

# Modeling Line Edge Roughness in Lamellar Block Copolymer Systems

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## ABSTRACT

Block copolymers offer an appealing alternative to current lithographic techniques with regard to fabrication of the next generation microprocessors. However, if copolymers are to be useful on an industrial manufacturing scale, they must meet or exceed lithography specifications for placement and line edge roughness (LER) of resist features. Here we discuss a field theoretic approach to modeling the LER of lamellar microdomain interfaces in a strongly segregated block copolymer system; specifically, we derive a formula for the LER as a function of the Flory Huggins parameter  $\chi$  and the index of polymerization  $\mathcal{N}$ . Our model is based on the Leibler-Ohta-Kawasaki energy functional. We consider a system with a finite number of phase separated microdomains and also show how the LER depends on distance of the microdomain interface from the system boundary. Our results suggest that in order to meet target LER goals at the 15 nm, 11 nm, and 6 nm nodes,  $\chi$  must be increased by a factor of at least 5 above currently attainable values.

## 1. INTRODUCTION

Self-assembling block copolymers have received considerable attention in recent years in the hope that they can augment state-of-the-art lithographic techniques as semiconductor features approach the 15 nm, 11 nm, and 6 nm nodes.<sup>1-4</sup> In particular, template directed self assembly (TDSA) – the spontaneous formation of polymer micro-domains between lithographically etched boundaries – is being studied as a way to decrease the domain spacing of semiconductor features without increasing the resolution of current lithography tools.<sup>5-8</sup> A critical task in assessing the usefulness of block-copolymers is therefore to characterize the fundamental limitations on the roughness of the patterns that polymers form. If the roughness cannot be controlled sufficiently well, devices fabricated using TDSA will have high soft-defect densities, precluding the use of block copolymers in many semiconductor manufacturing settings.

Our goal in this paper is to *analytically* predict the line edge roughness (LER) of block copolymer microdomain interfaces as a function of the Flory-Huggins parameter  $\chi$  and the index of polymerization  $\mathcal{N}$ ; in the present analysis, we focus on lamellar systems in the strong segregation regime (SSR). Similar tasks have been pursued by, e.g. Semenov,<sup>9</sup> Detcheverry and de Pablo,<sup>10-12</sup> and Bosse;<sup>13,14</sup> however, to the best of our knowledge, these treatments either (i) relied heavily on numerical methods and computer simulations (which we do not use here), or (ii) considered geometries and physical parameters corresponding to systems in a weak segregation regime. With this work, we aim to supply formulas that predict what values of  $\chi$  and  $\mathcal{N}$  are needed to bring the LER within acceptable levels for systems that are of direct interest to the semiconductor industry.

The starting point of our analysis will be the phase field model originally developed by Leibler, Ohta, and Kawasaki (LOK),<sup>15,16</sup> which gives the energy  $\mathcal{H}[\phi]$  of a polymer melt as a functional of the relative density  $\phi$  of the monomer species. In their original work,<sup>16</sup> Ohta and Kawasaki determined that the lowest-energy configuration (for equal molecular weights of the two monomer components) was indeed a lamellar phase-separated system (but with zero LER). Our main task will be to determine the fluctuation eigenmodes of the system about that mean-field configuration. We then construct the total LER as a weighted sum of the LERs associated with each eigenmode, where  $P[\psi]$  is the weighting function given by the Maxwell-Boltzmann distribution,

$$P[\psi] \propto e^{-\mathcal{H}[\psi]/k_B T}, \quad (1)$$

and  $\psi$  is a fluctuation eigenmode.

Our choice of model is driven in large part by consideration of the features that we wish to describe. On a microscopic scale, individual polymers exhibit complicated geometries and foldings, which are adequately described by Gaussian chain models, molecular dynamics, and Monte Carlo simulations. However, for polymer melts whose

domains span tens of nanometers, the computational expense of using such models becomes insurmountable, owing simply to the number of particles that must be taken into account. Moreover, at such length scales, one typically wishes to study mesoscopic features (such as the LER) of the melt as a whole, as opposed to the structure of individual molecules. Phase field models therefore enter as computationally tractable alternatives that (i) permit study of quantities relevant at the nanometer length scale, while (ii) coarse-graining microscopic features that otherwise render computation difficult. In our present analysis, we use the LOK phase field model, since recent studies suggest that it could be well suited to describe fluctuations in technologically relevant systems.<sup>13,14</sup> We note, however, that while this model is amenable to analytic computation, it is nonetheless complicated enough that we do not find exact expressions for the LER; rather, we arrive at our final results by means of perturbation theory and a series of asymptotic approximations that become increasingly accurate as  $\chi$  and/or  $\mathcal{N}$  increase, i.e. as the system parameters move further into the SSR.

The rest of the paper is organized as follows. In Section 2 we review the basic principles of the LOK model and derive the energy functional describing fluctuations. In Section 3 we use perturbation theory to approximately diagonalize the energy functional. In Section 4 we define and calculate the LER. In Section 5 we discuss our main results in the context of the requirements set forth by the International Technology Roadmap for Semiconductors, and Section 6 we briefly summarize our work.

We end this section by summarizing notation that will be used throughout the remainder of the text:

- i)  $a$  is the Kuhn statistical length, which measures the average distance between two adjacent monomers. This length is considered small relative to the system size.
- ii)  $\chi$  denotes the dimensionless Flory-Huggins parameter, which characterizes the repulsion between different monomer species.
- iii)  $\mathcal{N}$  denotes the index of polymerization, the number of monomers in a single chain.
- iv)  $f$  is the (normalized) molecular weight of A monomers;  $(1 - f)$  is the molecular weight of B monomers.
- v)  $\phi(\mathbf{x}) = \phi_A(\mathbf{x}) - \phi_B(\mathbf{x})$  denotes the relative density of A monomers  $[\phi_A(\mathbf{x})]$  and B monomers  $[\phi_B(\mathbf{x})]$ . We choose the normalization  $0 \leq \phi_A(\mathbf{x}), \phi_B(\mathbf{x}) \leq 1$  and impose the incompressibility condition  $\phi_A(\mathbf{x}) + \phi_B(\mathbf{x}) = 1$ , so that  $-1 \leq \phi \leq 1$ .
- v)  $\Omega$  is the physical domain of the polymer melt, whose volume will be denoted  $|\Omega|$ . The symbol  $\mathcal{V}$  will denote a unit volume.
- vi) Informally, the expression  $g = \mathcal{O}(\delta)$  means that  $g$  is roughly the same size as  $\delta$ .\*
- vii) Unless otherwise noted, italicized variables will represent quantities having dimensions, whereas non-italicized versions of the same variables will be dimensionless. For example, if  $x$  represents a length in some units (e.g. nm), the variable  $x$  will be a rescaled, dimensionless version of  $x$ . The scaling of non-italicized variables will always be defined at their first appearance.

## 2. PHYSICAL SYSTEM: GEOMETRY, KEY PARAMETERS, AND ANALYTIC DESCRIPTION OF FLUCTUATIONS

The system we wish to describe is a lamellar, diblock copolymer melt in the SSR (cf. Fig. 1). For simplicity, we take the molecular weights of the A and B subchains to be equal (i.e.  $f = 1/2$ ) and denote  $\ell$  as the mean-field width of A (or B) domains. Since the system is in the SSR, the boundaries (yellow regions in Fig 1) between the A and B domains are small compared to  $\ell$ . The parameter  $h$  denotes the height of the melt, and we assume that the system is infinite in the  $y$  direction.<sup>†</sup> If the polymers are placed between parallel, chemically templated boundaries that are separated by some multiple integer of  $\ell$ , then the polymer microdomains will form rows between the templated boundaries (cf. Fig. 1).

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\*More precisely,  $g = \mathcal{O}(\delta)$  signifies that  $g/\delta$  is bounded from above by a constant as  $\delta \rightarrow 0$ .

<sup>†</sup>Our analysis is trivially generalized to systems that are finite in the  $y$  direction by converting the appropriate integrals to sums. In Section 3 we indicate where these changes should be made.

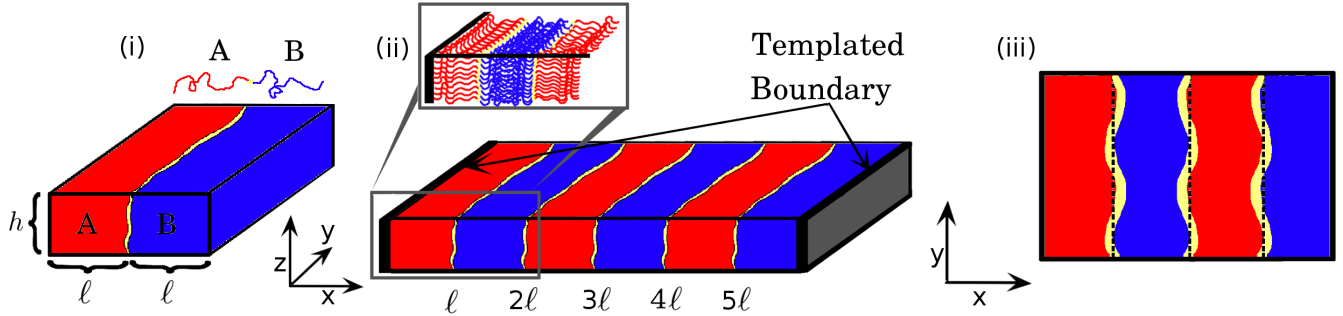


Figure 1. [Color online] Three views of a polymer melt in the lamellar phase. Inset (i) shows a single block copolymer with A (red or light gray) and B (blue or dark gray) components, alongside two microdomains comprising a full pitch; a boundary (yellow) separates regions of different monomer species (red and blue). We assume that the microdomains extend to  $\pm\infty$  in the  $y$  direction.<sup>†</sup> Inset (ii) shows a system with three full pitches. Boundaries are located at integer values of  $\ell$ . Inset (iii) shows a top down view of a system with different types of fluctuating boundaries. The black dotted lines indicate the mean field positions of the phase boundaries. All boundaries exhibit displacement fluctuations corresponding to  $f_1$  [cf. Eq. (11)].

We describe this system with the Leibler-Ohta-Kawasaki energy functional, which takes the general form

$$\mathcal{H}[\phi] = \frac{k_B T \chi}{\nu} \int dV \left\{ \frac{\xi^2}{2} (\nabla \phi)^2 - \frac{\phi^2}{2} + \frac{\phi^4}{4} + \frac{\varsigma}{2} \int_{\Omega} dV' \phi(\mathbf{r}) g(\mathbf{r}, \mathbf{r}') \phi(\mathbf{r}') \right\}, \quad (2)$$

$$\xi^2 := \frac{a^2}{3f(1-f)\chi} \quad \varsigma := \frac{36}{f^2(1-f)^2 a^2 \chi \mathcal{N}^2}. \quad (3)$$

The product  $k_B T$  is Boltzmann's constant times the temperature, and the function  $g(\mathbf{r}, \mathbf{r}')$  is the Green's function of the Laplacian. We assume that the normal derivatives of  $\phi$  and  $g$  vanish at the  $x$  boundary, but we impose periodic boundary conditions on both functions in the  $y$  and  $z$  directions.

Equation (2) is an *effective* field theory that views the melt from a coarse-grained perspective. Individual polymers are not considered; rather the configuration of the system is represented by the relative density  $\phi$  of A and B polymers, which is a continuous function of position. Interfaces between A and B domains will therefore be represented by *boundary layers* (cf. Fig. 2), i.e. narrow regions in which the density  $\phi$  changes rapidly. The parameter  $\xi$  determines the boundary layer thickness and is generally considered small in the sense  $\xi \ll \ell$  whenever  $\chi \mathcal{N} \gg 1$ , which corresponds to the system being in the SSR. We note in passing that  $\chi = \mathcal{O}(1)$  is considered a large value of the Flory-Huggins parameter.

The lowest energy polymer density  $\phi_0$  may be found by taking a variational derivative of Eq. (2) with respect to  $\phi$  and setting the resulting first variation equal to zero. This procedure yields a complicated integro-differential equation for  $\phi_0$ ; solutions to this equation were originally pursued by Ohta and Kawasaki, who determined to good approximation that,<sup>16</sup>

$$\begin{aligned} \phi_0 &= -\sum_{j=1}^N (-1)^j \tanh \left[ \frac{x - j\ell}{\sqrt{2}\xi} \right], & N \text{ odd} \\ \phi_0 &= 1 + \sum_{j=1}^N (-1)^j \tanh \left[ \frac{x - j\ell}{\sqrt{2}\xi} \right] & N \text{ even} \end{aligned} \quad (4)$$

where  $N$  (distinct from  $\mathcal{N}$ , the index of polymerization) is the number of interfaces. The form of  $\phi_0$  given above is essentially a square wave oscillating between  $\pm 1$ ; physically, this solution corresponds to a lamellar phase-separated system with perfectly straight interfaces having a width  $\xi$  (cf. Fig. 2). The domain spacing  $\ell$  was originally determined<sup>16</sup> by inserting Eq. (4) into Eq. (2) and minimizing the resulting expression with respect to  $\ell$ . This procedure yields  $\ell \propto \mathcal{N}^{2/3} \chi^{1/6}$ .

If  $\psi$  is some perturbation of the mean field density, then we write  $\phi = \phi_0 + \psi$  and expand Eq. (2) to second order in  $\psi$ ; doing so yields,

$$\mathcal{H}[\phi] \approx \mathcal{H}[\phi_0] + \mathcal{H}_1[\psi] + \mathcal{O}(\psi^3) \quad (5)$$

$$\mathcal{H}_1[\psi] := \frac{k_B T \chi}{\mathcal{V}} \int dV \left\{ \frac{\xi^2}{2} (\nabla \psi)^2 - \frac{\psi^2}{2} + \frac{3}{2} \phi_0^2 \psi^2 + \frac{\varsigma}{2} \int dV' \psi(\mathbf{r}) g(\mathbf{r}, \mathbf{r}') \psi(\mathbf{r}') \right\}, \quad (6)$$

where  $\mathcal{H}_1$  is the approximate energy perturbation due to  $\psi$ . Since fluctuations with large energies should have small probabilities [via Eq. (1)], the above approximation is justified for small enough temperatures by noting that the statistics of the system will be dominated by those states whose energies are near that of the ground state.

At this point, our main task should be to diagonalize Eq. (6) in terms of its eigenmodes, which determine the probability of the system expressing a particular configuration. Before continuing, however, we examine the terms in Eq. (6) to gain insight into the types of fluctuations that are actually allowed by our model, especially since we are looking specifically for fluctuations of *interfaces*. The term  $\xi^2 (\nabla \psi)^2$  yields a (small) energy penalty for non-constant fluctuations, while the non-local term multiplying  $\varsigma$  promotes oscillations. The term  $(1/2)(3\phi_0^2 - 1)\psi^2$  yields an energy penalty for non-zero fluctuations *except* in the boundary layers [i.e. when  $|x - \ell n| \leq \mathcal{O}(\xi)$ ], where this term *promotes* fluctuations. This last observation suggests that we look for eigenmodes localized at microdomain interfaces and define the LER in terms of those modes. We pursue this task in the following section.

### 3. DIAGONALIZATION OF $\mathcal{H}_1$

In this section, our goal is to diagonalize Eq. (6) and find the eigenmodes corresponding specifically to interface fluctuations. We begin by non-dimensionalizing space via  $dV \rightarrow d\tilde{V} := dV/\xi^3$ . Equation (6) then becomes

$$\mathcal{H}_1[\psi] := \frac{k_B T \chi \xi^3}{\mathcal{V}} \int d\tilde{V} \left\{ \frac{1}{2} (\tilde{\nabla} \psi)^2 - \frac{\psi^2}{2} + \frac{3}{2} \phi_0^2 \psi^2 + \frac{\xi^2 \varsigma}{2} \int d\tilde{V}' \psi(\tilde{\mathbf{r}}) \tilde{g}(\tilde{\mathbf{r}}, \tilde{\mathbf{r}}') \psi(\tilde{\mathbf{r}}') \right\}, \quad (7)$$

where  $\tilde{g}(\tilde{\mathbf{r}}, \tilde{\mathbf{r}}')$  is the dimensionless Green's function [ $g(\mathbf{r}, \mathbf{r}')$  has units of inverse length]. If  $\varsigma$  is sufficiently small, then to leading order we may diagonalize the energy by solving the eigenvalue problem,

$$-\tilde{\nabla}^2 \psi - (1 + 3\phi_0^2) \psi = E^{(0)} \psi, \quad (8)$$

where  $E^{(0)}$  is a dimensionless, leading-order energy eigenvalue. We may then use standard perturbation techniques to calculate corrections to  $E^{(0)}$  and  $\psi$ . At the end of our analysis, we will determine the values of  $\varsigma$  for which our perturbation theory is valid.

For a system having a single interface, Eq. (8) can be approximately solved if we cast it into the standard form,

$$\psi = \sum_{\mathbf{k}_z} \int \frac{dk_y}{2\pi} f(x) e^{ik_y y + ik_z z} \quad (9)$$

$$0 = \partial_{xx} f + \lambda^2 f + 6 \operatorname{sech}^2(x) f, \quad (10)$$

where  $\lambda^2 = 2E^{(0)} - 2q_{\parallel}^2 - 4$  and  $q_{\parallel}^2 = k_y^2 + k_z^2$  is a wave-vector parallel to the mean field interface profile. The above expression are written in terms of the rescaled variables  $x = x/(\sqrt{2}\xi)$ ,  $y = y/\xi$ , and  $z = z/\xi$ . The wave-vector  $k_y$  is a continuous parameter (in units of  $\xi^{-1}$ ), whereas  $k_z = 2\pi n \xi/h$ ,  $n = 0, \pm 1, \pm 2, \dots$  may only take discrete values.<sup>¶</sup>

<sup>¶</sup>If the system is finite in the  $y$  direction, then Eq. (9) is appropriately modified by changing the integral over  $k_y$  to a sum and allowing  $k_y$  to only take discrete values, in the same manner as  $k_z$ .

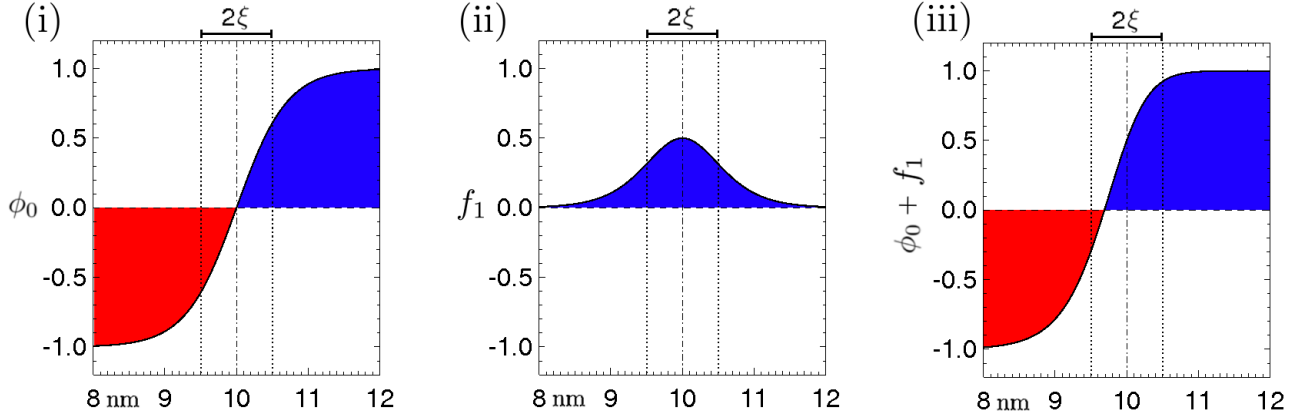


Figure 2. [Color online] The effect of  $f_1$  fluctuation modes on  $\phi_0$ . In inset (i), we plot  $\phi_0$  for a polymer system with a 10 nm half pitch and a single interface (located at 10 nm); we set  $\xi = 0.5$  nm. The yellow region is the boundary layer (or interface) separating A (red or light gray) and B (blue or dark gray) monomer domains. Inset (ii) shows the functions  $f_1$  localized at the interface. Inset (iii) shows that  $f_1$  displaces the interface (i.e. the boundary layer) away from its mean position. As in the previous figures, red and blue shading indicates regions A and B monomer micro-domains, respectively. Fluctuations of the type  $f_1$  determine the LER.

Equation (10) is in fact a version of the well-studied Pöschl-Teller equation used to model diatomic molecules; general solutions can be found in Ref. [18], for example. For the case at hand, there is a single type of interface fluctuation corresponding to the LER.<sup>19</sup> Specifically,

$$f_1(x) = \text{sech}^2(x), \quad E_1^{(0)} = q_{\parallel}^2, \quad (11)$$

where the dimensionless energy  $E^{(0)} = E^{(0)}\mathcal{V}/(k_B T \chi \xi^3)$ . A fluctuation of the form  $\psi = f_1(x)e^{ik_y y + ik_z z}$  corresponds to an oscillation of the interface about its mean position *without* a broadening of the width of the boundary layer itself [see Fig. 1(iii) and Section 4 for more explanation of this point].

For a system with  $N > 1$  interfaces, Eq. (10) takes the approximate form,

$$0 = \partial_{xx} f + \lambda^2 f + 6f \sum_{j=1}^N \text{sech}^2[x - jL], \quad (12)$$

where  $L = \ell/(\sqrt{2}\xi)$ . We subsequently assume that the bound state eigenfunctions  $f_1$  can be written in the form,

$$f_1(x) = \sum_{j=1}^N \Xi_j f_1[x - jL], \quad (13)$$

where  $\Xi_j$  are phase factors chosen to ensure that we have a complete basis of states. Since  $f_1$  asymptotes to zero away from an interface, the solutions that we construct in this way will then solve Eq. (12) up to exponentially small corrections. For the same reason, we may use any orthonormal basis of phase factors  $\Xi_j$  without introducing significant error. The most natural choice is a Kronecker delta function basis,  $\Xi_j = 1$  for  $j = m$  and zero otherwise, where  $j, m = 1, 2, \dots, N$ ; that is

$$f_1(x; m) = f_1[x - mL]. \quad (14)$$

This basis yields  $N$  independent fluctuation modes corresponding to the situations in which each one of the  $N$  interfaces fluctuates independently.

Considering only  $f_1$  fluctuation modes, Eq. (7) is written to leading order in diagonal form as,

$$\mathcal{H}_1 = \frac{\sqrt{2}k_B T h \chi \xi^2}{2\mathcal{V}} \sum_n \int \frac{dk_y}{2\pi} \sum_{m=1}^N \left\{ q_{\parallel}^2 |C_1^{(m)}(q_{\parallel})|^2 \langle f_1^2(x; m) \rangle \right\} \quad (15)$$

where  $\langle f_1^2(x; m) \rangle := \int dx f_1^2(x; m) \approx 2/3$ , and  $C_1^{(m)}(q_{\parallel})$  is the amplitude of a fluctuation of the  $m$ th interface having frequency  $q_{\parallel}$ .

From perturbation theory, it is well known that the first order correction to the energy eigenvalues can be written in general as  $\int dV \Psi^\dagger \hat{H} \Psi$ , where  $\hat{H}$  is some perturbing potential, and  $\Psi$  is an eigenfunction of the leading order problem.<sup>20</sup> In our case, we find,

$$E_1^{(1)}(m, q_{\parallel}) \approx G(m\ell, m\ell) = \frac{\pi\xi}{hq_{\parallel}} \left\{ \frac{\cosh[q_{\parallel}\ell N/\xi] + \cosh[(q_{\parallel}/\xi)(\ell N - 2\ell m)]}{\sinh(\ell N q_{\parallel}/\xi)} - \frac{2\pi\xi}{\ell N q_{\parallel}} \right\}, \quad (16)$$

where  $E_1^{(1)}$  is the first order energy corrections for the  $f_1$  states. In calculating Eq. (16), we approximate  $f_1(x) \approx \sqrt{2\xi}\delta(x - \ell m)$  and use the definition

$$\tilde{g}(\tilde{\mathbf{r}}, \tilde{\mathbf{r}}') = \sum_{m=-\infty}^{\infty} \sum_{k_z} \int \frac{dk_y}{2\pi} \frac{e^{ik_z(z-z') + ik_y(y-y')} \cos(\pi m x/LN) \cos(\pi m x'/LN)}{k_z^2 + k_y^2 + (\pi m/LN)^2}. \quad (17)$$

It is possible to continue perturbation theory indefinitely, computing corrections to the energy eigenfunctions and eigenvalues. We recall, however, that in the multiple interface problem, all  $f_1(m)$  have the same energy for a fixed value of  $m, q_{\parallel}$ . In order to calculate additional corrections to either the eigenmodes or eigenenergies, it is necessary to rewrite the  $f$  in a basis that is orthogonal in the non-local term. Applying the approximation  $f_1(x) \approx \sqrt{2\xi}\delta(x - \ell m)$ , this procedure is achieved by solving the eigenvalue problem

$$\lambda \Xi_i = \sum_j G(\ell i, \ell j) \Xi_j, \quad (18)$$

which determines the values of  $\Xi_j$  appearing in Eq. (13). For our present purposes, it suffices to stop with the first order correction to the eigen-energies.

Combining Eq. (15) with Eqs. (16), we can write the probability density of a given fluctuation as

$$P[\psi] \propto \exp \left\{ \frac{\sqrt{2}h\chi}{2\mathcal{V}} \sum_n \int \frac{dk_y}{2\pi} \sum_{m=1}^N (2/3)[q_{\parallel}^2 + G(m\ell, m\ell)] |C_1^{(m)}(q_{\parallel})|^2 \xi^2 \right\}, \quad (19)$$

where we restrict  $\psi$  to fluctuations containing only  $f_1$  modes.

#### 4. THE LINE EDGE ROUGHNESS AND INTERFACE THICKNESS

In Fig. 2 we show heuristically how the  $f_1$  modes give rise to the line edge roughness. It is also possible to show this analytically by deriving Eq. (6) in a way that manifestly yields  $f_1$  modes as LER fluctuations. Specifically, if we momentarily consider the 1-interface problem, we may write,

$$\phi = \phi_0[x + \xi\zeta(y, z)] \approx \phi_0(x) + \xi\phi_0'(x)\zeta(y, z) + \frac{\xi^2}{2}\phi_0''(x)\zeta(y, z)^2, \quad (20)$$

where  $\xi\zeta(y, z)$  is a small fluctuation in the position of the equilibrium interface and  $\phi_0', \phi_0''$  denote the first and second derivatives of  $\phi_0$ ; substitution of Eq. (20) into Eq. (2) (with  $\varsigma = 0$ ) then yields (note that  $\phi_0' \propto f_1$ )

$$\mathcal{H}[\zeta] \approx \frac{k_B T \chi}{\mathcal{V}} \int dV \left\{ \frac{\xi^2}{2} [(f_1')^2 \zeta^2 + f_1^2 (\nabla_{\parallel} \zeta)^2] - \frac{1}{2} f_1^2 \zeta^2 + \frac{3}{2} \phi_0^2 f_1^2 \zeta^2 \right\}. \quad (21)$$

Equation (21) is in fact just Eq. (6) with  $\psi = f_1 \zeta$ . Comparison to Eq. (9) reveals that Eq. (21) can be diagonalized if  $\zeta$  is written as product of Fourier modes in the  $y$  and  $z$  directions. Therefore, we may conclude that  $f_1$  modes correspond to a *shift in position* of the interface, where  $\xi$  is the physical amplitude of the actual fluctuation. <sup>‡‡</sup>

<sup>‡‡</sup>While this procedure is useful for physically interpreting the  $f_1$  modes, we cannot assume that an expansion having the form of Eq. (20) will yield all of the possible interface fluctuation modes; in fact, the assumptions underlying Eq. (20) completely neglect fluctuations that alter the *width* of the interface itself, which are predicted by the LOK model.

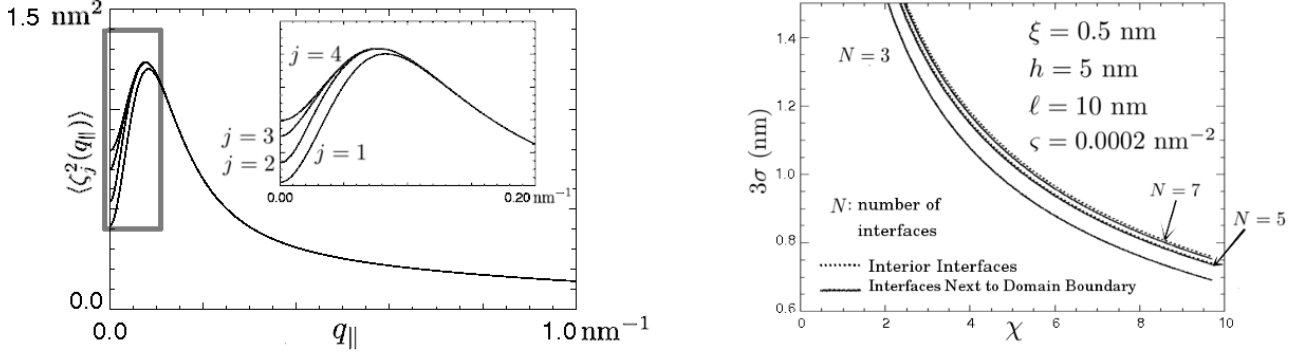


Figure 3. LER in frequency and real space. Inset (i) shows the amplitude  $\langle \zeta_j^2(q_{\parallel}) \rangle$  for a system with 7 interfaces, a 20 nm half-pitch, and an interface thickness  $\xi = 1$  nm. Note that  $\langle \zeta_j^2(q_{\parallel}) \rangle = \langle \zeta_{8-j}^2(q_{\parallel}) \rangle$ . Interfaces closer to the domain boundary in general have a smaller LER than interfaces in the middle of the domain; the figure shows, however, that only the low frequency fluctuations differ significantly among interfaces. Inset (ii) shows values of  $3\sigma$  given by Eq. (23); at the 11 nm node, industrial specifications require that  $3\sigma \leq 1.1$  nm. Our results therefore predict that  $\chi$  must be increased by at least a factor of five above previous limits in order to bring copolymers within reach of industrial specifications. This figure also shows that the *number* of microdomains has a larger impact on LER than the *position* of an interface relative to the system boundary.

This analysis may be extended to the case  $N > 1$  to show that the modes  $f_1(x, m)$  correspond to LER of the  $m$ th interface.

Based on the above analysis, we define the LER per Fourier mode  $q_{\parallel}$  and interface  $m$  as

$$\langle \zeta_m^2(q_{\parallel}) \rangle = \int D[\psi] \xi^2 |C_1^{(m)}(q_{\parallel})|^2 P[\psi] = \frac{\mathcal{V}}{h\chi} \left[ \frac{3}{2\sqrt{2}\xi q_{\parallel}^2 + 6\varsigma hG(\ell m, \ell m)} \right], \quad (22)$$

where  $D[\psi]$  is a functional measure over  $\psi$  and  $\mathcal{V}$  is a unit volume. We may define the LER of the  $m$ th interface in real space by integrating over Eq. (22); specifically,

$$\sigma^2(m) = \sum_n \int \frac{dk_y}{2\pi} \langle \zeta_m^2(q_{\parallel}) \rangle = \sum_n \int \frac{dk_y}{2\pi} \frac{\mathcal{V}}{h\chi} \left[ \frac{3}{2\sqrt{2}\xi q_{\parallel}^2 + 6\varsigma hG(\ell m, \ell m)} \right]. \quad (23)$$

Note that when  $\varsigma = 0$ , Eq. (23) *does not* depend on  $m$ ; this is a direct consequence of Eqs. (16) and (19), which states that each interface fluctuates independently from the others. Physically this makes sense; in the strong segregation regime, we expect polymer fluctuations to depend largely on the local behavior of the (mean-field) interface. It is the non-local term multiplying  $\varsigma$  that induces a position dependence (relative to the system boundary) on the LER. Again, this result is to be expected; following the analogy to electrostatics, the non-local term gives rise to *interactions* between components of the system.

We note that Eq. (23) is log divergent if we allow  $n$  and  $k_y$  to go to infinity. Following Semenov,<sup>9</sup> we define a cutoff frequency such that  $0 \leq q_{\parallel} \leq 2/\xi$ , which renders the integral finite and bounded; this cutoff signifies that the mean field theory breaks down for fluctuation wavelengths that are the same order of magnitude as the interface thickness.

## 5. DISCUSSION

Our goal in this section is to discuss the implications of Eq. (23) in the context of manufacturing specifications set forth by the ITRS. We also discuss key approximations and limitations of our approach.

The ITRS specifies that the LER must satisfy  $3\sigma < 1.1$  nm at the 11 nm node. In Fig. 3 we estimate values of  $\chi$  that will be required to reach these goals. Using the the values  $\mathcal{N} = 300$ ,  $\xi = 1$  nm,  $\ell = 10$  nm,<sup>6</sup> Eq. (23) predicts that values of  $\chi \approx 5$  will be required to reduce the LER to within acceptable limits. These values of  $\chi$  are extremely large relative to typical values seen in systems such as PS-PDMS and PS-PMMA, where typical

values of  $\chi$  range from  $\mathcal{O}(10^{-4})$  to  $\mathcal{O}(10^{-1})$ .<sup>21-25</sup> Although effective values of  $\chi$  as large as  $\mathcal{O}(1)$  have been reported in some systems, our results suggest that a significant increase in  $\chi$  is necessary to reach target goals at the 11 nm node.

Our analysis reveals a connection between the LER and the number and position of microdomains between template boundaries. Figure 3 provides a representative illustration, which shows that, for a fixed value of  $\chi$  and  $\ell$ , decreasing the number of microdomains can reduce the LER by a factor of 20 % or more. This amounts to reducing the distance between templated boundaries. On the other hand, the position of the individual microdomains within the actual system has a much smaller effect on the LER.

While the values of  $\chi$  that we consider here are relatively large, we caution that the analysis herein should be taken more as a qualitative estimate of the necessary system parameters as opposed to a strict, quantitative prediction. As noted earlier, the LOK energy is known to predict behavior seen in many experimental systems. Moreover, the physics of the  $f_1$  fluctuation modes appears naturally through an analysis of the relevant differential equations. However, at the length scales we consider, we begin to push the model to the limits of its validity. This fact is seen by a close examination of the parameters entering the model. Notably, for a physical system in which  $\xi = 1$  nm and  $\ell = 10$  nm, we find that  $a^2 = 3\chi\xi^2/4$ , which is  $\mathcal{O}(1)$  nm<sup>2</sup> for  $\chi = 1$ . Thus, for large enough values of the Flory-Huggins parameter, the Kuhn statistical length exceeds the interface thickness; we are forced to re-interpret  $a$  as an *effective parameter* if we choose to keep the model.

The perturbation methods we use, while approximate, pose minimal problems with regard to the validity of our results. We noted in Section 2 that the parameter  $\varsigma$  had to be small. From purely dimensional considerations, the product  $\ell^2\sigma$  should be the largest combination of terms (involving  $\varsigma$ ) appearing in the perturbation analysis; if  $\ell^2\varsigma \ll 1$  in the region of parameter space we consider, then our perturbation analysis should be valid. We can estimate  $\varsigma$  value from its definition in Eq. (3). Again taking  $\xi = 1$  nm and letting  $\mathcal{N} \approx 300$ , we find that  $\varsigma \approx 10^{-3}/\chi^2$  nm<sup>-2</sup>. If we use  $\chi = 10$  as an upper limit suggested by our analysis, then we find  $\ell^2\varsigma \approx 10^{-3}$  is sufficiently small in the regimes we consider. We expect that our analysis should be valid for values of  $\chi$  as low as  $\chi \approx 1$ , which yields  $\ell^2\varsigma \approx 10^{-1}$ .

## 6. CONCLUSION

In this paper, we have used the Leibler-Ohta-Kawasaki phase field model to calculate the LER associated with microdomains of block copolymers in the lamellar phase; we consider system geometries that are expected to be used in a manufacturing setting, i.e. a few microdomains on a templated substrate. Using values of the  $\mathcal{N}$  (the index of polymerization) and  $\xi$  the (interface thickness) that correspond roughly to the 11 nm half-pitch node, we predict that the Flory-Huggins parameter  $\chi$  must be increased by at least a factor of five above current experimental values in order to reach target goals for the LER set forth in the ITRS. Our analysis also reveals that the number of microdomains has a greater affect on LER than the position of the microdomain interfaces relative to the boundary of the system.

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