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Interfacial fluctuations in an ideal block copolymer resist†

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We use field-theoretic computer simulations to show that intrinsic interfacial fluctuations may place limits on the use of block copolymer materials as resists for the nanofabrication of next-generation electronics devices. Even for an ideal system, our simulations demonstrate that the magnitude of the interfacial fluctuations is of the same order as the maximum interfacial roughness requirements noted in the International Technology Roadmap for Semiconductors. Furthermore, we show that the interfacial fluctuations exhibit an inverse power law dependence on the copolymer molecular mass, and we identify a peak in the interfacial fluctuation spectrum indicative of fluctuations with a wavelength of order the mesophase interdomain spacing. Recent research on block copolymer resists has focused on controlling and measuring long-range order; however, given the relatively large magnitude of the interfacial fluctuations observed in our ideal model, accurate measurements of interfacial fluctuations in candidate block copolymer materials will be necessary in order to determine their ultimate effectiveness as nanofabrication resists.

1 Introduction

Block copolymer (BCP) thin films are under investigation as possible sacrificial lithographic masks (*i.e.*, resists) in the fabrication of next-generation nanoelectronic and high-density-storage devices.^{1–4} The development of BCP thin films as viable nanofabrication materials requires precise control over the BCP mesophase order and uniformity.^{4–6} Specifically, the 2007 International Technology Roadmap for Semiconductors (ITRS) Directed Self-Assembly Research Requirements for 2011–2012 establishes that next generation lithographic applications will require BCP resists to exhibit a mesophase defect density of $\leq 0.02 \text{ cm}^{-2}$, a microdomain size (*i.e.*, critical dimension) of $\leq 11 \text{ nm}$, a 3σ interfacial roughness (*i.e.*, line edge roughness [LER]) of $\leq 2.1 \text{ nm}$, and a 3σ microdomain width roughness (*i.e.*, line width roughness [LWR]) of $\leq 1.7 \text{ nm}$.⁶

BCP order can be significantly enhanced, and in some cases directly controlled, *via* template-directed self-assembly—where one directs the self-assembly of BCP materials using chemical or topographical pattern templates.^{7–9} While the long-range order of such systems is dramatically increased, the local uniformity of the individual microdomains is not guaranteed. Specifically, thermodynamic fluctuations can introduce fluctuations in the microdomain interfaces that can significantly alter the interface structure (*e.g.*, see refs. 10–13), and thus potentially compromise device function.^{4–6}

We use computer simulations of a field-theoretic model of BCP ordering in two dimensions to elucidate the physical origin and behavior of interfacial and microdomain width fluctuations in a lamellar diblock copolymer melt confined by a nano-scale

topographical template. Our results indicate that the magnitude of the interfacial and microdomain width fluctuations are the same scale as the ITRS maximum LER/LWR requirements for BCP resists. Furthermore, we show that the interfacial and microdomain width fluctuations exhibit a power law dependence on the copolymer index of polymerization (*i.e.*, molecular mass). Finally, the spectral density of fluctuations is found to have a peak at a “characteristic wavenumber” approximately equal to the mean-field diblock copolymer characteristic wavenumber q^* ,¹⁴ indicative of interfacial and microdomain width fluctuations with wavelengths of order the mesophase interdomain spacing L_0 .¹⁵ Given the ideal nature of the model under consideration, one can view the fluctuation magnitudes reported in this study as a possible “best case scenario” for the magnitude of interfacial fluctuations in similar, real-world BCP resist materials. The industry and academic research focus has been on achieving long-range order in BCP resists; however, this study highlights that careful consideration of LER/LWR in BCP resist materials is equally important. Accordingly, accurate measurements of fluctuations in candidate BCP materials is necessary in order to determine their ultimate effectiveness as nanofabrication resists. Our results also stress the importance of considering the *fluctuation spectrum* when analyzing or modeling interfacial fluctuations in BCP melts.

2 Model and methods

We use a coarse-grained polymer field theory to simulate LER and LWR in a diblock copolymer resist. It is important to note that LER/LWR with a wavelength of order the target feature size (*i.e.*, the *critical dimension*, *half-pitch*, or, in our case, the *mesophase interdomain spacing*) will adversely affect the desired pattern and thus have a pronounced negative effect on device function.^{4,6} Therefore, technologists in the nanofabrication industry are primarily concerned with managing low-frequency LER/LWR in BCPs—specifically, LER/LWR with a wavelength

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of order, or greater than, the mesophase interdomain spacing. This is precisely the length scale where coarse-grained polymer field theories are most applicable. In addition, coarse-grained, field-theoretic simulations offer increased speed and flexibility when compared to particle-based methods such as coarse-grained molecular dynamics (MD) and coarse-grained Monte Carlo (MC) simulations. For example, our field-theoretic framework can be used to simulate the equilibrium thermodynamics of a dense polymer melt on a modern personal computer. Similar simulations based on a MD or MC framework would necessarily require supercomputer resources.^{12,13} It is important to note, however, that while coarse-grained field theories are well suited for simulations of low-frequency fluctuations, detailed BCP simulations of high-frequency fluctuations, of ordering on the length- or time-scale of segment relaxations, or of low-concentration systems would almost certainly require a particle-based, MD framework.

We begin with a Gaussian thread model of n AB diblock copolymers and n_w fixed wall “particles” in a volume V .^{16,17} The fraction of A segments along the BCP is denoted f , and the BCP index of polymerization is denoted N . Interactions between unlike segments/particles are parametrized by χ_{IJ} , with $I, J = A, B$ and w for A segments, B segments and wall particles, respectively. The wall particles are used to confine the BCP *via* the melt incompressibility constraint $\phi_A(\mathbf{x}) + \phi_B(\mathbf{x}) + \phi_w(\mathbf{x}) = 1$, where $\phi_A(\mathbf{x})$, $\phi_B(\mathbf{x})$ and $\phi_w(\mathbf{x})$ are the local volume fractions of A segments, B segments and wall particles, respectively. The field-theoretic canonical partition function is given by

$$Z \propto \int \delta \Xi \int \delta W e^{-C\mathcal{H}[\Xi, W]}, \quad (1)$$

where $C = n/V$ is the dimensionless polymer concentration, $\mathcal{H}$ is the field-theoretic energy functional, Ξ is a “pressure” field that is thermodynamically conjugate to the local incompressibility condition, and W is a “composition” field that is thermodynamically conjugate to the melt composition. For an incompressible melt, $C \propto N^{1/2}$.¹⁶ Here and throughout, we use spatial units with $R_g = 1$, where R_g is the radius of gyration of an unperturbed AB BCP. Details of the derivation of eqn (1) can be found in nearly any recent reference on BCP field theory (e.g., see refs. 16 and 17).

Thermodynamics fluctuations in a polymer melt vanish in the limit $C \rightarrow \infty$ (i.e., $N \rightarrow \infty$). For $C \rightarrow \infty$, the functional integrals in eqn (1) can be evaluated using the saddle point approximation (i.e., an asymptotic expansion) yielding a mean field theory (MFT) or self-consistent field theory (SCFT).¹⁶ For a finite C , SCFT does not account for composition fluctuations, and thus a more general integration technique is required.¹⁶

We use an integration scheme based on the *partial saddle point approximation*, where the pressure field Ξ is integrated using the saddle point approximation, and the composition field W is integrated using a stochastic method.^{16,18–21} The partial saddle point approximation assumes that only fluctuations in W are relevant; accordingly, the pressure field Ξ is instantaneously relaxed to its mean-field value. This approximation has been shown to be accurate for intermediate to large C (e.g., $C \geq 50$).²¹

For the stochastic integration of W we use a force-biased *smart Monte Carlo* algorithm.^{16,20,22} The strength of fluctuations in the system is controlled by varying C or the effective segregation

strength (i.e., the quench depth) $\tau = \chi_{AB}N - (\chi_{AB}N)_{\text{ODT}}$, where $\chi_{AB}N \geq (\chi_{AB}N)_{\text{ODT}}$, and $(\chi_{AB}N)_{\text{ODT}} \sim C^{-2/3}$ is the order-disorder transition value of $\chi_{AB}N$.^{16,23} In general, larger values of C or deeper quenches (i.e., larger values of τ) yield lower-amplitude fluctuations.

3 Results and discussion

The symmetric ($f = 0.5$) AB BCP melt was simulated using periodic boundary conditions in a rectangular, two-dimensional simulation space of area $L_x L_y$, with $L_x = 12.8$ and $L_y = 5.4$ and step sizes $\Delta x = \Delta y = 0.1$. The melt was assumed to be uniform but finite in the z direction, characteristic of an ultra-thin, quasi-two-dimensional film. By construction, the boundary conditions in the y direction were periodic; however, the local, fixed wall particle volume fraction $\phi_w(\mathbf{x})$ was used to confine the melt in the y direction between two walls (*cf.* Fig. 1) *via* the melt incompressibility constraint. The physical confinement size (imposed by the wall particles) in the y direction L was selected to be $L \approx 3$. This value of L is approximately equal to the mean-field interdomain spacing $L_0 \approx 3.3$.¹⁴ Although not presented in this paper, our simulations demonstrated that the interfacial and microdomain width fluctuations exhibit no appreciable dependence on the confinement size L , for L near 3. Specifically, we performed simulations with $L = 2.6$ and $L = 3.4$, and the spectral density of fluctuations were nearly identical to the results presented here for $L = 3$ (*cf.* Fig. 4).

The system size and resolution examined here facilitates simulation of large-amplitude, long-wavelength interfacial and microdomain width fluctuations. At higher resolutions, our field-theoretic simulations were dominated by high-frequency fluctuations that significantly complicated the LER/LWR analysis. Given the length scales of interest to resist technologists, we found that the system size and resolution indicated above was more than sufficient for capturing LER/LWR over the range of parameters examined in this study.

The wall-segment interaction parameters were selected to be $\chi_{Aw}N = -6.0$ and $\chi_{Bw}N = 6.0$ so that the A segments were *attracted* to the walls and the B segments were *repelled* from the walls. With this size and boundary condition, the mesophase-separated symmetric AB BCP formed a single B -segment lamellar microdomain extending in the x direction (again, *cf.* Fig. 1). This configuration represents the smallest reasonable topographical confinement configuration of interest in industrial

Fig. 1 Snapshot of the local A segment fraction $\phi_A(\mathbf{x})$ for a symmetric AB BCP with $C = 150$ and $\chi_{AB}N = 12$. Recall, $C \propto N^{1/2}$. The two A – B interfaces are labeled h_1 and h_2 , as in the figure. The black “bars” running along the top and bottom edges of the snapshot represent the confining walls.

applications, and thus we view it as a model of an ideal topographically confined system. Furthermore, this configuration allowed for fast, easy, and accurate determination of the A – B interface position (discussed below).

For the system configuration presented in this article, there are two A – B interfaces extending in the x direction. The position of the i th interface is labeled $h_i(x)$, with $i = 1, 2$, and calculated via an image thresholding procedure such that $\phi_B(x, h_i(x)) = 0.5$. We calculated the fluctuations in $h_i(x)$ from its average value along the interface $\delta h_i(x) = h_i(x) - \bar{h}_i$ and the standard deviation of $\delta h_i(x)$ along the interface $\sigma_{h_i} = [(\delta h_i)^2]^{1/2}$.[§] In addition to interfacial fluctuations, we also observed fluctuations in the width of the single, confined lamellar microdomain. The local width of the single microdomain is defined by $w(x) = h_2(x) - h_1(x)$. Again, we calculated the fluctuations in $w(x)$ from its average value $\delta w(x) = w(x) - \bar{w}$ and the standard deviation of $\delta w(x)$ along the interface σ_w . Plotting the standard deviations of the two interfaces, h_1 and h_2 , yielded two curves that were statistically indistinguishable. Here and throughout, we examine only the $i = 1$ interface with the understanding that the $i = 2$ interface exhibits statistically indistinguishable behavior. We report $\langle \sigma_h \rangle$ and $\langle \sigma_w \rangle$, where $\langle \cdot \rangle$ represents a “time” average over multiple Monte Carlo snapshots.

In Fig. 2 we plot the standard deviation of interfacial fluctuations and the standard deviation of microdomain width fluctuations for $C = 50$. This value of C corresponds to $N = \mathcal{O}(10^5)$.²¹ We observe two obvious trends. First, the fluctuations in h and w both decrease monotonically as a function of $\chi_{AB}N$. Increasing $\chi_{AB}N$ for constant C amounts to increasing the effective segregation strength τ , and thus one would expect

Fig. 2 Plots of the standard deviation of interfacial fluctuations and the standard deviation of microdomain width fluctuations for a nano-confined, symmetric AB BCP with $C = 50$. Here $\langle \sigma_h \rangle$ and $\langle \sigma_w \rangle$ are presented as a fraction of the mean-field, lamellar interdomain spacing $L_0 \approx 3.3$.¹⁴ The error bars represent the standard deviation in σ_h/L_0 or σ_w/L_0 over a Monte Carlo run, and the lines are provided as a guide to the eye.

[§] The average value of $h_i(x)$ along the interface $\bar{h}_i$ is defined by $\bar{h}_i = (1/L_x) \int_{L_x} dx h_i(x)$.

Fig. 3 Plots of the standard deviation of interfacial fluctuations and the standard deviation of microdomain width fluctuations for a nano-confined, symmetric AB BCP with $\chi_{AB}N = 12$. Recall, $C \propto N^{1/2}$; thus $C^2 \propto N$. As in Fig. 2, $\langle \sigma_h \rangle$ and $\langle \sigma_w \rangle$ are presented as a fraction of the mean-field, lamellar interdomain spacing $L_0 \approx 3.3$. The error bars represent the standard deviation in σ_h/L_0 or σ_w/L_0 over a Monte Carlo run, and the lines represent a fit to the power law form $b(C^2)^{-\alpha}$, where α and b are fitting parameters.

a decrease in the magnitude of fluctuations.¹⁰ Second, the microdomain width fluctuations are larger than the interfacial fluctuations. Specifically, we find that $\langle \sigma_w \rangle \approx 1.4 \langle \sigma_h \rangle$ over the range of parameters presented in Fig. 2. Below we discuss the relationship between $\langle \sigma_h \rangle$ and $\langle \sigma_w \rangle$.

In Fig. 3 we plot the standard deviation of interfacial fluctuations and the standard deviation of microdomain width fluctuations for $\chi_{AB}N = 12$. Recall, $C \propto N^{1/2}$, and thus $C^2 \propto N$. Therefore, in Fig. 3 we are effectively plotting the amplitude of fluctuations versus the copolymer index of polymerization or molecular mass for constant $\chi_{AB}N$. Varying C while holding the product $\chi_{AB}N$ constant requires varying χ_{AB} . This procedure is not easily reproduced in the laboratory; however, examining how LER/LWR varies with C for constant $\chi_{AB}N$ can help elucidate the nature of the fluctuations, as discussed below. In general, we expect the interfacial fluctuations to decrease with increasing C —as the system becomes more “mean-field-like”—and that is exactly what is observed here. We note that both $\langle \sigma_h \rangle$ and $\langle \sigma_w \rangle$ have an observed power law dependence $(C^2)^{-\alpha}$ with an exponent $\alpha \approx 1/5$ to $1/4$ for all values of $\chi_{AB}N$ examined. Again, we note that $\langle \sigma_w \rangle \approx 1.4 \langle \sigma_h \rangle$ over the range of parameters presented in Fig. 3.

A fluctuating interface with tension has $\langle \sigma_h \rangle \propto \gamma^{-1/2}$, where γ is the interfacial tension.^{10,24,25} For polymer–polymer interfaces in a melt (including BCPs), $\gamma \propto C(\chi_{AB}N)^{1/2}$, and thus surface tension waves in polymer–polymer interfaces have $\langle \sigma_h \rangle \propto (C^2)^{-1/4}$, for constant $\chi_{AB}N$. This is approximately the scaling observed in Fig. 3. Further analysis of the real-space fluctuations demands careful consideration of a full three-dimensional system (e.g., dimensional considerations can play an important role in determining the precise behavior of real-space fluctuations under confinement and with respect to $\chi_{AB}N$; see refs. 10 and 24); however, we expect the trends observed here to remain qualitatively true, and the observed dependence on C should remain

quantitatively true provided that the BCP interfaces can be effectively modeled as fluctuating interfaces with surface tension.

The standard deviation of interfacial and microdomain width fluctuations are related *via* the *error propagation equation*. For the microdomain width we have $\delta w(\delta h_1, \delta h_2) = \delta h_2 - \delta h_1$, and thus δw is a function of δh_1 and δh_2 . Accordingly,

$$\sigma_w^2 = \sigma_{h_1}^2 + \sigma_{h_2}^2 - 2\text{Cov}(\delta h_1, \delta h_2) \quad (2)$$

where $\text{Cov}(\delta h_1, \delta h_2)$ is the *covariance* of δh_1 and δh_2 . If we assume that the two interfaces h_1 and h_2 are statistically independent (*i.e.*, uncorrelated) with equal standard deviations $\sigma_h = \sigma_{h_1} = \sigma_{h_2}$, then $\text{Cov}(\delta h_1, \delta h_2) = 0$, and

$$\sigma_w = \sqrt{2}\sigma_h. \quad (3)$$

In general, we do not expect this to be true for the lamellar phase of a diblock copolymer, as the natural connectivity of the copolymers and the thermodynamic fluctuation spectrum of the copolymer melt can lead to correlated fluctuations.^{15,26,27} In any event, the relationship between σ_w and σ_h can shed light on the interface–interface correlations without the need to examine a specific model of interacting, fluctuating interfaces. The data presented in Figs. 2 and 3 suggest that $\langle \sigma_w \rangle \approx 1.4 \langle \sigma_h \rangle \approx \sqrt{2} \langle \sigma_h \rangle$, with $|\langle \sigma_w \rangle / L_0 - \sqrt{2} \langle \sigma_h \rangle / L_0| < 0.01$ for all system parameters presented in this study. This suggests that there is minimal coupling between the two interfaces. The confining walls likely act to suppress interfacial fluctuations,¹² alter the fluctuation spectra,²⁸ and thus indirectly affect the coupling between the two adjacent interfaces. Further study of the specific effects of the confining wall on the magnitude of fluctuations and fluctuation spectra is beyond the scope of this paper and will be considered in a forthcoming publication.²⁸

In order to elucidate the nature of the observed fluctuations, we plot the spectral density of δh and δw in Fig. 4 for $C = 150$ and $\chi_{AB}N = 12$. In Fig. 4a we plot $\langle |\delta h(q_x)|^2 \rangle$ and in Fig. 4b we plot $\langle |\delta w(q_x)|^2 \rangle$. The peaks at $q_x \approx 2$ in Fig. 4 indicate that there is a characteristic wavelength for the interfacial and microdomain width fluctuations. We believe that these peaks are caused by *intrinsic* thermodynamic fluctuations in the ordered BCP melt.

For weakly segregated BCP melts, the *ordered phase* thermodynamic fluctuation spectrum can be estimated analytically and calculated numerically.^{15,27} One finds that for symmetric (*i.e.*, $f = 0.5$) diblock copolymers, the primary component of the fluctuation spectrum is *isotropic* and centered about $|\mathbf{q}| = q^*$, where q^* is the mean-field characteristic wavenumber for a diblock copolymer melt.^{15**} In other words, regardless of the direction examined, we expect to observe a peak in the fluctuation spectrum at q^* . Therefore, for the “in-plane”

Fig. 4 Plots of (a) the spectral density of interfacial fluctuations and (b) the spectral density of microdomain width fluctuations for a nano-confined, symmetric *AB* BCP with $C = 150$ and $\chi_{AB}N = 12$. In order to ensure sufficient Fourier space resolution for observing the peak, the system size was selected to be $L_x = 128.0$ and $L_y = 5.4$ with $\Delta x = \Delta y = 0.1$.

interfacial and microdomain width fluctuations examined here, a peak at $q_x = q^*$ should be observed. The mean-field characteristic wavenumber for a bulk *AB* BCP melt is $q^* \approx 1.9$,¹⁴ which agrees quite well with the peak positions observed in Fig. 4.

LER and LWR with a wavelength of order L_0 or larger can negatively impact device function. Fig. 4 illustrates that the diblock copolymer resist examined in this study exhibits *intrinsic* LER and LWR with wavelength $L_0 = 2\pi/q^*$. This could limit the use of diblock copolymers as lithographic resists, and thus careful examination of the fluctuation spectra in real-world diblock copolymers resists is necessary.

The simulations presented in this paper neglect dynamical issues often important in BCP experiments, most notable of which is the glass transition. Upon cooling (*i.e.*, upon increasing χ_{AB}), polymer systems experience a glass transition at a temperature $T_g \propto 1/(\chi_{AB})_g$. Near and below this temperature, the molecular-scale structural relaxation and the approach to equilibrium slow dramatically. We suppose that a quench from an initial temperature $T_i \gg T_g$ to a final temperature $T_f < T_g$ will freeze in the fluctuation spectrum from T_i , and the glassy nature

¶ Recall, for random variables a and b , $\text{Cov}(a, b) = \overline{(a - \bar{a})(b - \bar{b})}$.

¶ We use the notation $g(q_x)$ to represent the Fourier transform of the function $g(x)$.

** For bulk *AB* BCPs, q^* corresponds to the interdomain spacing L_0 of the mesophase (*i.e.*, $q^* = 2\pi/L_0$). In asymmetric diblock copolymers (*i.e.*, $f \neq 0.5$), the fluctuation spectrum also contains an *anisotropic* component that is centered about the wavevectors $\{\mathbf{K}_i\}$, where $\{\mathbf{K}_i\}$ is a set of reciprocal lattice vectors corresponding to an underlying order–order transition of the BCP. For example, for the lamellar phase of an asymmetric diblock copolymer, the set $\{\mathbf{K}_i\}$ has a hexagonal symmetry corresponding to the underlying “LAM-to-HEX” transition.^{15,27}

of the system will prevent the BCP from accessing the lower-amplitude fluctuations at T_f . Therefore, it is important to associate the values of χ_{AB} examined in this paper with values of $\chi_{AB} \ll (\chi_{AB})_g$ (i.e., $T \gg T_g$). Experimentalists and technologists should select diblock copolymers with as low a T_g as possible so that they can access the low-LER/LWR region of the phase space at high χ_{AB} .

Let us consider the potential implications of the fluctuation analysis presented above. We have outlined a series of resolution and roughness requirements for BCPs presented in the 2007 ITRS; now we consider these requirements in the context of our simulations. As a matter of convenience, we express all ITRS roughness requirements as a fraction of the ITRS required interdomain spacing for a hypothetical symmetric diblock copolymer $L_{0,\text{ITRS}} \approx 2 \times 11 \text{ nm} = 22 \text{ nm}$. This allows us to directly compare the ITRS requirements with the roughness results from our field-theoretic simulations—presented as a fraction of $L_0 = 3.3$ in Figs. 2 and 3. The ITRS established that self-assembling systems used in lithography (such as BCPs) will require 3σ LER less than $2.1 \text{ nm}/L_{0,\text{ITRS}} = 0.095$ and 3σ LWR less than $1.7 \text{ nm}/L_{0,\text{ITRS}} = 0.077$. We can see in Figs. 2 and 3 that the 3σ value of interfacial roughness (LER) and microdomain width roughness (LWR) are both $\mathcal{O}(0.1)$.^{††} That is, the simulation values of LER and LWR are roughly the same scale as, or slightly larger than, the ITRS requirements. In making this comparison, we are effectively extrapolating the observed behavior of our simulated $C = 50$ (i.e., $N = \mathcal{O}(10^5)$) diblock copolymer system down to the low-molecular mass diblock copolymer systems of interest to resist technologists. The situation for low-molecular mass diblock copolymers is likely even worse than suggested here. We have demonstrated that LER scales like $\langle\sigma_h\rangle \propto N^{-1/4}$, for constant $\chi_{AB}N$ (from Fig. 3), and the mesophase interdomain spacing scales like a positive power of N ; that is, $L_0 \propto N^\nu$, where $\nu > 0$. Therefore, the relative amplitude of LER with respect to L_0 scales like $\langle\sigma_h\rangle/L_0 \propto N^{-(1+4\nu)/4}$. This scaling relationship tells us that the relative amplitude of LER gets *larger* for smaller N . Clearly, this could represent a challenge for diblock copolymer resist applications.

It is important to note that the trend observed in Fig. 3 is at odds with the requirements of the BCP lithography community. As outlined in the ITRS,^{4,6} BCP resists will require mesophase interdomain spacings of order $\leq 10 \text{ nm}$. This will necessarily require selecting diblock copolymers with low molecular mass (i.e., low index of polymerization) because the interdomain spacing scales as a positive power of C . As can be seen in Fig. 3, a reduction in the index of polymerization results in larger interfacial fluctuations. In principle, one can offset the necessary decrease in C with an appropriate increase in χ_{AB} (cf. Fig. 2); however, it may not be possible to synthesize neat diblock copolymers with large enough values of χ_{AB} . Furthermore, large- χ_{AB} diblock copolymers often have an unreasonably slow approach to equilibrium, due in large part to their glassy nature

on both the molecular scale *and* mesoscale. All of these concerns will need to be thoroughly addressed before diblock copolymer resists can function as a viable nanofabrication tool.

4 Conclusion

We have presented a computational study of interfacial fluctuations (i.e., line edge roughness [LER]) and microdomain width fluctuations (i.e., line width roughness [LWR]) in a two-dimensional, nano-confined diblock copolymer melt. Our simulations indicate that BCP LER and LWR exhibit a power law dependence on $C^2 \propto N$, and the spectral densities of LER and LWR have peaks at q^* . The power law dependence on C^2 is consistent with a fluctuating interface with surface tension. The observed peak in the spectral density of fluctuations indicates a preferred wavelength $L_0 = 2\pi/q^*$ for the A – B interfacial and microdomain width fluctuations. The peak position is consistent with a weak segregation theory of fluctuations in an ordered symmetric diblock copolymer melt.

Much of the past and present research on BCP resist materials has focused on characterizing long-range order or improving long-range order with chemical or topographical templates, while very little work has focused on LER and LWR in candidate BCP resists. Given that the magnitude of the interfacial and microdomain width fluctuations in our *ideal* model are the same scale as the ITRS maximum LER/LWR requirements for BCP resists and that the characteristic wavelength of the observed LER and LWR is of order the desired featured size L_0 , accurate measurements of fluctuations and fluctuation spectra in real BCP resist materials is absolutely necessary in order to determine their effectiveness as lithographic masks in next-generation nanomanufacturing.

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^{††} Here we are interested in a rough, order-of-magnitude comparison. Clearly, $3\langle\sigma_h\rangle/L_0$ and $3\langle\sigma_w\rangle/L_0$ vary quite significantly over the range of parameters studied; however, from Figs. 2 and 3 we can see that all of the measured values are of the same order of magnitude, with microdomain width fluctuations slightly larger in magnitude than interfacial fluctuations.

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