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## Fast Inverse Design of Microstructures via Generative Invariance Networks

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# Fast Inverse Design of Microstructures via Generative Invariance Networks

## Abstract

The problem of efficient design of material microstructures exhibiting desired properties spans a variety of engineering and science applications. An ability to rapidly generate microstructures that exhibit user-specified property distributions transforms the iterative process of traditional microstructure-sensitive design. We reformulate the microstructure design process as a constrained Generative Adversarial Network (GAN). This approach explicitly encodes invariance constraints within a GAN to generate two-phase morphologies for photovoltaic applications obeying design specifications: specifically, various short circuit current density and fill-factor combinations. Such invariance constraints can be represented by deep learning-based surrogates of full physics models mapping microstructure to photovoltaic properties. To circumvent data generation bottlenecks, we utilize a multi-fidelity surrogate that reduces the requirements of expensive labels by 5X. Our approach enables fast generation of microstructures (in~190ms) with user-defined properties. Such physics-aware data-driven methods for inverse design problems are expected to democratize and accelerate the field of microstructure-sensitive design.

## Keywords

User-specified Property Distributions, Two-phase Morphologies, Data Generation Bottlenecks, Multi-fidelity Surrogate

## Disciplines

Computer-Aided Engineering and Design | Other Mathematics | Structural Materials

## Comments

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# Fast Inverse Design of Microstructures via Generative Invariance Networks

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## ABSTRACT

The problem of efficient design of material microstructures exhibiting desired properties spans a variety of engineering and science applications. An ability to rapidly generate microstructures that exhibit user-specified property distributions transforms the iterative process of traditional microstructure-sensitive design. We reformulate the microstructure design process as a *constrained* Generative Adversarial Network (GAN). This approach explicitly encodes invariance constraints within a GAN to generate two-phase morphologies for photovoltaic applications obeying design specifications: specifically, various short circuit current density and fill-factor combinations. Such invariance constraints can be represented by deep learning-based surrogates of full physics models mapping microstructure to photovoltaic properties. To circumvent data generation bottlenecks, we utilize a multi-fidelity surrogate that reduces the requirements of expensive labels by 5X. Our approach enables fast generation of microstructures (in  $\approx 190$ ms) with user-defined properties. Such physics-aware data-driven methods for inverse design problems are expected to democratize and accelerate the field of microstructure-sensitive design.

## 1 Introduction

Advances in manufacturing (additive manufacturing, 3D printing, layer-by-layer deposition, real time control) allow us to precisely tailor the spatial distribution (i.e. microstructure) of materials. This opens up the possibility of microstructure-sensitive design, which involves identification of optimal material configurations that produce a desired property. Microstructure-sensitive design can impact a diverse array of applications ranging from membranes design to enhance water reclamation, battery electrode design to improve energy transport, and organic electronics active layer design to improve energy harvesting or sensing<sup>1,2</sup>. The systematic creation of *fast methods* for microstructure-sensitive design is, however, a challenging problem, usually due to the complexity of the ‘forward model’ that maps the microstructure to property. The availability of a fast inverse design framework will transform the field of microstructure-sensitive design, and significantly impact how we harvest, store and distribute energy and mass.

Over the past decade, a wide variety of approaches have been explored for solving the inverse microstructure design problem<sup>3-5</sup>. Traditionally, iterative optimization approaches have been the most popular strategy to search for microstructures that yield desirable characteristics. These approaches are typically time-consuming, computationally expensive, and often require manual supervision by domain experts. Furthermore, such approaches often lack the ability to generalize to new design constraints and require repeated exploration of the design space for each new design constraint or user choice. More generally, optimization based approaches are susceptible to challenges arising from (a) the combinatorial explosion of plausible microstructures, and (b) the computational complexity of function evaluation, i.e., solving the forward problem, especially for complex multi-physics problems. Thus, conventional optimization strategies based on using the full-physics forward model is a very challenging proposition, with limited deployment by groups with the skill set to use, and dedicated access to large computational clusters. As an example, current microstructure optimization approaches which rely on multiple forward-model PDE calculations for the discovery of a single optimal (inverse) design may typically require up to 160,000 CPU-hours for a given chemical system<sup>3</sup>. The push to democratize microstructure-sensitive design led to efforts focused on relaxing the challenges described above. Some approaches relied on developing cheaper, but less accurate surrogates for the forward model (example, using graphs<sup>3</sup>), while other approaches made the problem computationally tractable by severely constraining the design space (allowing only specific parameterized shapes)<sup>4</sup>.

In this context, recent advances in the field of deep learning and scientific machine learning show promise for solving inverse design problems<sup>6-12</sup>. Particularly promising are Generative Adversarial Networks (GANs)<sup>13</sup>, a class of generative

28 deep learning models. Given a set of data, these generative models are capable of learning the underlying data distribution to  
29 generate new, realistic-looking samples. In the context of engineered systems, GANs have been successfully applied to areas  
30 such as differential equations<sup>14</sup>, system-modelling<sup>15,16</sup>, and material and drug discovery<sup>17-19</sup>.

31 Generative models can be trained to reconstruct realistic looking microstructures<sup>17,20,21</sup>. The challenge is to train them  
32 to reconstruct microstructures that satisfy a user-defined set of properties – or more generally, satisfying a set of constraints.  
33 Recently, a modified version of generative models called Invariance Networks (InvNets), have been proposed to enable  
34 imposition of explicit constraints on the model outputs<sup>22</sup>. The formulation of InvNet, which allows constraints to be defined  
35 independently, provides great flexibility in terms of incorporating domain knowledge or user specifications, cast as constraints.  
36 In this work, we formulate the microstructure-sensitive design problem into that of training an InvNet with physics-based  
37 constraints. We deploy this framework to generate candidate two-phase microstructures/morphologies for organic photovoltaic  
38 (OPV) applications due to this microstructure’s potential aspect in addressing a broad range of problems elaborated below.  
39 Flexible, light-weight, and wearable electronics and solar cells made from organic components provide a promising solution to  
40 wide array of societal needs. The potential application of these devices range from sensing (for precision and personalized  
41 medicine) to ambient energy harvesting (indoor solar cells) and energy efficient lighting and electronics. For example, the  
42 newest generation of small molecule acceptors have pushed single-junction organic photovoltaic (OPV) efficiencies over  
43 14% and tandem efficiencies over 17%, potentially revolutionizing cheap, flexible, and green energy harvesting. A large  
44 body of work has demonstrated that the morphology in the active layer of OPV devices is key to enabling high-performance  
45 devices<sup>23-25</sup>. Thus, controlling the morphology in the active layer of these devices continues to be crucial for maximizing  
46 performance. Tremendous advances in self-assembly of flexible polymers suggest remarkable control of hierarchical structure,  
47 but the impact on high-performance organic electronics has been limited. This is because, despite the importance of active layer  
48 mesoscale morphology to OPV device performance, it remains a challenge to identify “ideal” microstructures that maximize  
49 power conversion efficiencies. A key question is whether multiple ‘families’ of optimal morphology exist, and whether these  
50 morphological characteristics depend on material specific parameters such as electron mobility, exciton diffusion length and  
51 biomolecular recombination (i.e. the molecular chemistry). Thus, a rapid, physics-aware microstructure design strategy will  
52 enable practitioners to systematically explore and unravel questions of how materials limitations affect optimal morphological  
53 features, thereby accelerate materials design leading to high-performance devices.

54 Here, we train an InvNet to generate microstructures that simultaneously obey multiple constraints, specifically a user  
55 defined short-circuit current density  $J_{sc}$  and fill-factor  $FF$ . These two properties characterize the current-voltage performance  
56 of an OPV (for a given material system). The short circuit current density,  $J_{sc}$ , represents the maximum amount of current per  
57 unit area that can be drawn across a solar cell (when the applied voltage is zero). Meanwhile, the fill-factor  $FF$  denotes the  
58 maximum amount of power that can be supplied by the solar cell as a ratio of peak theoretical power. These properties are  
59 intimately (and non-trivially) influenced by morphology<sup>26</sup>. We first demonstrate that a deep neural network (DNN) can be  
60 trained as a surrogate model to accurately predict the values of  $J_{sc}$  and  $FF$  given a two-phase microstructure. This requires a  
61 substantial amount of full fidelity training data, and only serves as a baseline forward model surrogate. Next, we propose a  
62 multi-fidelity neural network that achieves a similar predictive accuracy while utilizing a small fraction of full fidelity labels  
63 alongside low-fidelity labels. We then formulate an InvNet based inverse design framework where these surrogate (baseline as  
64 well as multi-fidelity) models are used as invariances to generate morphologies that satisfy user-design specifications.

## 65 Results

66 We begin with a brief overview of our proposed methodology of using InvNets for fast generation of targeted two-phase  
67 morphologies. Figure 1(a) illustrates the overall InvNet framework with a Wasserstein-GAN (WGAN)<sup>27</sup> formulation. The  
68 WGAN model architecture ensures that the distribution of generated morphologies matches the true data distribution. Design  
69 specifications are enforced via an explicit invariance constraint, whereby the invariance loss is computed using the surrogate  
70 physics model represented by a deep neural network. This invariance loss ensure that the generator produces morphologies that  
71 satisfies the invariance constraint. Figure 1(b) and (c) shows two alternatives of surrogate models: a high-fidelity convolutional  
72 neural network (CNN) trained with a large amount of expensive labels obtained from high-fidelity simulations, and a multi-  
73 fidelity network trained with a mixture of high- and low-fidelity labels. The multi-fidelity network is trained on low-fidelity but  
74 computationally cheap labels alongside a fraction of high-fidelity labels to reduce the overall computation cost.

75 We present our results in the following order. First, we validate our CNN surrogate model for accurately predicting  
76 photovoltaic properties of a given morphology, followed by comparative assessment of prediction results of the multi-fidelity  
77 surrogate model. We then provide illustrative results of the microstructures generated by InvNet using both the high-fidelity  
78 network as well as the multi-fidelity network.

79 [Figure 1 about here.]

## 80 High-fidelity short circuit current and fill-factor estimation

81 In this section, we validate the approach of utilizing a deep neural network as a surrogate of the physics-based forward model.  
82 This surrogate model also serves as the invariance constraint within the InvNet framework. As deep neural networks are  
83 known to be powerful function approximators with fast prediction times<sup>28</sup>, we hypothesize that they are suitable candidates for  
84 representing the physics-based forward models.

85 We previously showed that a CNN can accurately classify microstructures in binned classes of  $J_{sc}$ <sup>5</sup>. Here, extend the idea to  
86 train a regressor that is capable of predicting  $J_{sc}$  and  $FF$  as continuous values. We train a CNN-based high-fidelity regressor  
87  $R_{HF}$  on a dataset of two-phase morphology images with high-fidelity simulated  $J_{sc}$  values ranging from 0 to 7 mA/cm<sup>2</sup> and  $FF$   
88 values ranging from 0.4 to 0.8 (see Methods and Supplemental Materials for details on full physics simulations and material  
89 properties). The output of this surrogate model is a vector which consists of the estimated  $J_{sc}$  and  $FF$  values. Training this  
90 model yields a  $R^2$  statistic of 0.994 for the estimations of  $J_{sc}$  and  $R^2$  statistic of 0.928 for estimations of  $FF$  as seen in bottom  
91 two scatter plots of Figures 2(a). This suggests that the surrogate model is capable of estimating the properties with sufficiently  
92 high accuracy. The top two plots in Figure 2(a) shows the histograms of absolute error for both  $J_{sc}$  and  $FF$  respectively. The  
93 error distribution of  $J_{sc}$  has a mean of  $0.002 \pm 0.08$  mA/cm<sup>2</sup> while the error distribution of  $FF$  have a mean magnitude of  
94  $7.68E-5 \pm 0.02$ .

95 [Figure 2 about here.]

## 96 Reducing data cost with multi-fidelity labels

97 An expected bottleneck of training a surrogate model on high-fidelity labels is the challenge of initial data generation, which  
98 may be computationally expensive. Various works in literature have previously explored the idea of leveraging both high- and  
99 low-fidelity data to accelerate computational models<sup>29–32</sup>. We exploit a similar idea by proposing a multi-fidelity surrogate  
100 model to circumvent the challenge of generating computationally expensive high-fidelity labels.

101 Having demonstrated that the high-fidelity surrogate is capable of estimating  $J_{sc}$  and  $FF$ , we next provide a preliminary  
102 overview of the computation of low-fidelity labels, followed by results of estimating such low-fidelity labels via another neural  
103 network surrogate. Then, we present the results of the multi-fidelity surrogate model which alleviates the requirement of a  
104 large-labelled dataset required for training. We achieve this by using computationally inexpensive low-fidelity labels and a  
105 fraction of expensive high-fidelity labels to train the multi-fidelity surrogate model.

106 We use our prior work where a mechanistic consideration of the photo physics as three distinct processes (absorption and  
107 generation of excitons; exciton diffusion and dissociation; charge transport and collection) allowed identification of three  
108 morphology descriptors that together showed high correlation with  $J_{sc}$ <sup>33,34</sup>. These descriptors are computed by representing the  
109 two-phase morphology as a weighted-graph and evaluating standard graph measures (like connected components and path  
110 lengths). Since the complexity of graph-based problems and corresponding algorithms are well understood, these descriptors are  
111 computationally inexpensive to compute. These morphology descriptors thus provide a low fidelity link between morphology  
112 and performance. We next describe how using only a small fraction of simulated high-fidelity labels ( $J_{sc}$  and  $FF$ ) along  
113 with information from low-fidelity labels (morphology descriptors), we train a surrogate model that has the similar predictive  
114 performance as a model trained purely using high-fidelity labels. Training this multi-fidelity surrogate required availability of  
115 differentiable low-fidelity descriptors, which we get by training another neural network,  $R_g$ , that maps a morphology to the  
116 low-fidelity descriptors (see SI for results, network and training details)

## 117 Multi-fidelity short circuit current and fill-factor estimation

118 We train a multi-fidelity model that estimates the magnitudes of  $J_{sc}$  and  $FF$  using a limited amount of high-fidelity labels with  
119 the help of the low-fidelity descriptors. This multi-fidelity network consists of the low-fidelity network,  $R_g$  and a separate  
120 shared-embedding network, as illustrated in Figure 1(c). The purpose of the shared embedding network is to learn additional  
121 features that are useful in estimating the properties which were not captured by the low-fidelity model. In our experiments, we  
122 used only 20% of randomly sampled high-fidelity labels to train the multi-fidelity network, which resulted in a  $R^2$  of 0.989 and  
123 0.894 for  $J_{sc}$  and  $FF$  respectively. The absolute error distributions of the  $J_{sc}$  and  $FF$  predictions have a mean of  $-0.009 \pm 0.12$   
124 mA/cm<sup>2</sup> and  $-0.003 \pm 0.02$  respectively. Figure 2(b) shows the scatter plots of the properties estimated by the multi-fidelity  
125 model against the ground truth values as well as the distribution of errors. As seen in Figure 2(a) and (b), the  $R^2$  of the  $R_{HF}$  and  
126  $R_{MF}$  models are similar although the label requirements of the multi-fidelity model is reduced by 80%. We stress that while the  
127 low-fidelity network was trained using the entire dataset, the multi-fidelity model was only trained with 20% of the high-fidelity  
128 labels, which are significantly more expensive to generate (e.g., evaluating the  $J_{sc}$  and  $FF$  of one morphology needs about 1  
129 cpu-hr, whereas the low fidelity metrics can be computed in less than a minute). Hence, by using the multi-fidelity network,  
130 we alleviate the problem of requiring a large labelled dataset to train a surrogate physics model as the invariance constraint  
131 evaluator in the InvNet.

## 132 Targeted Microstructure Generation

133 [Figure 3 about here.]

134 We present the results of generating targeted morphologies that are tailored to design specifications using our proposed  
135 *InvNet* with multi-fidelity surrogate model framework. In Figure 3(a), we show samples of microstructures generated with  
136 *InvNet* for different design specifications. In the top row, we show examples of morphologies with low  $J_{sc}$  values and high  $FF$   
137 values. As we traverse down the rows of Figure 3(a), the specified  $J_{sc}$  values are increased while the  $FF$  values are decreased.  
138 It is observed that the *InvNet*-trained generator is able to generate a variety of candidate microstructures with different  
139 morphologies given the same design specifications. This signifies that the generator has learnt the underlying distribution of the  
140 actual data and no mode collapse occurred during training which can result in only similar morphologies being generated. This  
141 also anecdotally validates a hypothesis in the OPV community that there exist multiple families of morphologies that produce  
142 identical performance.

143 To further verify that the generated morphologies satisfy the imposed design constraints, we generated an additional 1000  
144 morphologies for different ranges of  $J_{sc}$  and  $FF$  values and compared the estimated properties of these morphologies with the  
145 actual design specifications. The values of these estimated properties and design specifications are plotted as densities and  
146 shown in Figure 3(b). We observe that the specified values and generated values for both  $J_{sc}$  and  $FF$  have highly overlapping  
147 densities. These overlapping densities show that generator is capable of creating morphologies that satisfy the imposed design  
148 specifications, hence enabling targeted design of candidate two-phases microstructures.

149 Nonetheless, we observe that there are situations where the generated morphologies do not adhere to the design specifications,  
150 as seen in the first row of Figure 3(b), where the density of generated morphologies (in solid green) had a range of  $J_{sc}$  values that  
151 are higher than the specified range of  $J_{sc}$  values (in dotted blue). Since the proposed framework is fundamentally data-driven,  
152 we hypothesize that this failure mode was caused by an imbalanced dataset where samples from the low  $J_{sc}$  and high  $FF$   
153 regions might be sparse. To confirm this hypothesis, we visualize the training data distribution in Figure 3(c). Based on the  
154 visualization of the joint density, we observe that there are indeed very few samples in the top left region, where morphologies  
155 have a low  $J_{sc}$  and high  $FF$  values. However, it is interesting to recognize that even when the generator fails to generate  
156 morphologies with specified  $J_{sc}$  in such sparse training data regions, the rank order of the morphologies'  $J_{sc}$  are still preserved.  
157 Instead of generating morphologies with random  $J_{sc}$ 's, the generated morphologies defaulted to morphologies with low  $J_{sc}$  and  
158 high  $FF$  values which are well supported with data.

## 159 Comparing high-fidelity and multi-fidelity *InvNets*

160 Next, we provide qualitative results to compare the effects of using the high-fidelity,  $R_{HF}$ , and multi-fidelity  $R_{MF}$  surrogate  
161 model as the invariance constraint evaluator in *InvNet* framework. In Figure 2, we have shown that the performances of the high-  
162 and multi-fidelity surrogate models are comparable. Moreover, we are also interested in investigating if the higher variance  
163 of the multi-fidelity surrogate will compound and affect the results of the generated morphologies. To study this, we trained  
164 *InvNet* with the same network architecture and replaced the  $R_{MF}$  with  $R_{HF}$ . We illustrate the results from both methods in  
165 Figure 4. In terms of the generated morphologies, we do not observe any significant difference between the two methods. Both  
166 the high- and multi-fidelity *InvNets* are capable of generating microstructures of varying morphologies without signs of mode  
167 collapse. However, the density plots which are used to validate the constraint invariances reveal two interesting observations.

168 First, we observe that the high-fidelity *InvNet* is more capable of generating low  $J_{sc}$ /high  $FF$  morphologies in comparison  
169 with the multi-fidelity *InvNet*. This is evident in the first row, where the density of morphologies generated by high-fidelity  
170 *InvNet* has a higher overlapping area with the design specifications as compared to the density of morphologies created by  
171 multi-fidelity *InvNet*. We attribute this to the fact that  $R_{HF}$  was exposed to a much larger and diverse set of morphologies as  
172 compared to  $R_{MF}$ , which results in the high-fidelity *InvNet* being able to learn the underlying structure of the low  $J_{sc}$ /high  $FF$   
173 morphologies better when training for the invariance. Thus, this suggests we can expect the performance of high-fidelity *InvNet*  
174 to be more robust and consistent when queried in regions where training data is sparser.

175 The second interesting observation we make is that the high-fidelity *InvNet* also tends to generate morphologies that are  
176 a little more biased in terms of the  $FF$ . This can be observed in the second, third, and fourth rows where the densities of  
177 high-fidelity  $FF$  are slightly shifted from the  $FF$  design specifications. Referring back to Figure 3(c), we observe that the  
178 marginal density of  $FF$  data is highly skewed towards the lower regions. Therefore, it is possible that by training  $R_{HF}$  on  
179 the entire high-fidelity dataset and subsequently using it as the invariance constraint evaluator to train *InvNet* does result in  
180 generated morphologies that are more biased in terms of the design specifications. This highlights the importance of having a  
181 balanced dataset when using our proposed framework for morphology generation.

182 [Figure 4 about here.]

## Efficiency of neural-network based methods versus physics-based models

In Table 1, we compare the wall-clock running times of our proposed neural-network based methods with physics-based methods for a few different scenarios. All timings were performed on the same platform using a NVIDIA Titan RTX GPU and averaged across 100 function evaluations. In the first two columns, we show the average computation times for evaluating the  $J_{sc}$  and  $FF$  properties of a given morphology. We observe that both multi- and high-fidelity methods are several orders of magnitude faster than a high-fidelity physics simulation. A second advantage is that with the surrogate models, only one evaluation is required to estimate both  $J_{sc}$  and  $FF$  simultaneously. In comparison, performing the physics simulation requires separate individual evaluations for  $J_{sc}$  and  $FF$ . Comparing the multi-fidelity surrogate model  $R_{MF}$  with the high-fidelity surrogate model  $R_{HF}$ , we note that  $R_{HF}$  is an order of magnitude faster than  $R_{MF}$ . However, training  $R_{HF}$  comes at the cost of requiring a large dataset with high-fidelity labels. On the other hand,  $R_{MF}$  requires a smaller amount of high-fidelity labels, but requires training a more complex model architecture, which increases computation time. Hence, we view the benefits of each method as a trade-off between availability of data with computation time.

In the third column, we show the total time required to train InvNet for 1E5 epochs. We observe that the high-fidelity InvNet is  $\approx 3X$  faster than multi-fidelity InvNet, which is expected since the training of InvNet is dependent on the surrogate model to compute the invariance loss. We also include an estimate of the time required to train the InvNet if we were to replace the invariance constraint evaluator with an actual physics-based model to compute the invariance loss. As observed, training such an InvNet will require  $\approx 60k$  hours, which is not tractable in compared to using a neural network-based surrogate model.

Last but not least, we provide the morphology generation time for a single morphology. Since the process of generating a morphology using InvNet during inference is independent of surrogate model, there is no significant difference time difference between using the high-fidelity versus multi-fidelity InvNet. In summary, we conclude that there is no significant difference in terms of the querying a trained high-fidelity versus multi-fidelity InvNet to generate targeted morphologies. Instead, the deciding factor of which model to apply depends on the availability of high-fidelity labels or computation resources. The high-fidelity InvNet framework is faster to train but requires a large dataset of high-fidelity labels to pre-train the surrogate model. Conversely, the multi-fidelity InvNet model requires less high-fidelity labels but requires a more complex network architecture which results in longer training times.

[Table 1 about here.]

## Discussion

The ability to rapidly synthesize targeted microstructure designs is essential in a broad range of scientific and engineering applications. We propose a data-efficient generative framework (InvNet) that casts user-specifications as explicit invariance constraints to generate candidate two-phase microstructures that adheres to design specifications. While recent works with similar objectives have proposed frameworks that demonstrated promising results<sup>12,22</sup>, we highlight that those approaches is not capable of solving our specific application in a tractable manner. This is particularly due to the extremely long and expensive computation required to evaluate the constraints, which is a common bottleneck in the community. Hence, to remedy this challenge, we leverage neural network-based surrogates for the purpose of fast constraint evaluation. Using a surrogate, our framework addresses the challenge of expensive constraint evaluation while simultaneously circumventing the need of having a differentiable and explicit, closed-form expression of the constraints. Combining these advantages, we believe that our method results in a far more general-purpose framework that is applicable to a wider range of inverse design problems. Additionally, we have also supplemented our surrogate-based generative framework with a multi-fidelity approach to improve the data requirements of the model. This multi-fidelity approach reduces foreseeable expensive label generation procedures, which is an obstacle that is not present in inverse design problems where design constraints can be tractably computed. For further discussion on the motivation of our framework, we defer reader to the Methods section. From our experimentation, our results illustrate that neural networks are capable of being accurate surrogates of expensive full-physics simulations and the InvNets trained with multi-fidelity surrogates are capable of generating various candidate morphologies which caters to both  $J_{sc}$  and  $FF$  specifications. Furthermore, comparing the results of InvNet trained with high-fidelity and multi-fidelity surrogates reveals no significant differences in performance, thus reinforcing the fact of data-efficiency benefits of using the multi-fidelity surrogate. A wall-clock comparison of training times reveals that a trade-off exists between the high-fidelity and multi-fidelity modes, with the multi-fidelity version of the surrogate and InvNet having longer training times.

While we have demonstrated our proposed framework through the lens of a material microstructure design problem that uses a data-driven surrogate, we emphasize that our InvNet framework is certainly not limited to purely data-driven surrogate approaches. Since the invariance constraint of InvNet is explicit, it can be easily replaced or combined with other data-free approaches. In this regard, a key future direction is to develop InvNets that explicitly incorporate complex physics/domain knowledge in a computationally tractable manner. This approach will significantly reduce the dependency of the proposed framework on data availability and extend the capability of the framework to extrapolate beyond the support of data. Other

236 promising directions include extending the current framework to generate morphologies with more than two phases as well as  
 237 validating the generalizability of the framework on a dataset with more than two target properties. To conclude, our vision is  
 238 that the computational tools developed in this paper will serve to democratize and accelerate the area of microstructure-sensitive  
 239 design.

## 240 Methods

### 241 Description of two-phase morphology microstructures

242 **Microstructures:** We use a large dataset of microstructure images arising from solving the Cahn-Hilliard (CH) equation  
 243 with varying initial conditions. The Cahn-Hilliard equation<sup>35</sup> describes phase separation occurring in a binary alloy under  
 244 thermal annealing. It tracks the evolution of local volume fraction of each phase, in the presence of spatial gradients in  
 245 chemical potential of the system. Hence, in the time evolution process, one first observes an initial rapid separation of the  
 246 well-mixed system into its constituent phases, followed by slow coarsening of the respective domains. Thus, the microstructures  
 247 generated will have lower energy compared to those at the beginning stages of the simulation, according to the second law of  
 248 thermodynamics. Image data arising from the simulations provide a rich dataset for design of microstructures. Specifically, the  
 249 morphologies obtained through the simulation will be similar to the morphologies in a real active layer of organic photovoltaic  
 250 cells<sup>5</sup>. We use an in-house solver for generating the microstructure images.

251 **Photophysics Annotation of Microstructures:** Each of the morphologies is virtually interrogated to extract its current-  
 252 voltage characteristics, by solving a morphology aware (i.e. spatially heterogeneous) photophysics device model. We deploy  
 253 a validated, in-house software that uses a finite element based solution strategy for solving the photophysics device model.  
 254 The photophysics model is described by the steady state *excitonic drift diffusion (XDD) equations*. The XDD equations are a  
 255 set of four tightly coupled partial differential equations that model the optoelectronic physics of energy harvesting in organic  
 256 photovoltaic devices. The photophysics consists of the following stages:

- 257 • Incident solar radiation causes the generation of energetically active electron-hole pairs, called excitons (denoted by  $X$ ),  
 258 in the donor regions of the microstructure. These excitons diffuse across the microstructure and have a finite lifetime  
 259 before becoming ground state electron-hole pairs;
- 260 • Excitons that diffuse and reach the donor-acceptor interface undergo dissociation into electrons (denoted by  $n$ ) and holes  
 261 (denoted by  $p$ ) at the donor-acceptor interface. The dissociation mechanism is material and field dependent (denoted by  
 262  $D$ );
- 263 • These generated charges ( $n,p$ ) traverse the microstructure and reach their corresponding electrodes (cathode and anode)  
 264 to produce a current. Two mechanisms are responsible for driving carrier transport or current flow. First, the drift, which  
 265 is caused by the presence of an electric field (denoted as the gradient of the potential,  $\nabla\phi$ , and second, the diffusion,  
 266 which is caused by a spatial gradient of electron or hole concentration;
- 267 • The distribution of electrons and holes in the microstructure interacts with the applied voltage and influences the  
 268 electrostatic potential  $\phi$  across the microstructure. Finally, electrons and holes can recombine (denoted by  $R$ ) to create  
 269 excitons

The photophysics described above is encoded as the exciton drift diffusion (XDD) equations<sup>26</sup>.

$$\nabla \cdot J_n - R + D = 0 \quad (1)$$

$$-\nabla \cdot J_p - R + D = 0 \quad (2)$$

$$\nabla \cdot (\epsilon_r \epsilon_0 \nabla \phi) = q(n - p) \quad (3)$$

$$-\nabla \cdot (V_t \mu_x \nabla X) - f D_{[\nabla \phi, X]} - R_{[X]} = -G - R_{[n,p]} \quad (4)$$

270 Here,  $X$ ,  $n$ ,  $p$  represent the exciton, electron and hole distributions respectively.  $\phi$  represents the electric potential.  $q$   
 271 represents the elementary charge.  $V_t$  represents the thermal voltage.  $\epsilon$  is the dielectric constant in the donor and recipient  
 272 materials.  $\mu_{n/p/X}$  are the mobilities of electron/hole/exciton respectively. The current densities  $J_n$  and  $J_p$  are given by the  
 273 constitutive equations

$$J_n = -qn\mu_n \nabla \phi + qV_t \mu_n \nabla n \quad (5)$$

$$J_p = -qp\mu_p \nabla \phi - qV_t \mu_p \nabla p \quad (6)$$

274 These set of high-dimensional, complex PDEs are solved to get the performance of the solar cell device, which is  
 275 characterized by the short-circuit current, and fill factor.

## 276 Framework development

277 In this section, we discuss the motivation of creating a data-driven framework capable of generating microstructures with  
278 various targeted morphologies while adhering to design specifications. Previous studies have demonstrated that InvNets can  
279 effectively generate novel two-phase microstructures that satisfy explicit constraints such volume fractions and domain size and  
280 also generate poly-crystalline microstructures (a discrete-valued generation problem) by relaxing the generation problem to a  
281 probabilistic assignment problem<sup>22</sup>. However, we consider a couple of drawbacks of the existing InvNet in terms of scalability.  
282 Previously, it has only been shown that InvNet worked with explicitly defined constraints or invariances. Nevertheless, existing  
283 works have not addressed what happens when the invariances cannot be explicitly defined. Additionally, evaluation of the  
284 invariances are often computationally expensive and time-consuming. For example, in the case of our application, evaluating  $J_{sc}$   
285 and  $FF$  of a given morphology involves solving a set of differential equations that can take up to approximately several hours.

286 As such, this limitation hampers the scalability of training InvNet. Hence, we represent such invariance constraints, which  
287 cannot be explicitly expressed or are too computationally expensive with a deep neural network. By representing the invariance  
288 with a deep neural network surrogate, the evaluation of the invariance constraints can be significantly accelerated since the  
289 forward evaluation of a neural network is fast once the model is trained. Utilizing a neural network surrogate also has the benefit  
290 of not requiring the invariances, such as the equations governing a physical system, to be explicitly known as long as labeled  
291 data are available to train the surrogate model. Also, a neural network representation of the invariance simplifies the training of  
292 InvNet. During training of the InvNet, the parameters of the entire model are optimized by utilizing gradient information from  
293 the invariance loss function. Since neural network models are differentiable, gradient information with respect to the invariance  
294 loss can be easily computed using modern deep learning libraries with automatic-differentiation capabilities. In comparison,  
295 using other forms of explicit invariances will necessitate the constraints to be differentiable, and the gradients will have to be  
296 calculated separately.

297 Nonetheless, as alluded above, representing the invariance with a deep neural network does result in a second drawback,  
298 which is the availability of labeled data. In the context of our application, creating a labeled data set of morphologies with  
299 corresponding  $J$  and  $FF$  values is computationally expensive, and defeats the goal of avoiding costly physics-based simulations.  
300 This second drawback motivates the development the multi-fidelity surrogate which alleviates the problem of generating  
301 expensive labels.

## 302 Training details

303 **High-fidelity surrogate model:** To improve the robustness of the surrogate model, we first performed standard image  
304 augmentation techniques, image rotation and flipping, which resulted in an augmented dataset of  $\approx 38k$  images of augmented  
305 morphologies. To ensure a stable training process, we also scaled the labels of  $J_{sc}$  and  $FF$  to belong in the same numerical  
306 range. Following standard practices, we partitioned 80% of the data as training data and reserved 20% of data as a test data.  
307 Since the task of the surrogate model is to essentially perform a multi-target regression, the loss function of the regressor is  
308 formulated as:

$$L_{R_{HF}} = \|R_{HF_{\phi, J_{sc}}}(I) - J_{sc}\|_2^2 + \|R_{HF_{\phi, FF}}(I) - FF\|_2^2 \quad (7)$$

309 where  $R_{HF}$  denotes the high-fidelity surrogate model, parameterized by parameters  $\phi$ ,  $I$  is the input image of the microstructure  
310 and  $J_{sc}$  and  $FF$  are the true label values. The high-fidelity model architecture we used is a sequential model which consists  
311 of two convolution layers, each followed by batch normalization layer, ReLU activation, and a max pooling layer. Two dense  
312 layers were used after the two convolution blocks along with dropout layers to avoid over-fitting during training. The output  
313 of  $R_{HF}$  is a vector of two values that corresponds to the estimated  $J_{sc}$  and  $FF$  values. The model was trained using Adam  
314 optimizer with a learning rate of  $3E-4$  for 25 epochs. Additionally, we also investigated network architectures with separate  
315 final layers that do not share parameters. We observed no significant improvement in prediction accuracy while the cost of  
316 computational memory requirement was increased.

317 **Multi-fidelity surrogate model:** Before describing the training details, we briefly justify the need to replace the graph-based  
318 computation of low-fidelity descriptors with another neural network surrogate,  $R_g$  in the multi-fidelity model. While multi-  
319 fidelity frameworks are effective in reducing the requirement of expensive labels<sup>32</sup>, they are currently not tractable for application  
320 as an invariance constraint in InvNets. This is because updating the generator’s parameters in InvNet requires the gradient  
321 computation of the invariance-loss function. However, graph-based methods used to compute the low-fidelity descriptors are  
322 often non-differentiable. Therefore, optimizing the parameters of the generator via conventional back-propagation becomes  
323 a non-trivial problem. Additionally, evaluating the low-fidelity descriptors using previously proposed graph-based method  
324 requires that the generated images be converted into nodes and edges on-the-fly during training, which incurs additional  
325 computational cost and time. Hence, a neural network surrogate which is differentiable and can directly evaluate graph features  
326 of morphologies in the pixel domain circumvents both of these challenges.

327 As illustrated in Figure 1(c), the multi-fidelity network encompasses both low-fidelity network (described in SI) and a  
 328 shared-embedding network. The purpose of the shared-embedding network is to learn additional features that are not already  
 329 captured by the low-fidelity network for estimating  $J_{sc}$  and  $FF$ . During training of the multi-fidelity network, the low-fidelity  
 330 network predicts the low-fidelity descriptors of a given microstructure, which are combined with the image embeddings from  
 331 the shared embedding network. These two vectors are then passed through a dense layer to estimate  $J_{sc}$  and  $FF$ . As we are only  
 332 using a limited amount of high-fidelity labels, it is possible that training the multi-fidelity network might lead to a biased model  
 333 due to label imbalance. To avoid such issues, we constructed the following weighted loss function with empirically-determined  
 334 scaling constants that balances the errors between the estimations of  $J_{sc}$  and  $FF$ .  $R_{MF}$  denotes the multi-fidelity surrogate  
 335 model where  $\chi$  and  $\omega$  represents the parameters of the shared-embedding network and low-fidelity network respectively.

$$L_{R_{MF}} = L_{J_{sc}} + L_{FF} \quad (8)$$

$$L_{J_{sc}} = \lambda_1 (J_{sc}^2 + J_{sc}) \|R_{MF_{\chi, \omega, J_{sc}}}(I) - J_{sc}\|_2^2 \quad (9)$$

$$L_{FF} = \lambda_2 \|R_{MF_{\chi, \omega, FF}}(I) - FF\|_2^2 \quad (10)$$

338 with  $\lambda_1$  and  $\lambda_2$  heuristically set to 0.008 and 0.0005, respectively. We highlight that in principle, the weights of the low-fidelity  
 339 network  $R_g$  are already trained and can be frozen. Nevertheless, in practice, we find that allowing the weights of the low-fidelity  
 340 network to optimized alongside the entire network does result in a slightly better estimations. To train the multi-fidelity network,  
 341 we used SGD optimizer with a learning rate of 1E-3 and trained the network for 100 epochs.

342 **Generator and Discriminator of InvNet:** In this section, we provide the training description of InvNet with the multi-fidelity  
 343 surrogate model as the invariance constraint evaluator. Since the main modification that we've proposed occurs in the invariance  
 344 constraint, the formulation of InvNet's loss function remains as

$$L_{InvNet} = L_G(\theta, \psi) + L_I(\theta) \quad (11)$$

345 where  $L_G$  denotes the standard loss function of the WGAN, with  $\theta$  being the parameters of the generator,  $\psi$  being the  
 346 parameters of the discriminator. Both the generator and discriminator are also represented using deep neural networks. The  
 347 invariance loss  $L_I$  is expressed as:

$$L_I = \|R_{MF_{J_{sc}}}(G_\theta(z)) - R_{MF_{J_{sc}}}(I)\|_2^2 + \|R_{MF_{FF}}(G_\theta(z)) - R_{MF_{FF}}(I)\|_2^2 \quad (12)$$

348 with  $G_\theta$  denoting the generator,  $z$  denoting a latent vector sampled from a uniform distribution,  $G_\theta(z)$ , denoting the image of  
 349 generated morphology and  $I$  denoting a real morphology sampled from the dataset. During training, the weights of the surrogate  
 350 physics model,  $\phi$  are kept frozen, and  $R_{MF}$  acts purely as an invariance constraint evaluator that estimates the morphological  
 351 properties of the generated microstructures. Only the parameters of the discriminator  $\psi$ , and generator  $\theta$  are optimized.

352 To train the InvNet, we instantiate the generator with an architecture that consists of one dense layer, five residual blocks  
 353 with skipped connections, and one convolution layer. Each residual block is made up of two batch-normalization layers and  
 354 two convolution layers with up-sampling operations. ReLU activation functions were used after every layer, except for the  
 355 last convolution layer. We used the sigmoid activation function on the output of the convolution layer to generate 128 x 128  
 356 images of microstructures. The Discriminator network consists of one convolution layer, four residual blocks, and a dense  
 357 layer. The residual blocks are similar to the blocks used in the Generator, with the exception that the convolution layers are  
 358 paired with down-sampling operations and layer-normalization is used instead of batch-normalization. As we've chosen to use  
 359 the WGAN-GP<sup>36</sup> variant of GAN, the output of the discriminator is a single scalar value estimating the Wasserstein distance  
 360 between the distributions of generated and real microstructures. To compute the invariance loss, we use the multi-fidelity  
 361 surrogate model  $R_{MF}$  to ensure that generated morphologies had properties that are similar to the properties of the real  
 362 morphologies. Both the Generator and Discriminator are trained alternatively using Adam optimizer with a learning rate of  
 363 3E-4 for 1E5 epochs. We include specific details of network layers we used in the generator, discriminator and multi-fidelity  
 364 network in the Supplementary Materials. Note that in the methodology presented, we have described the InvNet framework  
 365 using the multi-fidelity surrogate,  $R_{MF}$  as the invariance constraint evaluator. We highlight that the methodology for training  
 366 the framework using high-fidelity network is exactly the same, with only  $R_{HF}$  replacing  $R_{MF}$ .

## References

1. Anthony, J. E. Addressing challenges. *Nat. materials* **13**, 773–775 (2014).
2. Yao, H.-B. *et al.* A flexible and highly pressure-sensitive graphene–polyurethane sponge based on fractured microstructure design. *Adv. Mater.* **25**, 6692–6698 (2013).
3. Du, P., Zebrowski, A., Zola, J., Ganapathysubramanian, B. & Wodo, O. Microstructure design using graphs. *npj Comput. Mater.* **4**, 1–7 (2018).
4. Noruzi, R., Ghadai, S., Bingol, O. R., Krishnamurthy, A. & Ganapathysubramanian, B. Nurbs-based microstructure design for organic photovoltaics. *Comput. Des.* **118**, 102771 (2020).
5. Pokuri, B. S. S., Ghosal, S., Kokate, A., Sarkar, S. & Ganapathysubramanian, B. Interpretable deep learning for guided microstructure-property explorations in photovoltaics. *npj Comput. Mater.* **5**, 1–11 (2019).
6. Stoecklein, D., Lore, K. G., Davies, M., Sarkar, S. & Ganapathysubramanian, B. Deep learning for flow sculpting: Insights into efficient learning using scientific simulation data. *Sci. reports* **7**, 46368 (2017).
7. Lore, K. G., Stoecklein, D., Davies, M., Ganapathysubramanian, B. & Sarkar, S. A deep learning framework for causal shape transformation. *Neural Networks* **98**, 305–317 (2018).
8. Bock, F. E. *et al.* A review of the application of machine learning and data mining approaches in continuum materials mechanics. *Front. Mater.* **6**, 110 (2019).
9. Lee, X. Y., Balu, A., Stoecklein, D., Ganapathysubramanian, B. & Sarkar, S. A case study of deep reinforcement learning for engineering design: Application to microfluidic devices for flow sculpting. *J. Mech. Des.* **141** (2019).
10. Balu, A. *et al.* A deep learning framework for design and analysis of surgical bioprosthetic heart valves. *Sci. reports* **9**, 1–12 (2019).
11. Geng, Y., van Anders, G., Dodd, P. M., Dshemuchadse, J. & Glotzer, S. C. Engineering entropy for the inverse design of colloidal crystals from hard shapes. *Sci. advances* **5**, eaaw0514 (2019).
12. Kim, B., Lee, S. & Kim, J. Inverse design of porous materials using artificial neural networks. *Sci. advances* **6**, eaax9324 (2020).
13. Goodfellow, I. *et al.* Generative adversarial nets. In *Advances in neural information processing systems*, 2672–2680 (2014).
14. Yang, L., Zhang, D. & Karniadakis, G. E. Physics-informed generative adversarial networks for stochastic differential equations. *SIAM J. on Sci. Comput.* **42**, A292–A317 (2020).
15. de Oliveira, L., Paganini, M. & Nachman, B. Learning particle physics by example: location-aware generative adversarial networks for physics synthesis. *Comput. Softw. for Big Sci.* **1**, 4 (2017).
16. Wu, J.-L. *et al.* Enforcing statistical constraints in generative adversarial networks for modeling chaotic dynamical systems. *J. Comput. Phys.* **406**, 109209 (2020).
17. Yang, Z. *et al.* Microstructural materials design via deep adversarial learning methodology. *J. Mech. Des.* **140** (2018).
18. Sanchez-Lengeling, B. & Aspuru-Guzik, A. Inverse molecular design using machine learning: Generative models for matter engineering. *Science* **361**, 360–365 (2018).
19. Prykhodko, O. *et al.* A de novo molecular generation method using latent vector based generative adversarial network. *J. Cheminformatics* **11**, 74 (2019).
20. Fokina, D., Muravleva, E., Ovchinnikov, G. & Oseledets, I. Microstructure synthesis using style-based generative adversarial networks. *Phys. Rev. E* **101**, 043308, DOI: [10.1103/PhysRevE.101.043308](https://doi.org/10.1103/PhysRevE.101.043308) (2020).
21. Gayon-Lombardo, A., Mosser, L., Brandon, N. P. & Cooper, S. J. Pores for thought: generative adversarial networks for stochastic reconstruction of 3d multi-phase electrode microstructures with periodic boundaries. *npj Comput. Mater.* **6**, 1–11 (2020).
22. Joshi, A. *et al.* Invnet: Encoding geometric and statistical invariances in deep generative models. In *AAAI 2020* (2020).
23. Yu, G., Gao, J., Hummelen, J. C., Wudl, F. & Heeger, A. J. Polymer photovoltaic cells: enhanced efficiencies via a network of internal donor-acceptor heterojunctions. *Science* **270**, 1789–1791 (1995).
24. Shaheen, S. E. *et al.* 2.5% efficient organic plastic solar cells. *Appl. Phys. Lett.* **78**, 841–843 (2001).
25. Guerrero, A. & Garcia-Belmonte, G. Recent advances to understand morphology stability of organic photovoltaics. *Nano-micro letters* **9**, 10 (2017).

- 415 **26.** Kodali, H. K. & Ganapathysubramanian, B. Computer simulation of heterogeneous polymer photovoltaic devices. *Model.*  
416 *Simul. Mater. Sci. Eng.* **20**, 035015 (2012).
- 417 **27.** Arjovsky, M., Chintala, S. & Bottou, L. Wasserstein generative adversarial networks. In *Proceedings of the 34th*  
418 *International Conference on Machine Learning-Volume 70*, 214–223 (2017).
- 419 **28.** Csáji, B. C. *et al.* Approximation with artificial neural networks. *Fac. Sci. Eötvös Loránd Univ. Hungary* **24**, 7 (2001).
- 420 **29.** Sarkar, S. *et al.* Multifidelity and multiscale bayesian framework for high-dimensional engineering design and calibration.  
421 *J. Mech. Des.* **141** (2019).
- 422 **30.** Costabal, F. S., Perdikaris, P., Kuhl, E. & Hurtado, D. E. Multi-fidelity classification using gaussian processes: accelerating  
423 the prediction of large-scale computational models. *Comput. Methods Appl. Mech. Eng.* **357**, 112602 (2019).
- 424 **31.** Babae, H., Bastidas, C., DeFilippo, M., Chryssostomidis, C. & Karniadakis, G. A multifidelity framework and uncertainty  
425 quantification for sea surface temperature in the massachusetts and cape cod bays. *Earth Space Sci.* **7**, e2019EA000954  
426 (2020).
- 427 **32.** Yang, C.-H. *et al.* Multi-fidelity machine learning models for structure-property mapping (2020). ArXiv.
- 428 **33.** Wodo, O., Zola, J., Pokuri, B. S. S., Du, P. & Ganapathysubramanian, B. Automated, high throughput exploration of  
429 process–structure–property relationships using the mapreduce paradigm. *Mater. discovery* **1**, 21–28 (2015).
- 430 **34.** Wodo, O., Tirhappura, S., Chaudhary, S. & Ganapathysubramanian, B. Computational characterization of bulk heterojunc-  
431 tion nanomorphology. *J. Appl. Phys.* **112**, 064316 (2012).
- 432 **35.** Cahn, J. W. & Hilliard, J. E. Free energy of a nonuniform system. I. interfacial free energy. *The J. chemical physics* **28**,  
433 258–267 (1958).
- 434 **36.** Gulrajani, I., Ahmed, F., Arjovsky, M., Dumoulin, V. & Courville, A. C. Improved training of wasserstein gans. In *Adv.*  
435 *Neural Inf. Proc. Sys. (NeurIPS)*, 5767–5777 (2017).

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### 437 **General**

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### 443 **Author contributions**

444 CH, BG, SS initiated the project; BP, BG planned and generated dataset. XYL, AJ, CH, BG, SS designed the ML framework;  
445 XYL, JW, CH-Y performed the training; XYL, AJ and AB analyzed the data; all authors contributed to writing the manuscript.

### 446 **Competing interests**

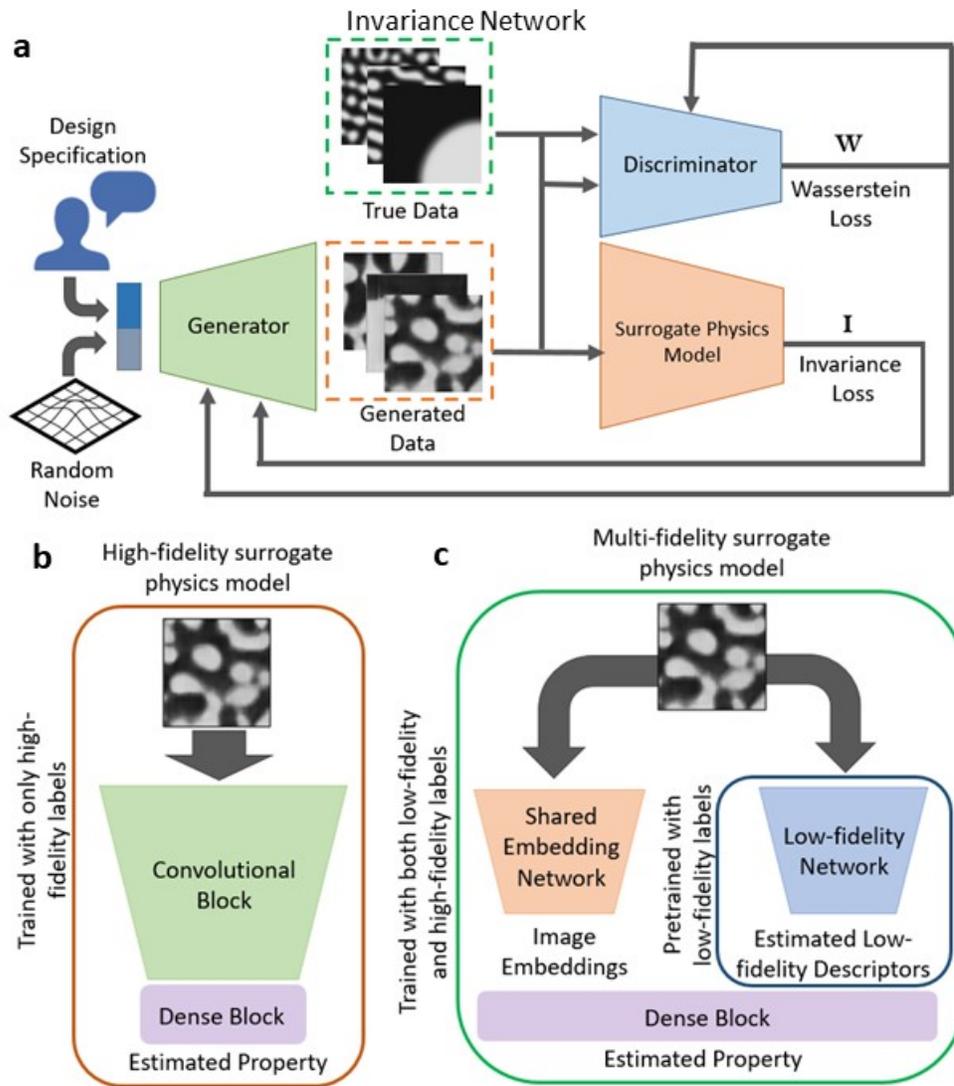
447 The authors declare no competing interests.

### 448 **Data and materials availability**

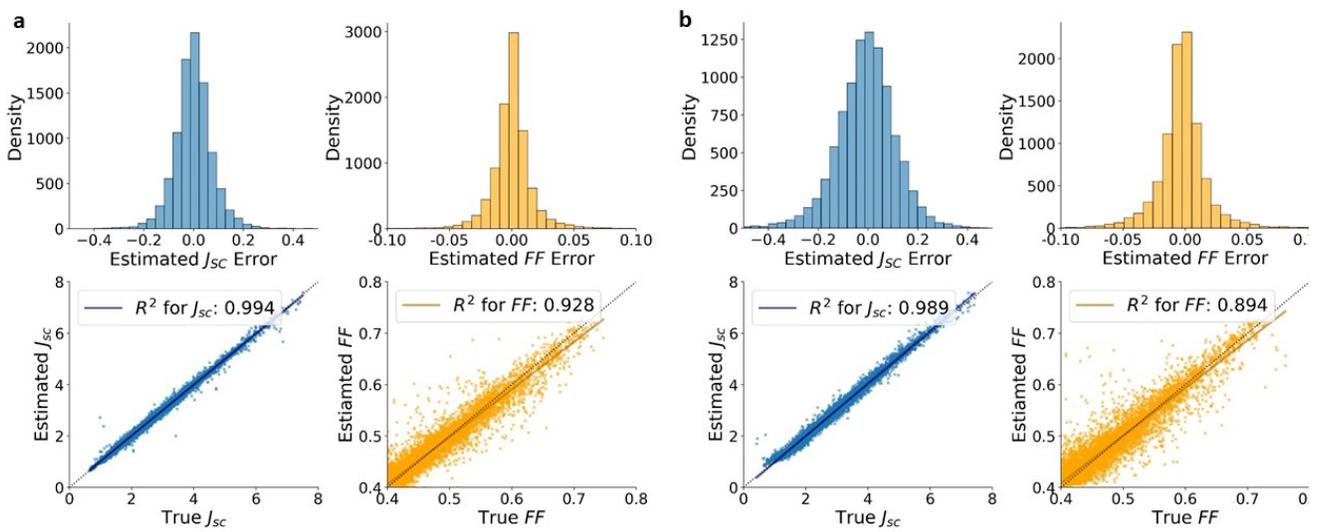
449 The datasets generated and/or analysed during the current study will be available upon acceptance of the paper.

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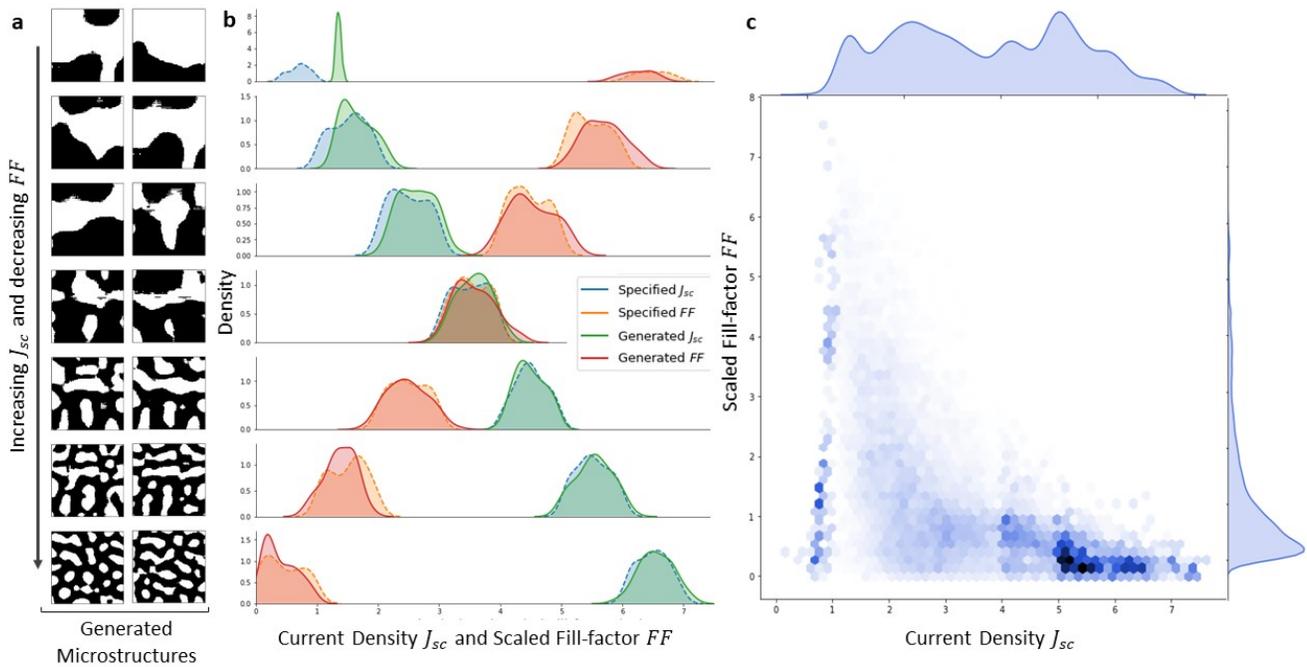
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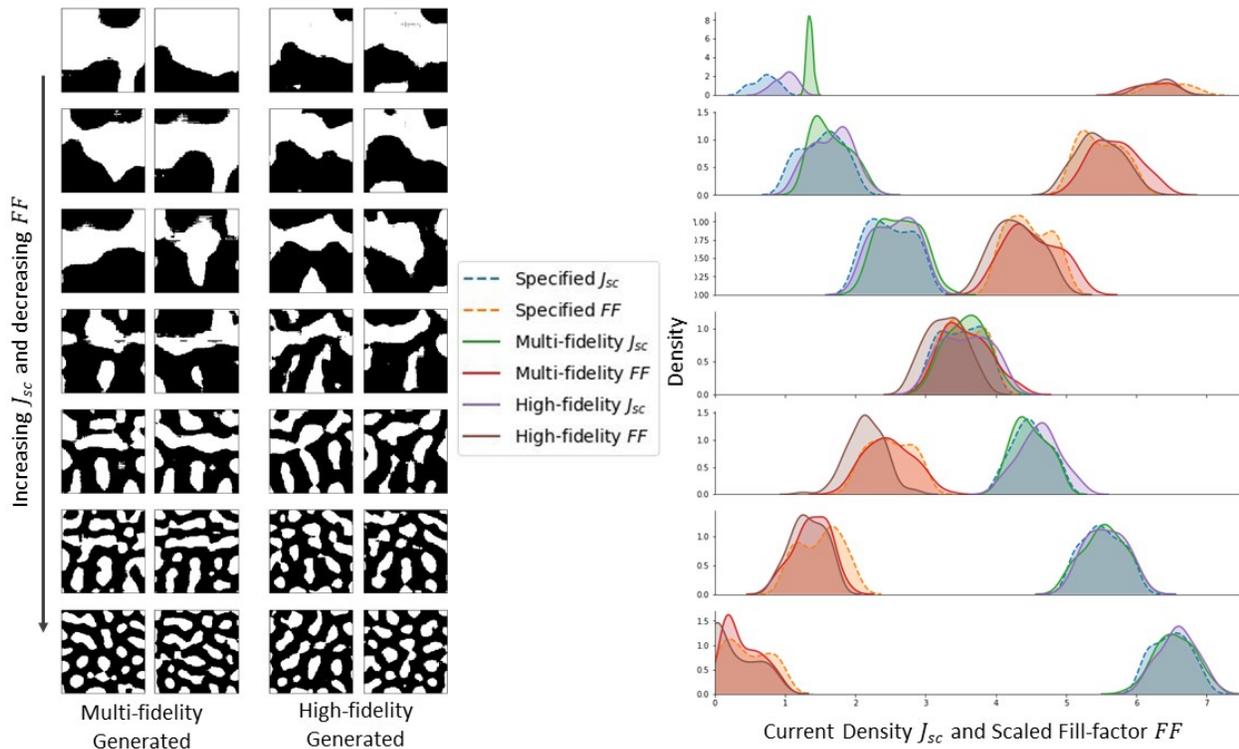
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**Figure 2. Results of high-fidelity and multi-fidelity surrogate models.** (a) Left figures summarize the distribution of errors for both  $J_{sc}$  and  $FF$  estimation using the high-fidelity surrogate physics model. Bottom plots visualize the correlation plot of the estimated properties with respect to the ground truth values. In both cases, the predicted values have high correlation coefficients,  $R^2$  values of greater than 0.9. (b) Summary of error distributions for  $J_{sc}$  and  $FF$  estimation using the multi-fidelity surrogate model which was trained with only 20% of high-fidelity labels. We observe that while there is slight drop in  $R^2$  and increase in variance, there is a huge marginal gain in terms of decreasing the amount of expensive simulations required to generate the high-fidelity labels.



**Figure 3. Results of targeted microstructure design using multi-fidelity InvNet.** (a) Examples of morphologies generated by InvNet for the specified  $J_{sc}$  and  $FF$  ranges shown on the right densities. (b) Densities of estimated  $J_{sc}$  and  $FF$  from generated morphologies compared with a range of respective design specifications for 1000 samples. Observe that the densities of the design specifications and generated morphologies properties in the mid- and high-ranges (rows 2 to 7) are highly overlapping, signifying that the invariances are satisfied. In contrast, the densities at the region of low  $J_{sc}$  are more deviated, signifying a more biased model at the region where the training data is sparse. (c) Visualization of joint and marginal densities of training data for both  $J_{sc}$  and  $FF$ . Notice that the marginal density of  $J_{sc}$  labels is relatively well balanced, while the marginal density of  $FF$  is extremely skewed, resulting in sparser data around certain regions.



**Figure 4. Qualitative comparison of morphologies generated by the high-fidelity InvNet vs multi-fidelity InvNet.**

Visually, we observe that both models are capable of generating varying morphologies which follows a similar trend as we varied the design specifications. Looking at the densities of property invariances, we observe that the high-fidelity InvNet performs slightly better than multi-fidelity InvNet by generating morphologies which are closer to design specifications in the low  $J_{sc}$  high  $FF$  regions where training data is sparse. However, the high-fidelity InvNet also tend to generate morphologies which are slightly biased in terms of the  $FF$ , as observed in rows 3, 4 and 5.

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485 **for different processes.**  $J_{sc}$  and  $FF$  columns denotes the time required to evaluate the corresponding properties  
486 given a morphology. InvNet training times are based on our training scheme of 1E5 epochs. \*Physics model-  
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488 physics simulation. Morphology Generation column denotes the time required for a trained InvNet to generate  
489 a single morphology given design specification values of  $J_{sc}$  and  $FF$ . . . . . 17

	$J_{sc}$ Evaluation	$FF$ Evaluation	InvNet Training	Morphology Generation
High-Fidelity	5.9 ms	5.9 ms	5.8 hr	191.0 ms
Multi-Fidelity	55.3 ms	55.3 ms	18.7 hr	192.0 ms
Physics-Model	9.0 min	72.0 min	60,017.0 hr*	N/A

**Table 1. Comparison of average computation times of neural network-based methods vs physics-based methods for different processes.**  $J_{sc}$  and  $FF$  columns denotes the time required to evaluate the corresponding properties given a morphology. InvNet training times are based on our training scheme of  $1E5$  epochs. \*Physics model-based InvNet training is based on an estimate if the invariance loss were to be computed using high-fidelity physics simulation. Morphology Generation column denotes the time required for a trained InvNet to generate a single morphology given design specification values of  $J_{sc}$  and  $FF$ .