

SIZE DISTRIBUTIONS AND SCALING IN HETEROGENEOUS NUCLEATION WITH SIZE-LINEAR RATE CONSTANTS

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Abstract. We present a theoretical analysis of rate equations for heterogeneous nucleation of nanomaterials with linear size dependences of the aggregation and fragmentation rate constants. Two scenarios are considered, one relating to stable growth and the other describing unstable situation with a time-dependent critical size. An interesting analytical solution is obtained which is exact in the stable case and only asymptotic in the unstable growth. This solution is expressed through the Polya distribution. Its continuum form features scaling properties for all but very small sizes, which is an intrinsic property of the model. Our scaled size distribution is capable of reproducing both monomodal and monotonically decreasing shapes depending on the value of the dimerization constant. The obtained solution is shown to reproduce fairly well some experimental size spectra of linear chains of metal adatoms on Si(100) surfaces.

1. Introduction

Theoretical analysis of the size distributions (SDs) of different “clusters” (droplets, nanopores, surface islands, nanowires, linear rows) is paramount from the fundamental viewpoint as well as for controlling physical and mechanical properties of nanomaterials. Nucleation of clusters from a metastable mother phase (supersaturated three-dimensional vapor or two-dimensional “sea” of adatoms) can occur either homogeneously, i.e. with all the clusters consisting of only one type of “monomers”, or heterogeneously, where the clusters form on a foreign nucleation centers (charged ions, impurities, surface defects *etc.*). In two-dimensional systems such as solid surfaces at submonolayer coverage, new adatoms are continuously brought to the system from a vapor flux. At very high degree of supersaturation of a mother phase (for example, for surface layers at a low temperature), the critical size of classical nucleation theory approaches one and the entire growth process proceeds almost irreversibly before the coalescence stage. In this case, the rate equations (REs) for completely irreversible growth or irreversible growth above a specified and time-independent critical size can be applied [1-21], with the effective capture coefficients (more generally – rate constants) depending on the cluster size and coverage. At lower supersaturations, decay of clusters becomes important and should be accounted for in the corresponding REs, as in classical nucleation theory [22-26]. Discrete REs comprise infinite set of the connected non-linear differential equations. Consequently, despite the important inputs from mathematical community (see, e.g., Refs. [27, 28]), the exactly solvable cases [4, 15, 29-31] are rare.

One of the most interesting properties of these REs (usually considered in the case of

2. Rate equations of heterogeneous nucleation

We consider a heterogeneous growth process involving aggregation and fragmentation of clusters via the scheme $A_s B + A_1 \leftrightarrow A_{s+1} B$ with $s = 0, 1, 2, 3, \dots$. Here, B denotes the nucleation center (such as a charged ion in vapor or reactive defect on a solid surface), A_1 is the free monomer and $A_s B$ is the cluster consisting of s monomers and one nucleation center. By definition, $A_0 B \equiv B$ is the free seed. The set of REs for the concentrations of free monomers $n_A(t) = [A_1]_t$ and clusters $n_s(t) = [A_s B]_t$ (at $i = 0$, $n_0(t) = [B]_t$ is the concentration of free nucleation centers) is given by

$$\begin{aligned} \frac{dn_A}{dt} &= -\sum_{s=1}^{\infty} J_s + R; \\ \frac{dn_s}{dt} &= J_s (1 - \delta_{s0}) - J_{s+1}, \quad s \geq 0; \\ J_s &= K_s^+ n_A n_{s-1} - k_s^- n_s, \quad s \geq 1. \end{aligned} \quad (1)$$

Here, K_s^+ is the aggregation rate constant, k_s^- is the fragmentation rate constant, J_s is the flux along the size axis showing how many clusters are transferred from the state with the size $s-1$ to s per unit time, and $R = \text{const}$ is the external influx of monomers A_1 . Kroneker's symbol δ_{s0} disables the decay of nucleation centers B . The first equation shows that the concentration of free monomers changes whenever a monomer is attached to or detached from a cluster, and due to the material influx. According to the second equation, the concentration of clusters of a given size changes in time due to the difference between the two fluxes going into and from the state with the size s and becomes time-independent at equilibrium ($J_s = 0$) or in a steady state (i.e., when $J_s = J$ is size-independent). Clearly, the equilibrium state can be reached only in systems with fragmentation, while for irreversible growth $J_s = K_{s-1}^+ n_A n_{s-1}$ can become zero only in a materially isolated system when $n_A \rightarrow 0$.

REs given by Eqs. (1) yield the following conservation laws:

$$\sum_{s=0}^{\infty} n_s(t) = n_B^{\text{tot}}; \quad n_A(t) + \sum_{s=1}^{\infty} s n_s(t) = n_A(0) + Rt. \quad (2)$$

The first equation gives the conservation of total number of nucleation centers, and the second one shows that the total number of monomers increases with time only due to the external flux. As the major simplification within this heterogeneous growth model, the total number of all clusters plus the number of free nucleation centers equals the total number of nucleation centers $n_B^{\text{tot}} = \text{const}$ at any time. This feature is distinctly different from homogenous nucleation where the total number of clusters is not conserved [26]. It is therefore convenient to consider the normalized size distribution $f_s(t) = n_s(t) / n_B^{\text{tot}}$ for which $\sum_{s \geq 0} f_s(t) = 1$, and the normalized monomer concentration $f_A(t) = n_A(t) / n_B^{\text{tot}}$. The aggregation rate constants can be put in the form $K_i^+ = k_i^+ n_B^{\text{tot}}$, where k_i^+ have the same dimension (of the inverse time) as k_i^- .

As in Ref. [31], we now assume that the rate constants of whatever origin depend linearly on the number of monomers in the cluster:

This ζ depends on time and can have either signs. The mean size of clusters, $\langle s \rangle = \sum_{s \geq 1} s f_s$, obeys the equation

$$\frac{d \langle s \rangle}{d\tau} = \zeta \langle s \rangle + (1-a)(\zeta + 1)(qf_0 - 1), \quad q \equiv (b+1-a)/(1-a). \quad (5)$$

The material balance of monomers given by Eq. (2) can be put in the form

$$\Theta = \langle s \rangle + (k^- / k^+) \zeta. \quad (6)$$

The appropriately normalized coverage, $\Theta = (k^- / k^+) \zeta(0) + (R\tau)/(n_B^{tot} k^-)$, equals the so-called ‘‘ideal supersaturation’’ in the absence of any clustering, i.e., when $\langle s \rangle = 0$ [26]. The coverage increases linearly with time in open systems and is conserved in isolated systems with $R=0$. In principle, Eqs. (5) and (6) allow for the determination of the mean size and supersaturation as functions of time. At the beginning of the growth process, the system does not contain any clusters, i.e. $f_0(\tau=0)=1$ and $f_s(\tau=0)=0$ for all $s \geq 1$, yielding also $\langle s \rangle(\tau=0)=0$.

We note that the system considered is expected to have rather different behaviors depending on whether $a > 1$ or $a < 1$. Indeed, the growth rate of a cluster consisting of s monomers can be generally defined as $ds/dt = k_s^+ f_A - k_s^-$. With our size-linear rate constants given by Eqs. (3), this is reduced to $ds/d\tau = (a-1)(\zeta + 1) + \zeta s$. At $a > 1$, the cluster growth rate is positive for all s at $\zeta > 0$. Hence, a positive supersaturation yields an unlimited growth of clusters regardless of their size. This is effectively the irreversible growth case studied in Refs. [4, 5, 12, 16]. When $\zeta < 0$, the growth rate becomes zero at a stable point such that smaller clusters would grow and larger clusters decay, corresponding to a limited stable growth. Whenever $a < 1$, a positive supersaturation leads to the growth instability such that smaller clusters decay and larger clusters grow, as in nucleation theory with a supersaturation-dependent critical size [22-26]. In our model, this critical size corresponds to the condition $ds/d\tau = 0$ and equals $i = (1-a)(\zeta + 1)/\zeta$. When $\zeta < 0$, all the clusters would decay regardless of their size. Consequently, we will call the situation with $a > 1$ the stable growth (which can be either unlimited or limited depending on the monomer influx), while the case $a < 1$ will be referred to as the unstable growth. The mathematical difficulty in the unstable case arises from the fact that the parameter q in Eq. (5) cannot be put to zero and hence the mean size depends on the concentration of free nucleation centers f_0 . The latter is generally unknown and requires a solution of the entire infinite set of the REs.

3. Analytical solutions

We start the analysis with the general case of arbitrary positive a and b . As in Refs. [4, 26, 27, 30, 31], we introduce the generating function for the discrete SD by definition

$$f(x, \tau) = \sum_{s=0}^{\infty} f_s(\tau) x^s, \quad (7)$$

having the obvious properties $f(0, \tau) = f_0(\tau)$ and $f(1, \tau) = 1$. Differentiating Eq. (7) with respect to time and using Eqs. (4), we obtain the first-order partial differential equation for $f(x, \tau)$:

$$\frac{\partial f}{\partial \tau} = (x-1) \left\{ [(\zeta + 1)x - 1] \frac{\partial f}{\partial x} - (1-a)(\zeta + 1)f + (b+1-a)(\zeta + 1)f_0 \right\};$$

corresponding approximate solution has the form

$$F(x, \tau) \cong [1 + \alpha(\tau)(1-x)]^{1-a} [1 - qf_0(\tau)]. \quad (16)$$

Using the normalization condition, we arrive at the following SD

$$f_0(\tau) = \frac{[1 + \alpha(\tau)]^{1-a}}{1 + q[1 + \alpha(\tau)]^{1-a} - q}; \quad f_s(\tau) = \frac{\langle s \rangle(\tau)}{s} f_{s-1}^{(a)}[\alpha(\tau)], \quad s \geq 1. \quad (17)$$

Here,

$$\langle s \rangle(\tau) = \frac{b\alpha(\tau)}{1 + q[1 + \alpha(\tau)]^{1-a} - q}, \quad (18)$$

is the mean size of the approximate SD. Here and below, we use the notation

$$f_s^{(c)}[\alpha] = \frac{\Gamma(c+s)}{\Gamma(c)s!} \frac{\alpha^s}{(1+\alpha)^{c+s}}, \quad s \geq 0 \quad (19)$$

for the Polya distribution with the mean size $\langle s \rangle = c\alpha$, where $\Gamma(y)$ denotes the gamma-function.

This SD has the following important properties:

1) In the stable growth with $a > 1$, we can put

$$b = a - 1, \quad (20)$$

corresponding to $q = 0$ and $k_s^+ = k^+(b+s)$ for all $s = 0, 1, 2, \dots$, i.e., a purely linear size dependence of the aggregation rate constants. In this case, Eqs. (17) are reduced to the exact solution which has the form of the Polya distribution with the parameter $b > 0$ and the mean size $b\alpha$ [31]:

$$f_s(\tau) = f_s^{(b)}[\alpha] = \frac{\Gamma(b+s)}{\Gamma(b)s!} \frac{\alpha^s}{(1+\alpha)^{b+s}}, \quad s \geq 0, \quad \langle s \rangle = b\alpha. \quad (21)$$

This can be represented in terms of the mean size $\langle s \rangle$ as

$$f_s^{(b)}[\langle s \rangle] = \left(1 + \frac{\langle s \rangle}{b}\right)^{-b} \frac{\Gamma(b+s)}{\Gamma(b)s!} \left(\frac{\langle s \rangle}{\langle s \rangle + b}\right)^s. \quad (21a)$$

In the formal limit $b \rightarrow \infty$, for which the rate constants become size-independent, the Polya distribution is reduced to the Poissonian SD

$$f_s[\langle s \rangle] = e^{-\langle s \rangle} \frac{\langle s \rangle^s}{s!}. \quad (22)$$

2) In the unstable case with $a < 1$, it can be shown that the approximate solution given by Eqs. (17) matches exactly the equilibrium SD at $\tau \rightarrow \infty$ in a closed system ($\Theta = \text{const}$) or upon the flux termination ($\Theta \rightarrow \Theta_{\max}$). Of course, the equilibrium solution exists only when the final supersaturation is negative ($\zeta \rightarrow \zeta_{\infty} < 0$).

3) By its form, Eq. (17) might seem to be a discrete analogue of the continuum Lifshitz-Slyozov transformation of variables if the mean size equals or is proportional to the critical size: $\langle s \rangle = \zeta_i$ [32]. Indeed, regardless of the particular form of the normalized SD in the

$$\Theta = \frac{\tau}{\tau_0}, \quad (23)$$

where τ_0 is the macroscopic time related to the influx R in Eq. (1) (e.g., vapor flux for surface clusters) as $1/\tau_0 = R/(k^-n_B^{tot})$. From the material balance given by Eq. (6) and assuming $\zeta \rightarrow 0$, we get: $\langle s \rangle \cong \Theta = \tau/\tau_0$, i.e., the mean size asymptotically equals the coverage and increases linearly with time due to the monomer influx. Using this in Eq. (5) at $q=0$, $b=a-1>0$ and $\zeta \rightarrow 0$, and then in Eq. (15), we obtain

$$\zeta \cong \frac{1-b\tau_0}{\tau}; \quad \alpha = \frac{\langle s \rangle}{b} \cong \frac{\tau}{b\tau_0}. \quad (24)$$

Therefore, supersaturation scales with time as $1/\tau$ and remains infinitely small positive when $b\tau_0 < 1$, while α increases linearly with time along with the mean size.

It is noteworthy that the approximate solution given by Eqs. (17) in the unstable case cannot hold if the material influx remains on at $\tau \rightarrow \infty$. Indeed, at $\Theta \rightarrow \infty$ we should also observe $\alpha \rightarrow \infty$. From Eq. (18) at $\alpha \gg 1$ it then follows that

$$\langle s \rangle \cong \frac{b}{q} \alpha^a, \quad (25)$$

showing that the mean size increases slower than α . Further, from Eq. (17) for f_0 at $\alpha \gg 1$ we get

$$f_0 \cong \frac{1}{q} \left[1 + \frac{(q-1)}{q} \alpha^{a-1} \right]. \quad (26)$$

For the non-trivial $(1-a)(\zeta+1)(qf_0-1)$ term in Eq. (5) for the mean size, this yields $(1-a)(\zeta+1)(qf_0-1) \cong (b/q)(\zeta+1)\alpha$. Using this and substituting Eq. (25) into Eq. (5), we arrive at

$$a \frac{d\alpha}{d\tau} = \zeta\alpha + \zeta + 1. \quad (27)$$

Obviously, this is consistent with Eq. (15) only when $d\alpha/d\tau \rightarrow 0$, so both α and $\langle s \rangle$ must saturate at $\tau \rightarrow \infty$, which necessarily requires $\zeta \rightarrow \zeta_\infty < 0$. Asymptotic properties of the unstable system with a continuous influx require a separate study which will be presented elsewhere.

5. Continuum size distributions and scaling

Using the Stirling formula and the asymptote $(\alpha/(1+\alpha))^s = \exp(-s/\alpha)$ for large s and α , we arrive at the continuum form of the Poly distribution at $s \gg 1$

$$f^{(c)}(s, \alpha) = \frac{s^{c-1}}{\Gamma(c)\alpha^c} \exp(-s/\alpha). \quad (28)$$

This continuum SD obeys the normalization condition and preserves the correct mean size:

$$\int_0^\infty ds f^{(c)}(s, \alpha) = 1; \quad \int_0^\infty ds s f^{(c)}(s, \alpha) = c\alpha. \quad (29)$$

$s/\langle s \rangle$, as discussed in Ref. [16], while the formal limit at $\langle s \rangle \rightarrow \infty$ yields the non-analytic Bartelt-Evans scaling [4] for both small and large sizes.

In the unstable case with $a < 1$, in situations described by the approximate solutions given by Eqs. (15) (we already know that these expressions apply near equilibrium for isolated systems and do not apply under the monomer influx), we can still use Eq. (26) at $s \gg 1$ and $\alpha \gg 1$. Using Eq. (26) at $c = a$ in Eq. (15) and then Eq. (23), we obtain

$$f(s, \langle s \rangle) = \frac{1}{\langle s \rangle^{2-a}} \frac{p}{\Gamma(a)} \left(\frac{s}{\langle s \rangle} \right)^{a-2} \exp\left(-p^{1/a} \frac{s}{\langle s \rangle^{1/a}} \right), \quad (34)$$

with $p = b/q$. It is seen that this function does not have a scaling form in terms of the scaled cluster size $s/\langle s \rangle$. Indeed, the scaled SD is given by

$$f(s, \langle s \rangle) \langle s \rangle = \frac{p}{\Gamma(a)} \frac{1}{\langle s \rangle^{1-a}} x^{a-2} \exp\left(-\frac{p^{1/a}}{\langle s \rangle^{1/a-1}} x \right), \quad (34a)$$

which is not a unique function of $x = s/\langle s \rangle$ and tends to a singularity at $x = 0$ when $\langle s \rangle \rightarrow \infty$ (see Fig. 4). The absence of scaling is explained by the non-linear relation between $\langle s \rangle$ and α given by Eq. (23). As for the Lifshitz-Slyozov theory of ripening [32], it requires that ζ at the asymptotic stage is small positive. In the linear model, this occurs only in systems with continuous influx where our approximate solution is not valid, as discussed above. In a closed system, supersaturation tends to a negative value and thus the ripening process is disabled.

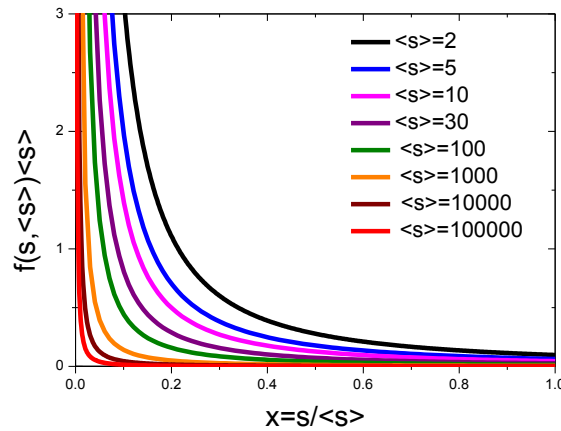


Fig. 4. Evolution of the scaled SD $f(s, \langle s \rangle) \langle s \rangle$ in the case $a = b = 1/2$ showing the absence of scaling in the unstable case with $a < 1$.

6. Fitting experimental size distributions

Let us now see how the obtained analytical solutions can be used for fitting the experimental data on different one-dimensional surface chains. For surface islands, assuming a heterogeneous character of nucleation, the growth rate constants would have the form $k_s^+ = n_{tot} D \sigma_s$, where n_{tot} is the surface concentration of the nucleation centers (e.g., surface steps [33] or defects [17]), D is the diffusion coefficient of a mobile adatom, and σ_s is the size-dependent capture number. Linear surface rows are formed rather than islands if the disusing adatoms can bond with rows only at the two end sites [7].

In Ref. [7], room temperature deposition of Ga onto Si(100) with a deposition rate 10^{-3} monolayers (MLs) per second produced single-atom-wide Ga rows which lie orthogonal to

curves demonstrate that the scaled SDs for both In coverages are purely decreasing, with a very high population near $x = 1$. The best fit by Eqs. (31) and (32) is obtained, however, at $b = 2.3$, and has a maximum at $x \cong 0.6$, and thus we are unable to describe correctly the two maximum points at the 0.08 ML coverage. The discrepancy between the theoretical fit and the data might be due to the postdeposition relaxation, as discussed by the authors [17]. Overall, the fits shown in Figs. 5 and 6 are quite reasonable, however, they require the assumption on the heterogeneous character of nucleation.

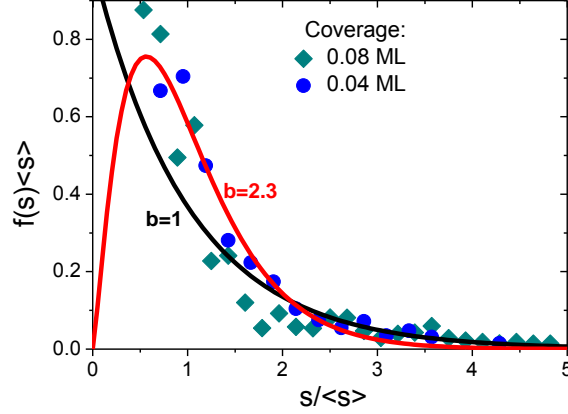


Fig. 6. Scaled size distributions of In chains on Si(100)- 2×1 at two different coverages [17], and their best fit by Eqs. (31) and (32) at $b = 2.3$ (red line). The black line at $b = 1$ corresponds to the exponential fit.

The role of heterogeneous versus homogeneous nucleation of metal surface rows on Si surfaces such as described in Refs. [7, 17, 39] remains an open question. In particular, Albao *et al.* [41] argued and presented some evidence that growth of Ga chains on Si(100) is not dominated by heterogeneous nucleation at C defects, contrasting the proposal of Kocán *et al.* [39] and the assumption of Ref. [17]. Probability of heterogeneous nucleation should depend on concentration of the reactive defects. As demonstrated theoretically in Ref. [42], concentration of the defects larger than 0.0025 per site may change the island SDs from monomodal to monotonically decreasing in the case of the irreversible growth model. Fragmentation of islands in systems close to equilibrium has been also found important for the SD shapes [42-44]. For example, the results of Ref. [43] suggest that the scaling functions are monotonously decreasing in quasi-equilibrium case with fragmentation, while in the case of irreversible growth they exhibit a monomodal character.

A comparison of heterogeneous versus homogeneous irreversible growths with size-linear capture coefficients will be presented elsewhere. We note, however, that the homogeneous irreversible growth model with size-linear capture coefficients $k_s^+ = k^+(1+s)$ for all s yields scaling in the form of Eq. (31) with the scaling function $\varphi_1(x) = \exp(-x)$, which is a particular case of Eq. (32) at $b = 1$. Thus, the scaling functions of homogeneous and heterogeneous SDs at $b = 1$ are exactly identical (which is of course not the case for arbitrary b). These exponential fits are shown in Figs. 5 (b) and 6 for comparison with the scaled heterogeneous SDs at $b \neq 1$. It is seen that the homogeneous exponential scaling function also provides a good fit to the scaled experimental SDs, especially in the case of Ga rows (Fig. 5 (b)). This is not surprising, because the fitting values of b are close to one.

7. Conclusion

In conclusion, we have presented the exactly solvable case of the stable heterogeneous nucleation including decay of clusters, with size-linear rate constants for aggregation and

fragmentation. The obtained SDs are given by the Polya distribution whose shape is controlled by the dimensionless dimerization constant b . Despite its simplicity, this SD has a rather general form and is capable of describing a monotonically decreasing SDs at $b \leq 1$ and monomodal SDs with well-defined peak at $b > 1$. Similar distribution shapes apply for irreversible growth with no decay, in both isolated or open systems, and also when desorption is included (this would only change the kinetics of the mean size). Very importantly, the continuum form of the Polya SD automatically leads to scaling in terms of the $s/\langle s \rangle$ variable. The model SDs have been shown to fit fairly well some experimental data for linear surface rows of metal adatoms on Si(100). The particular case of heterogeneous scaling function at $b = 1$ is exponential, and is identical to the case of homogeneous irreversible growth.

In the unstable model with a time-dependent critical size, we have obtained the asymptotic solution which applies near the equilibrium state in a closed system or upon termination of the influx. This solution does not yield a scaling form of the SD, because the mean size increases more slowly due to enhanced fragmentation of clusters. However, our approximate SD does not work for systems with a continuous influx, for example, in the case of surface islands deposited from a time-independent flux. Here, non-trivial effects such as breaking the SD into stable and irreversibly growing branches near the critical size and the asymptotic ripening stage are anticipated [32, 45, 46], which remain to be studied.

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