16, 12].

4 Experimental Insights

We show the comparison of our method against NSGA-II in Figure 2a on optimizing the random nanocone structure (13 design parameters) with a simulation budget of 500. We see that our MOBO method vastly outperforms NSGA-II. The performance of genetic algorithms is highly sensitive to the population size used. A small population size is susceptible to getting stuck in local minima; a large population size can find a more global optima, but it requires more generations to converge. We show the effect of population size in Figure 2b, for a fixed function evaluation budget of 500.

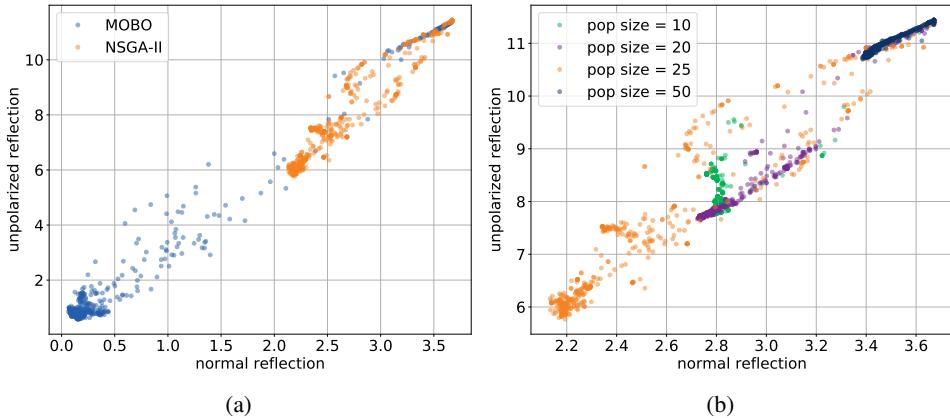


Figure 2: In panel (a), multiobjective Bayesian optimization versus Matlab NSGA-II on the random nanocone design optimization. Both optimization methods are run with 500 function evaluations. NSGA-II uses a population size of 25 and ran for 20 generations. In panel (b), results for NSGA-II with different population sizes and a fixed evaluation budget of 500 are shown.

Due to the limitation of modern fabrication strategies, we only compare the simulation results from uniform nanocones and uniform nanowires configurations, which can be more quickly and easily fabricated. From Figure 3a, we observe that by searching a larger design space, we can drastically improve the reflection performance of the materials. In Figure 3b, the Pareto optimal designs of nanocones completely dominate those of nanowires (which confirms that our method can efficiently search in a higher dimension in practice).

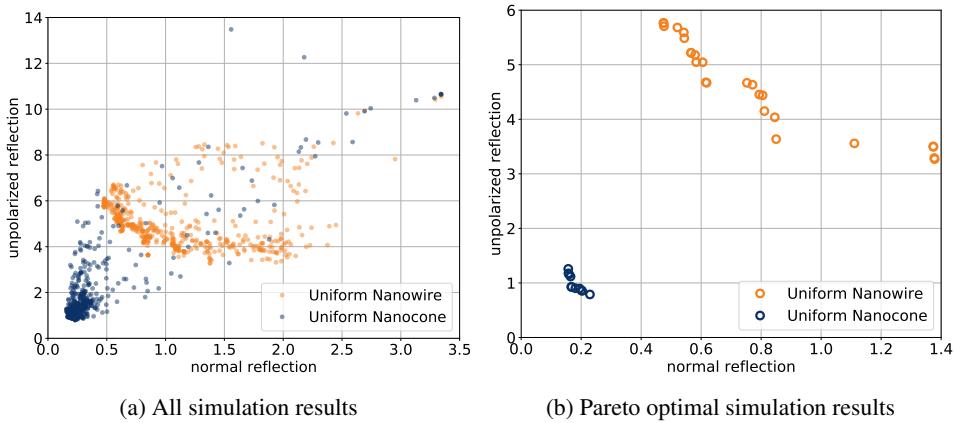


Figure 3: Simulation results for both uniform nanocone and nanowire structures. We ran our method for 500 function evaluations in all settings. In panel (b), we zoom in to inspect the Pareto-optimal results for each setting.

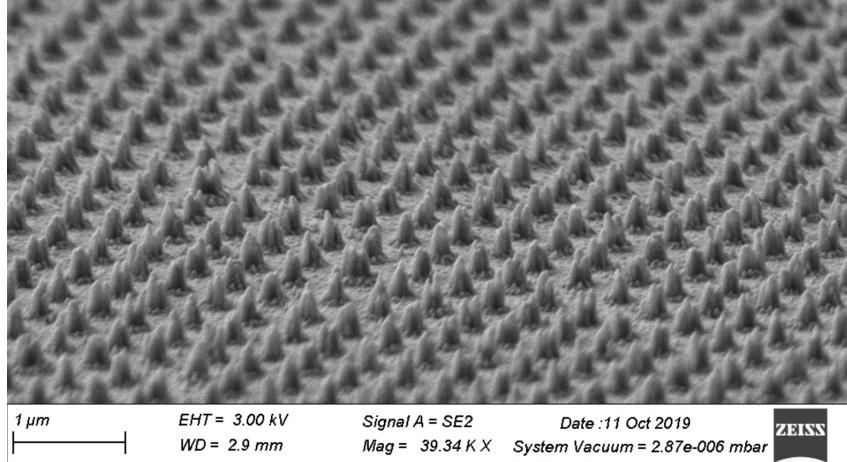


Figure 4: Sample fabrication of a Pareto efficient nanocone structure, identified using the frontier presented in Figure 3b, on a 2 cm by 2 cm glass substrate using nanosphere lithography.

4.1 Discussion on fabrication limitation

With the current state of the art, it is impossible to fabricate exactly the results that are being explored through computational simulations. Conventional methods for texturing surfaces such as sand blasting, powder blasting, and acid etching are unable to create features with sub-wavelength resolution. There are currently some 3D printers that can print features down to 200 nm with a spacing of less than 400 nm between features, but the areas are limited to about 1 cm^2 and the time it would take to print such structures would be over 24 hours and prohibitive. There are a variety of patterning methods such as the use of nanospheres and dewetted metal, that can be used for creating better defined structures in the glass, though there are still issues with minimum feature size and the uniformity of such samples over large areas. We show one sample of nanocone structures fabricated using this method in Figure 4. We have also been working with maskless reactive ion etching methods which create structures with more randomness in features [8].

5 Conclusion

In this paper, we investigate optimizing the reflectance properties of nanostructured glass through numerical simulations, with the specific goal of quantifying the impact of permitting nonuniformity/randomness in the nanostructures. Our results confirm conjecture within the community that nanocones provide better broadband and omnidirectional antireflection. While there exists some gap between simulation results and current fabrication methods, these BO assisted simulations help provide valuable information with regards to where in the vast design space we should be striving to fabricate structures. As nanomanufacturing methods continue to improve, these results offer guidance with regards to how we can improve the performance of such structures.

References

- [1] Gregory E. Fasshauer and Michael J. McCourt. *Kernel-based Approximation Methods Using Matlab*. World Scientific, 2015.
- [2] Peter I. Frazier. Bayesian optimization. In Esma Gel and Lewis Ntaimo, editors, *Recent Advances in Optimization and Modeling of Contemporary Problems*, pages 255–278. INFORMS, 2018.
- [3] Jacob Gardner, Matt Kusner, Zhixiang Xu, Kilian Weinberger, and John Cunningham. Bayesian optimization with inequality constraints. In Eric P. Xing and Tony Jebara, editors, *Proceedings of the 31st International Conference on Machine Learning*, volume 32 of *Proceedings of Machine Learning Research*, pages 937–945, Bejing, China, 22–24 Jun 2014. PMLR.

- [4] Michael A. Gelbart. *Constrained Bayesian optimization and applications*. PhD thesis, Harvard, 2015.
- [5] Yong H. Ghymn, Kinam Jung, Myunghun Shin, and Hyungduk Ko. A luminescent down-shifting and moth-eyed anti-reflective film for highly efficient photovoltaic devices. *Nanoscale*, 7(44):18642–18650, November 2015.
- [6] Sajad Haghifar, Tongchuan Gao, Rafael T. Rodriguez De Vecchis, Bradley Pafchek, Tevis D. B. Jacobs, and Paul W. Leu. Ultrahigh-transparency, ultrahigh-haze nanograss glass with fluid-induced switchable haze. *Optica*, 4(12):1522–1525, December 2017.
- [7] Sajad Haghifar, Ping Lu, Md Imrul Kayes, Susheng Tan, Ki-Joong Kim, Tongchuan Gao, Paul Ohodnicki, and Paul W. Leu. Self-cleaning, high transmission, near unity haze OTS/silica nanostructured glass. *Journal of Materials Chemistry C*, 6(34):9191–9199, 2018.
- [8] Sajad Haghifar, Michael McCourt, Bolong Cheng, Jeffrey Wuenschell, Paul Ohodnicki, and Paul W Leu. Creating glasswing-butterfly inspired durable antifogging omniphobic supertransmissive, superclear nanostructured glass through Bayesian learning and optimization. *Materials Horizons*, 2019.
- [9] C.L. Hwang and A.S.M. Masud. *Multiple objective decision making, methods and applications: a state-of-the-art survey*. Lecture notes in economics and mathematical systems. Springer-Verlag, 1979.
- [10] Donald R. Jones, Matthias Schonlau, and William J. Welch. Efficient global optimization of expensive black-box functions. *Journal of Global Optimization*, 13(4):455–492, Dec 1998.
- [11] Eric Jones, Travis Oliphant, Pearu Peterson, et al. SciPy: Open source scientific tools for Python, 2001–. [Online; accessed 2019].
- [12] Deb Kalyanmoy. *Multi-Objective Optimization Using Evolutionary Algorithms*. John Wiley & Sons, Inc., New York, NY, USA, 2001.
- [13] Dieter Kraft. A software package for sequential quadratic programming. Technical Report DFVLR-FB 88-28, DLR German Aerospace Center – Institute for Flight Mechanics, Koln, Germany, 1988.
- [14] Jung Woo Leem, Sehwan Kim, Soo Hyun Lee, John A. Rogers, Eunkyoung Kim, and Jae Su Yu. Efficiency Enhancement of Organic Solar Cells Using Hydrophobic Antireflective Inverted Moth-Eye Nanopatterned PDMS Films. *Advanced Energy Materials*, 4(8):1301315, 2014.
- [15] Lumerical device multiphysics simulation suite, fdtd solver, 2019. Lumerical Inc.
- [16] Matlab global optimization toolbox, 2019. The MathWorks, Natick, MA, USA.
- [17] Carl Edward Rasmussen and Christopher K. I. Williams. *Gaussian Processes for Machine Learning (Adaptive Computation and Machine Learning)*. The MIT Press, 2005.
- [18] Hemant Kumar Raut, V. Anand Ganesh, A. Sreekumaran Nair, and Seeram Ramakrishna. Anti-reflective coatings: A critical, in-depth review. *Energy Environ. Sci.*, 4:3779–3804, 2011.
- [19] Enzheng Shi, Hongbian Li, Long Yang, Luhui Zhang, Zhen Li, Peixu Li, Yuanyuan Shang, Shiting Wu, Xinming Li, Jinquan Wei, Kunlin Wang, Hongwei Zhu, Dehai Wu, Ying Fang, and Anyuan Cao. Colloidal Antireflection Coating Improves Graphene–Silicon Solar Cells. *Nano Letters*, 13(4):1776–1781, April 2013.
- [20] Radwanul Hasan Siddique, Guillaume Gomard, and Hendrik Hölscher. The role of random nanostructures for the omnidirectional anti-reflection properties of the glasswing butterfly. *Nature Communications*, 6:6909, April 2015.
- [21] Robert L. Smith. The hit-and-run sampler: A globally reaching markov chain sampler for generating arbitrary multivariate distributions. In *Proceedings of the 28th Conference on Winter Simulation*, WSC ’96, pages 260–264, Washington, DC, USA, 1996. IEEE Computer Society.