Stochastic modeling of photoresist development in two and three dimensions

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Abstract. The concepts of dynamical scaling in the study of kinetic roughness are applied to the problem of photoresist development. Uniform, open-frame exposure and development of photoresist corresponds to the problem of quenched noise and the etching of random disordered media and is expected to fall in the Kadar-Parisi-Zhang (KPZ) universality class for the case of fast development. To verify this expectation, simulations of photoresist development in 1 + 1 and 2 + 1 dimensions were carried out with various amounts of random, uncorrelated noise added to an otherwise uniform development rate. The resulting roughness exponent $\alpha$ and the growth exponent $\beta$ were found to match the KPZ values nearly exactly. The impact of the magnitude of the underlying development randomness on the values of these exponents was also determined, and an empirical expression for predicting the kinetic roughness over a wide range of conditions is presented. © 2010 Society of Photo-Optical Instrumentation Engineers. [DOI: 10.1117/1.3494607]

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1 Introduction

Stochastic models of lithography consider fundamental events such as the absorption of a photon or the chemical reaction of a molecule as stochastic events. As such, these events are described probabilistically, with the mean-field “rate” equation describing the probability that the event occurs. Of course, such a probabilistic description will not make deterministic predictions—instead, quantities of interest will be derived by their probability distributions, which in turn are characterized by their moments, such as the mean and variance. While stochastic modeling has been successfully applied to photoresist exposure and post-exposure bake processes in recent years, the stochastic behavior of resist dissolution is much less understood. Ultimately, the final result will be a roughness of the resist feature sidewalls that leads to line-edge roughness (LER) and linewidth roughness (LWR) of the resist feature. One common approach to studying LER formation is through the use of Monte Carlo simulations and mesoscale modeling. While Monte Carlo methods can be extremely useful, there is also a need for the development of simple, analytical expressions that capture the essence of the LER formation mechanisms. One approach to studying the stochastic nature of photoresist dissolution, which will be employed here, involves the characterization of scaling relationships as a means for elucidating fundamental mechanisms.

Since the final LER of a high-resolution lithographic feature will include all resist and aerial image contributions, studying LER to extract the contribution of only resist development can be difficult. A simpler approach is to remove the aerial image from the experiment and study the resist surface roughness after a uniform open-frame exposure and development. The use of surface roughness after open-frame exposure and development as a probe for understanding the stochastic nature of resist development will be examined in detail in this paper. In particular, an analysis approach known as dynamical scaling will be applied to photoresist development, and simulated open-frame dissolution results will be analyzed in this way.

2 Dynamical Scaling

Over the last 25 years, fractal concepts have been successfully applied to the appearance of roughness in surface growth phenomena, with many applications including deposition and etching. Called disorderly surface growth or kinetic roughness, research has focused on determining how experimental roughness or the roughness predictions of specific models scale with time and distance. While the magnitude of the roughness is unique to the specific experiment or model and their circumstances/parameters, the scaling behavior of the roughness tends to be more universally applicable to a wide range of conditions as long as the basic mechanism remains consistent. This section will review current understanding of kinetic roughness as applied to a simple lithographic case: open-frame exposure and development of photoresist.

Open-frame exposure of photoresist coated on a planar wafer leads, after development, to a certain resist height remaining, $h$. Including the effects of roughness, this surface height will be a function of wafer position, $h(x, y)$. If the possibility of overhangs is ignored, this surface height will be a single-valued function. (The validity of this assumption will be explored shortly.) Such surfaces are expected to be (or eventually become) self-affine, since the nominally vertical direction of the isotropic development will cause the $z$ dimension (the height dimension) of the resist surface to scale differently from the $x$-$y$ dimensions. For a statistical self-affine surface, the surface is characterized by statistical
properties. The RMS surface height difference, often called the interface width or the surface roughness, of a statistically self-affine surface scales with the measurement length $L$ as

$$\sigma_w \propto L^\alpha,$$

where $\alpha$ is called the roughness exponent. As you will see, however, resist surfaces are self-affine only over a region smaller than the correlation length of the roughness.

For the development of a uniform, open-frame exposure of photoresist, the resist surface is initially perfectly smooth. But as development proceeds, stochastic effects lead to an increased roughening of the surface. Thus, the statistical properties of the interface [the mean height $\langle h \rangle$ and the amount of roughness $\sigma_w$] are a function of time. Ignoring absorption so that the resist receives a uniform exposure through its depth (and assuming a perfect antireflection coating so that no standing waves are present), the mean development rate will be constant. Thus, the mean surface height will scale linearly with time. Empirically, many problems in etching and deposition show a roughness that, for moderately small times, grows as

$$\sigma_w \propto t^\beta,$$

where $\beta$ is called the growth exponent. The growth in roughness as development proceeds does not continue indefinitely. For a given measurement size $L$, the interface roughness saturates after a long enough time. However, since the roughness varies with $L$ according to Eq. (1), the point of saturation with development time depends on the size of the measurement region. The overall scaling can be summarized as

$$\sigma_w \propto L^\alpha f \left( \frac{t}{L^2} \right),$$

where

$$f(u) = \begin{cases} u^\beta & u \ll 1 \\ 1 & u \gg 1 \end{cases}$$

and $z = \alpha/\beta$ is called the dynamic exponent. The proper choice of $\alpha$ and $\beta$ allows dynamic roughness data [$\sigma_w(L, t)$] to collapse to a single universal curve for all $L$, giving a very sensitive method for determining these exponents.

For our problem, the source of interface roughness is a statistical uncertainty in the resist development rate $r$ as a function of position. Thus, the development rate can be separated into a mean dissolution rate plus a random variable $\eta$:

$$r(x, y, h) = \langle r \rangle + \eta(x, y, h).$$

Obviously, $\eta$ has been formulated to have zero mean, and its standard deviation is $\sigma_r$. It is very important to note that even if the underlying noise is uncorrelated, the resulting rough resist surface will exhibit height-to-height correlations. The cause of these correlations is the isotropic nature of dissolution. If, due to random fluctuations, one point in the resist interface develops down more quickly than the rest, this dimple in the resist surface will begin to spread laterally. Thus, the neighboring points on the interface will have a resist height that is correlated with the original fast-developing point. The typical distance over which heights interact is called the parallel correlation length, $\xi_p$. Initially, this $x$-$y$ plane correlation length is small, but it grows with time as

$$\xi_p \propto t^{1/2}.$$ (5)

The correlation length cannot grow to more than the measurement domain, so that eventually it saturates at $L$.

Another approach to characterizing the roughness of a surface is to determine its power spectral density (PSD), the magnitude squared of the Fourier transform of the relative surface height:

$$\text{PSD}(f) = \lim_{L \to \infty} \frac{1}{L^2} \left| \int_{-L/2}^{L/2} \int_{-L/2}^{L/2} \tilde{h}(x, y) \times \exp[-i2\pi(f_x x + f_y y)]dx dy \right|^2,$$ (6)

where $f = (f_x^2 + f_y^2)^{1/2}$, and $\tilde{h} = h - \langle h \rangle$. The self-affine scaling hypothesis of Eq. (3) can be translated into a scaling relationship for the surface roughness PSD. In the long-time limit and the long-wavelength limit, we expect that

$$\text{PSD}(f) \propto \frac{1}{f^{d+2\alpha}},$$ (7)

where a $d+1$ dimensional problem is characterized by a $d$-dimensional interface. Thus, $d = 2$ for the case of open-frame exposure and development of photoresist.

### 3 The KPZ Equation

The evolution of a resist surface under the assumption of a surface-rate-limited reaction mechanism during dissolution can be described by a simple differential equation. Since $r$ is the development rate normal to the resist surface, the rate at which the surface height at a specific $(x, y)$ point changes is given by

$$-\frac{\partial h}{\partial t} = r(1 + |\nabla h|^2)^{1/2},$$

where $\nabla h$ is the gradient along the resist surface and represents the maximum slope of the interface at a given $(x, y)$ point. If the surface is essentially horizontal with a small amount of roughness, $|\nabla h|^2 \ll 1$, and this equation can be approximated as

$$-\frac{\partial h}{\partial t} \approx r + \frac{r}{2} |\nabla h|^2.$$ (9)

Combining with Eq. (4) and assuming that the noise level is low ($\sigma_r \ll r$),

$$-\frac{\partial h}{\partial t} \approx \langle r \rangle + \frac{r}{2} |\nabla h|^2 + \eta.$$ (10)

This result is a simplification of a common stochastic growth model called the Kadar-Parisi-Zhang (KPZ) equation. In its full form, the KPZ equation is generally written as

$$\frac{\partial \tilde{h}}{\partial t} = v \nabla^2 \tilde{h} + \frac{\lambda}{2} |\nabla \tilde{h}|^2 + \eta,$$ (11)

where $\lambda = \langle r \rangle$, $\bar{h} = \langle r \rangle t - h$, and $v$ is a surface tension or diffusion term that relaxes the interface and contributes to smoothing. Lithography simulators assume $v = 0$, but it is possible that there is some relaxation mechanism at work in...
actual development. Here, we will assume that the nonlinear terms dominate (both the $|\nabla h|^2$ and $\eta$ terms are nonlinear) and that $\nu$ is very small (that is, we are in the so-called strong-coupling limit).

For high-temperature deposition and etching processes, $\eta$ in the KPZ equation is dominated by thermal noise—that is, it is a random variable in time. For the case of resist development, however, the noise is a spatial variation in development rate that, for a given resist instantiation, does not vary with time. Kessler et al. described this type of noise as quenched.\(^\text{14}\) Materials that exhibit quenched noise are called disordered media or random disordered media. If, however, the dissolution rate is moderately high, the quenched noise varies with time since the value of $z$ varies with time: $\eta(x, y, z) = \eta(x, y, h) \approx \eta(x, y, r(t))$. As such, in this “fast” developing regime, dissolution with quenched noise is expected to be in the same universality class as the KPZ equation with thermal noise. In the $d = 1$ case, the KPZ exponents can be determined exactly using renormalization group techniques\(^\text{9,15,16}\) to be $\alpha = 1/2$ and $\beta = 1/3$. For $d = 2$, however, an exact determination of the exponents is not possible. For the case where $\nu$ is small, where $L \gg \sigma_r$, and where the underlying quenched noise is uncorrelated, Hentschel and Family\(^\text{17}\) have shown that a $d = 2$ interface should have $\alpha = 0.4$ and $\beta = 0.25$, in agreement with simulation results by Kim and Kosterlitz.\(^\text{18}\) In general, scaling arguments (reinforced by renormalization group methods) show that $\alpha + z = 2$ in all dimensions\(^\text{9}\) (sometimes referred to as Galilean invariance).

4 Simulations of Photoresist Development

Simulation was used to predict the resist height as a function of development time for an open-frame exposure/development in the presence of stochastic dissolution rate noise. The dissolution rate of the photoresist followed Eq. (4), with a mean $\langle r \rangle$ and stochastic term $\eta$, set to be an uncorrelated, Gaussian-distributed random variable of standard deviation $\sigma_r$. Since development rate can never be less than zero, the noise term was symmetrically limited so that $r$ was always between 0 and $2|\langle r \rangle|$. The initial study considered the $d = 1$ case, with a maximum $L = 2048$ nm and a resist thickness of $10,000$ nm. The simulation grid was $1 \text{ nm}$ in both $x$ and $z$. A level-set algorithm\(^\text{19}\) was used to turn the $r(x, z)$ data into a resist surface as a function of development time. Linear variation of development rate between grid points was assumed, and repeating boundary conditions in $x$ and $y$ were used.

Development time was varied from $2$ s to the time needed to clear the resist in increments of $2$ s. At each development time, the mean and standard deviation of the resist height $h(x)$ was determined. Since the development algorithm is capable of predicting overhangs in the resist surface, these overhangs were ignored by setting $h(x)$ to be the first surface encountered when looking up from the substrate. (This is equivalent to eroding the overhangs to create a single-valued resist height function.) $L$ was varied by taking the $2048$-nm width and breaking it up into two $1024$-nm regions, four $512$-nm regions, etc., down to $16$-nm-sized regions. When multiple regions were cut from the one simulation, the analysis results were averaged over these regions.

An example of the results of these simulations with $\langle r \rangle = 10 \text{ nm/s}$ and $\sigma_r = 2 \text{ nm/s}$ (Gaussian distribution) is shown in Fig. 1. Figure 1(a) shows the results of a single simulation, and Fig. 1(b) shows the average of $500$ simulations. By setting the roughness and growth exponents to their expected KPZ $d = 1$ values ($\alpha = 1/2$, $\beta = 1/3$), the data of Fig. 1(b) collapses into one universal curve, as shown in Fig. 2. (This is accomplished by simply plotting the scaled values of roughness, $\sigma_r/L^{\alpha\nu}$, versus the scaled development time, $L|\langle r \rangle|^\beta )$. This is compelling evidence that the photoresist etching mechanism follows the KPZ universality class, as expected for the case of uncorrelated noise and a moderately large value for $\langle r \rangle$.

Simulations such as those shown in Fig. 2 were repeated as $\sigma_r/\langle r \rangle$ was varied from $0.02$ to $0.28$. For each value of
σ_r/⟨r⟩, the α and β exponents were adjusted to give the best collapse of the data into a single curve. The errors in determining α and β in this way were estimated by varying these exponents until the data collapse was notably worse. Figure 3 shows the resulting values of α and β. Gaussian-distributed development rate noise was used in all cases, but repeating the simulations with uniformly distributed noise produced almost identical results.

Simulations in 2 + 1 dimensions were also carried out, although L was limited to 512 nm and the resist thickness was 2000 nm. The development time increment was set to 1 s. The resulting values of α and β are shown in Fig. 4, and they match well with expected KPZ values of α = 0.4 and β = 0.25.

While overhangs were ignored in the analysis of the scaling exponents given earlier, the number of overhangs in the simulations shown earlier was measured and is shown in Fig. 5. The number of overhangs is defined as the number of x [or (x, y)] points above which an overhang can be seen. As expected, the number of overhangs grows rapidly as σ_r/⟨r⟩ increases. No overhangs were detected when σ_r/⟨r⟩ < 0.12.

The 2 + 1 simulations showed roughly 15% more overhangs per 1000 grid points than the 1 + 1 simulations.

The impact of ⟨r⟩ and σ_r on the mean resist height was also examined. According to Eq. (10), the rate at which the mean resist height decreases with time is slightly greater than the mean dissolution rate due to the nonzero resist surface gradient:

$$\frac{1}{2} \left\langle |\nabla h|^2 \right\rangle = -\frac{1}{\langle r \rangle} \frac{\partial \langle h \rangle}{\partial t} - 1.$$ (12)

By measuring the slope of the mean resist height versus development time curve from the simulations, the mean square resist surface gradient can be determined.

To study this effect, the mean square gradient of the resist surface was calculated from the simulation results using Eq. (12). The results are shown as the data points in Fig. 6. Empirically, the mean squared surface gradient is found to
Fig. 6 Plots of the mean square gradient, $\langle |\nabla h|^2 \rangle$, as a function of the relative dissolution rate standard deviation. Data points are simulation results; solid lines are from the empirical expression of Eq. (13).

scale as

$$\langle |\nabla h|^2 \rangle \sim \left( \frac{\sigma_r}{\langle r \rangle} \right)^{1.67}. \tag{13}$$

This empirical model is shown in Fig. 6 as the solid curves.

The power spectral density approach was also applied to the rough surfaces generated in the preceding $1 + 1$ simulations. Figure 7 shows a typical result. The linear (mid- to high-frequency) region follows the self-affine behavior of Eq. (7), allowing the roughness exponent $\alpha$ to be extracted. At the highest frequencies, the resolution limit (grid size) of the simulation causes a loss of information and a resulting leveling off of the PSD. The low-frequency leveling of the PSD occurs for wavelengths greater than the correlation length of the roughness. If the correlation length can be extracted from the PSD for different development times, a fit to Eq. (5) will give the exponent $z$.

Typically, the correlation length is defined using the autocorrelation function for the rough surface. Empirically, many systems exhibit autocorrelation behavior that can be well modeled as

$$R(\tau) = \lim_{L \to \infty} \frac{1}{L} \int_{-L/2}^{L/2} h(x)h(x + \tau) \, dx \approx \sigma^2 \exp\left[-\left(\frac{\tau}{\xi} \right)^{2\alpha}\right]. \tag{14}$$

where the $1 + 1$ dimension definition of the autocorrelation function is shown here. Since the process can be treated as a wide-sense stationary random process ($\langle h \rangle$ and $\sigma_w$ do not vary with position), the PSD is the Fourier transform of the autocorrelation function. For the $1 + 1$ dimension case ($d = 1$) and with $\alpha = 0.5$, this Fourier transform can be carried out analytically using the model autocorrelation function of Eq. (14):

$$\text{PSD}(f) = \frac{2\sigma^2 \xi_0}{1 + (2\pi f \xi_0)^2}. \tag{15}$$

The form of Eq. (15), along with the self-affine behavior of Eq. (7), inspires a form of the PSD for general values of $d$ and $\alpha$:

$$\text{PSD}(f) = \frac{\text{PSD}(0)}{1 + (2\pi f \xi_0)^{2\alpha + 2}}. \tag{16}$$

In a simulation with a nonzero grid size (or a measurement with a resolution limit), the high-frequency response is limited by a resolution noise limit. Assuming that this resolution limit results in white noise added in quadrature to the PSD, a final semiempirical expression should describe well the observed PSD:

$$\text{PSD}(f) = a \left[ \left( \frac{1}{f_c^s + f^s} \right)^2 + \left( \frac{1}{f_n^s} \right)^2 \right]^{1/2}, \tag{17}$$

where $a$ is a constant, $s = d + 2\alpha = \text{the slope of the linear portion (on a log-log scale)}$ of the PSD, $f_c = 1/(2\pi \xi_0)$, and $f_n$ is the high-frequency limit (resolution noise limit) of the

Fig. 7 Power spectral density (PSD) results for $1 + 1$ simulations of open-frame resist surface roughness with $\langle r \rangle = 10 \text{ nm/s}$ and $\sigma_r = 2 \text{ nm/s}$ (Gaussian distribution) and a development time of 9 s: (a) results from a single simulation, and (b) the average of 6000 simulations.
Fitting the PSDs resulting from simulations for different development times allows the correlation length to be extracted (symbols). A fit of correlation length versus development time to Eq. (5) gives the dynamic exponent, $z$ (line).

Simulation due to the grid size (which is assumed to be much larger than $f_c$).

Over a range of development times from 9 to 900 s, the extracted value of $\alpha$ (from the best fit slope $s$) was in the range of 0.483 to 0.498 (compared to the expected value of 0.5). Fitting the resulting correlation length versus development time to Eq. (5) gave $\varepsilon = 1.52$ (compared to the expected value of 1.5) and $\beta = 0.32$ (expected value of 1/3). Figure 8 shows these results. The noise limit was determined to be $1/f_n \approx 2.4$ nm for a grid size of 1 nm, about equal to the inverse of the Nyquist critical frequency (twice the grid spacing).

5 Discussion of Results

Simulation results presented in the previous section clearly show that a straightforward surface-limited dissolution/etch model falls into the KPZ universality class when the dissolution rate is sufficiently high. The collapse of data into a single curve as shown in Fig. 2 confirms the Family-Vicsek scaling of Eq. (3), with the resulting roughness and growth exponents matching the expected KPZ results. But while Eq. (3) expresses the limits of small and large time behavior of the surface roughness, the simulations done in this work follow a very specific scaling function that remains the same for all of the conditions simulated in both 1+1 and 2+1 dimensions. The data from Fig. 2 and all other simulations done for this work can be fit extremely well by the following empirically determined expression:

$$\hat{\sigma} = \hat{\sigma}_{sat} \left[ 1 + \frac{\hat{t}_x}{\hat{t}} \right]^{-\beta},$$

where $\sigma_{sat}$ is the saturation (long time) value of the surface roughness, $\hat{t}_x$ is the cross-over time between the roughness growth and saturation regimes, and the hat over the variable represents the Family-Vicsek scaled versions of the quantities:

$$\hat{\sigma} = \hat{\sigma}_{sat}, \quad \hat{t} = \frac{t}{L^z}. $$

It is easy to see that Eq. (18) satisfies the basic scaling requirements of Eq. (3). Equation (18) also shows that four parameters, $\alpha$, $\beta$, $\hat{\sigma}_{sat}$, and $\hat{t}_x$, completely describe the dynamic roughness behavior for all $L$ and $t$. The dependence of the scaling exponents on the magnitude of the underlying development rate randomness were shown in Figs. 3 and 4. By fitting Eq. (18) to the scaled data, best-fit values of $\hat{\sigma}_{sat}$ and $\hat{t}_x$ were also generated. These results are shown in Figs. 9 and 10.

The universal curve of Eq. (18) provides a predictive capability for the resulting surface roughness given the four parameters $\alpha$, $\beta$, $\hat{\sigma}_{sat}$, and $\hat{t}_x$. The two scaling exponents $\alpha$ and $\beta$ are known approximately based on their KPZ universality class, but have been shown here to vary slightly (and about linearly) with $\sigma_r/\langle r \rangle$. The remaining two parameters $\hat{\sigma}_{sat}$ and $\hat{t}_x$ vary significantly with $\sigma_r/\langle r \rangle$. It will be interesting to see whether the form of Eq. (18) could be derived based on physical arguments and whether this form is appropriate for other dynamic scaling problems.
6 Conclusions

In this work, the concepts of dynamical scaling were employed to study the growth of surface roughness during photoresist development. All evidence suggests that resist surfaces become self-affine over length scales less than a few correlation lengths and are governed by the scaling laws commonly used in the study of kinetic roughness. This scaling is characterized by two basic exponents, the roughness exponent $\alpha$ and the growth exponent $\beta$. The simple experiment of open-frame (uniform) exposure and development followed by measurement of the mean thickness and RMS surface roughness is a powerful tool for extracting these exponents and confirming the applicability of dynamical scaling.

Simulation of development in $1+1$ and $2+1$ dimensions also shows very nicely that the expected scaling behavior is followed. Additionally, and not unexpectedly, these $1+1$ simulations match the KPZ universality class almost perfectly, giving $\alpha \approx 1/2$ and $\beta \approx 1/3$. In $2+1$ dimensions, simulations match the KPZ universality class in the strong-coupling limit, giving $\alpha \approx 0.40$ and $\beta \approx 0.25$. The match to this universality class is not exact, however, since the transformation from quenched random noise (spatial variations) to thermal noise (temporal variations) under fast developing conditions is not exact. As a result, $\alpha$ and $\beta$ vary slightly with the relative magnitude of the underlying development rate uncertainty.

While the Family-Vicsek scaling of Eq. (3) is a foundation for the dynamical scaling approach to kinetic roughness formation, little work has been done on establishing the exact form of the function in Eq. (3) for any specific case. Here, an empirical function is proposed in Eq. (18) that matches all of the simulation data generated for this paper in both $1+1$ and $2+1$ dimensions. As a result, the surface roughness can be predicted for any development time and domain size once the four parameters of this expression have been determined. While two of those parameters, $\alpha$ and $\beta$, are essentially determined by the universality class of the problem (varying only slightly with the specific conditions of the experiment), the remaining two parameter values are particular to the problem—that is, to the value of $\sigma_r/(r)$.

Much future work is needed to fully apply the concepts of dynamical scaling to photoresist development. In particular, the simulations shown here are for reasonably fast development (small values of $\sigma_r/(r)$). It is unclear whether slower development rates will still exhibit KPZ scaling behavior, and this important region of response will be simulated in future work. Further, simulations using correlated development rate noise should show whether kinetic roughness during development dominates the final lithographic roughness or whether the underlying development rate noise, coming from earlier stochastic processes such as exposure and reaction-diffusion, controls the final surface characteristics. Last, a thorough understanding of the uniform, open-frame development case will lead the way to the more difficult and interesting case of roughness formation during development in the presence of a steep development rate gradient, as is found at the edge of a photoresist line, and low mean development rates. Of course, experimental data with which to compare these simulation results is needed and should also be furnished in the future.

References

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