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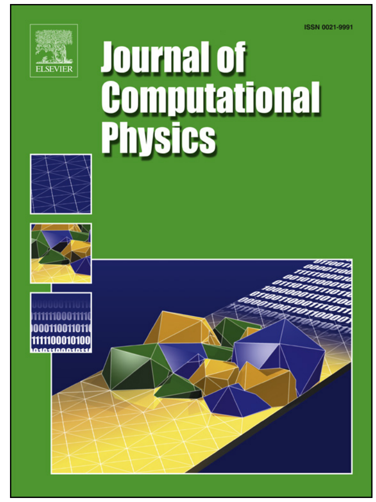

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Functional Level-Set Derivative for a Polymer Self Consistent Field Theory Hamiltonian

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Abstract

We derive functional level-set derivatives for the Hamiltonian arising in self-consistent field theory, which are required to solve free boundary problems in the self-assembly of polymeric systems such as block copolymer melts. In particular, we consider Dirichlet, Neumann and Robin boundary conditions. We provide numerical examples that illustrate how these shape derivatives can be used to find equilibrium and metastable structures of block copolymer melts with a free surface in both two and three spatial dimensions.

1. Introduction

The use of polymers in confined domains is ubiquitous in science and engineering, ranging from novel lithography techniques at the sub 22-nanometer scale for next generation computer chips to the study of drug delivery carriers with controlled release to the study of thin films [15, 17, 16, 18, 36, 35, 14, 26]. Self-consistent field theory (SCFT) provides an accurate description of the self-assembly of dense collections of long polymers at equilibrium by considering a Fokker-Planck equation that describes the probability of polymer segments to be at a certain spatial location and given chain contour length. This technique has been successfully used in the case of periodic as well as confined domains. The more difficult case of free boundaries is still in its infancy, although some authors have recently provided specialized solutions in the context of directed self-assembly, by considering a parametric description of the free boundary [20, 19]. An important question in the context of free surfaces is how the self-assembled solution responds to a change in the free boundary's *shape*. Answering this question requires to either explicitly solve the direct problem several times in order to *a posteriori* estimate the sensitivity or to derive appropriate functional derivatives with respect to the level set to *a priori* estimate it. The advantage of an *a priori* approach is that it enables the development and implementation of algorithms that optimize the shape.

Shape derivatives have been derived for many systems described by partial differential equations with various boundary conditions; a rigorous derivation of the methods and examples can be found in the seminal book of Sokolowski and Zolesio [39] and the more recent book of Delfour and Zolesio [8]. The level-set framework introduced by Osher and Sethian [28] which uses a scalar function to implicitly represent any arbitrary shape, has been implemented to study shape optimization for a variety of physical applications such as structural optimization, tomography and inverse obstacle problems in fluids, among others [29, 3, 1, 41, 5, 6].

On the other hand, functional shape derivatives in the context of SCFT have only been derived for orthorhombic shapes [2]. In the present, we derive functional shape derivatives with arbitrary shapes using the level-set description recently proposed by Ouaknin *et al.* [31, 30]. In [31], the authors showed that the use of Neumann boundary conditions, as modeled by deGennes [7], is needed in order to robustly evaluate physical quantities at the free boundary. In [30], they use that framework in the context of directed self-assembly to propose an algorithm that will find the geometry of a mask that will guide the self-assembly of

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