

A QMC-Deep Learning Method for Diffusivity Estimation in Random Domains

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Abstract. Exciton diffusion plays a vital role in the function of many organic semi-conducting opto-electronic devices, where an accurate description requires precise control of heterojunctions. This poses a challenging problem because the parameterization of heterojunctions in high-dimensional random space is far beyond the capability of classical simulation tools. Here, we develop a novel method based on Quasi-Monte Carlo sampling to generate the training data set and deep neural network to extract a function for exciton diffusion length on surface roughness with high accuracy and unprecedented efficiency, yielding an abundance of information over the entire parameter space. Our method provides a new strategy to analyze the impact of interfacial ordering on exciton diffusion and is expected to assist experimental design with tailored opto-electronic functionalities.

AMS subject classifications: 35K57, 65C05, 65M06, 65M32

Key words: Exciton diffusion length, deep learning, Quasi-Monte Carlo sampling, diffusion equation.

1. Introduction

Over the past decades, much attention has been paid on organic semiconductors for applications in various opto-electronic devices [16, 44, 51, 63]. These materials include small molecules [33, 50], oligomers [43, 56], and polymers [39, 49]. Exciton diffusion is one of the key processes behind the operation of organic opto-electronic devices [4, 5, 14]. From a microscopic perspective, exciton, a bound electron-hole pair,

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is the elementary excitation in opto-electronic devices such as light emitting diodes and organic solar cells. The exciton diffusion length (EDL) is the characteristic distance that excitons are able to travel during their lifetime [33]. A short diffusion length in organic photovoltaics limits the dissociation of excitons into free charge [38, 65]. Conversely, a large diffusion length in organic light emitting diodes may limit luminous efficiency if excitons diffuse to non-radiative quenching sites [2].

As quasi-particles with no net charge, excitons are difficult to probe directly by electrical means [41]. This is particularly true in organic semiconductors where the exciton binding energy is 1 electronvolt [64]. Reported techniques to measure EDL include photoluminescence (PL) surface quenching [19, 33, 36, 54, 58, 60, 65, 66, 70], time-resolved PL bulk quenching modeled with a Monte Carlo (MC) simulation [39, 40], exciton-exciton annihilation [12, 31, 37, 59], modeling of solar cell photocurrent spectrum [6, 18, 22, 27, 49, 50, 52, 55, 62, 66, 67, 74], time-resolved microwave conductance [17, 30], spectrally resolved PL quenching [3, 34, 53] and Förster resonance energy transfer theory [34, 35, 42]. From a theoretical perspective, the minimal modeling error is given by the diffusion equation model [9], which is employed in the current work.

To be precise, the device used in PL surface quenching experiment includes two layers of organic materials with thickness ranging from dozens of nanometers to hundreds of nanometers. One layer of material is called donor and the other is called acceptor or quencher according to the difference of their chemical properties. Under the illumination of solar lights, excitons are generated in the donor layer and diffuse in the donor. Due to the exciton-environment interaction, some excitons die out and emit photons which lead to the PL. The donor-acceptor interface serves as the absorbing boundary while other boundaries serve as reflecting boundaries due to the tailored properties. Since the donor-acceptor interface is not exposed to the air/vacuum and the resolution of the surface morphology is limited by the resolution of atomic force microscopy, the interface is subject to an uncertainty. It is found that the fitted EDL is sensitive to the uncertainty in some scenarios. From a numerical perspective, the random interface requires a parametrization in high-dimensional random space, which is prohibitively expensive for any simulation tool. For example, MC method overcomes the curse of dimensionality but has very low accuracy [26]. Stochastic collection method has high accuracy but is only affordable in low dimensional random space [72]. Asymptotics-based method is efficient but its accuracy relies heavily on the magnitude of randomness [10]. In the current work, we propose a novel method based on deep learning with high accuracy and unprecedented efficiency.

Recently, increasing attentions have been paid to apply machine learning (ML) techniques to materials-related problems. For example, the classification of crystal structures of transition metal phosphide via support vector machine [48] leads to the discovery of a novel phase [47]. Likewise, a hybrid probabilistic model based on high-throughput first-principle computation and ML was developed to identify stable novel compositions and their crystal structures [24]. Physical parameters such as band gap [28, 71], elastic constants [13, 28], and Debye temperature [28] have also been