Quantum ring growth simulation algorithm

April 9, 2025

1 Main equations

Diffusion equations for Ga and As. $R_d\left(t\right)$ is the droplet radius.

$$\frac{\partial C_{Ga}}{\partial t}=D_{Ga}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C_{Ga}}{\partial r}\right)+F_{Ga}\left(r,t\right)-k_rC_{Ga}C_{As} \tag{1}$$

$$\frac{\partial C_{As}}{\partial t}=D_{As}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C_{As}}{\partial r}\right)+F_{As}-k_rC_{Ga}C_{As}-\frac{C_{As}}{\tau_{As}} \tag{2}$$

Parameters in detail

$$D_{Ga}=a_0^2\nu\exp\left(-\frac{E_{Ga}}{kT}\right) \tag{3}$$

$$D_{As}=a_0^2\nu\exp\left(-\frac{E_{As}}{kT}\right) \tag{4}$$

Arsenic desorption time:

$$\tau_{As}=\frac{1}{\nu}\exp\left(\frac{E_a}{kT}\right) \tag{5}$$

$$\nu=\frac{kT}{\pi\hbar} \tag{6}$$

Note that temperature kT and the As flux F_{As} are free, user-defined parameters.

Parameter	Value	Units
k_r	0.1	cell^2/time
a_0	1	cell
h_0	1	cell
Ω_{GaAs}	1	cell^3
Ω_{Ga}	0.1	cell^3
C_0	2	cell^{-2}
$E_{Ga} - E_{As}$	0	eV
$E_a - E_{Ga}$	0.1	eV

Table 1: Numerical values of the parameters

1.1 Numerical values

Let's set the distance variable in such a way that:

$$a_0 = 1$$

$$\Omega_{GaAs} = 1$$

Then concentrations are per lattice cell. Droplet radius is measured in cells. Flux is per cell per unit time. Time units we set in such a way that:

$$\nu \exp\left(-\frac{E_{Ga}}{kT}\right) = 1$$

$$D_{Ga} = 1$$

$$D_{As} = \exp\left(-\frac{E_{As} - E_{Ga}}{kT}\right)$$

$$\tau_{As} = \exp\left(\frac{E_a - E_{Ga}}{kT}\right)$$

1.2 Boundary and initial conditions

Boundary conditions for $r \in [0, R_\infty]$:

$$C_{Ga}\left(R_\infty, t\right) = 0$$

$$C_{As}\left(R_\infty, t\right) = F_{As}\tau_{As}$$

Initial conditions for $r \in [0, R_\infty]$:

$$C_{Ga}\left(r, 0\right) = 0$$

$$C_{As}\left(r, 0\right) = 0$$

2 Finding the Ga flux

We require an additional concentration of Ga atoms at the droplet boundary according to:

$$\tilde{C}_{Ga}\left(r, t\right) = C_0 \exp\left(-\frac{\left(r - R_d\left(t\right)\right)^2}{w^2}\right) \tag{7}$$

This leads to an additional flux:

$$F_{Ga}\left(r, t\right) = \frac{C_0}{\tau_{Ga}\left(t\right)} \exp\left(-\frac{\left(r - R_d\left(t\right)\right)^2}{w^2}\right) \tag{8}$$

Which is our only source of Ga atoms! To find the parameter τ_{Ga} (w is a free constant value), we need to account for the loss of Ga atoms. In a lengthy derivation, which we omit here, we obtained:

$$\tau_{Ga} = \frac{w^2}{2D_{Ga}}\left[\exp\left(-\frac{R_d^2}{w^2}\right) - \exp\left(-\frac{\left(R_\infty - R_d\right)^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{\operatorname{erf}\left(\frac{R_\infty - R_d}{w}\right) + \operatorname{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln\frac{R_\infty}{R_d} \tag{9}$$

If $R_\infty \gg R_d$, then this equation simplifies:

$$\tau_{Ga} = \frac{w^2}{2D_{Ga}}\left[\exp\left(-\frac{R_d^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{1 + \operatorname{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln\frac{R_\infty}{R_d} \tag{10}$$

When $R_d \to 0$, then naturally $\tau_{Ga} \to +\infty$ and $F_{Ga} \to 0$.
So we obtain:

$$F_{Ga}(r,t) = \frac{2D_{Ga}C_0}{w^2} \frac{\exp\left(-\frac{(r-R_d(t))^2}{w^2}\right)}{\left[\exp\left(-\frac{R_d^2}{w^2}\right) - \exp\left(-\frac{(R_\infty - R_d)^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{\operatorname{erf}\left(\frac{R_\infty - R_d}{w}\right) + \operatorname{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln\frac{R_\infty}{R_d}} \tag{11}$$

Note that if we define the variables:

$$x = \frac{R_d}{w}, \qquad p = \frac{R_\infty}{w}$$

Then the function:

$$q(x,p) = \exp\left(-x^2\right) - \exp\left(-(p-x)^2\right) + \sqrt{\pi}x\left\{\operatorname{erf}(p-x) + \operatorname{erf}(x)\right\}$$

Can be approximated by a straight line:

$$q(x,p) \approx a(p)\,x + b(p)$$

Where:

$$b(p) \approx \frac{0.187}{p - 3.156}$$

$$a(p) \approx 3.545$$

So we obtain:

$$F_{Ga}(r,t) = \frac{2D_{Ga}C_0}{w^2} \frac{\exp\left(-\frac{(r-R_d(t))^2}{w^2}\right)}{\left[3.545\frac{R_d}{w} + \frac{0.187w}{R_\infty - 3.156w}\right] \ln\frac{R_\infty}{R_d}} \tag{12}$$

3 Droplet geometry

Droplet height H, droplet contact angle θ. Where θ is the contact angle, which could be between 30° and 80°.
Form-factor:

$$B(\theta) = \frac{8 - 9\cos\theta + \cos 3\theta}{3\sin\theta - \sin 3\theta} \tag{13}$$

$$\frac{dR_d}{dt} = \frac{\Omega_{Ga}}{\pi B\left(\theta\right) R_d^2} \frac{dN_{Ga}}{dt} \tag{14}$$

$$\frac{d}{dt} \frac{R_d}{R_\infty} = -\frac{2\Omega_{Ga} D_{Ga} C_0}{B\left(\theta\right) R_\infty^3} \frac{R_\infty^2}{R_d^2 \ln \frac{R_\infty}{R_d}} \tag{15}$$

$$\frac{R_d}{R_\infty} = \rho < 1$$

$$\frac{2\Omega_{Ga} D_{Ga} C_0}{B\left(\theta\right) R_\infty^3} = \sigma \tag{16}$$

$$\frac{d\rho}{dt} = \frac{\sigma}{\rho^2 \ln \rho} \tag{17}$$

But this ODE can be easily solved:

$$\rho^2 \ln \rho d\rho = \sigma dt \tag{18}$$

$$\frac{\rho^3}{9} \left(\ln \rho^3 - 1\right) = \sigma t + C \tag{19}$$

The initial state is a known value:

$$\rho\left(0\right) = \rho_0 < 1 \tag{20}$$

$$C = \frac{\rho_0^3}{9} \left(\ln \rho_0^3 - 1\right) \tag{21}$$

We obtain the following equation for droplet radius:

$$\rho^3 \left(\ln \rho^3 - 1\right) = \rho_0^3 \left(\ln \rho_0^3 - 1\right) + 9\sigma t \tag{22}$$

Now this can be solved by Newton's method for each time.

$$p\left(\rho\right) = \rho^3 \left(3 \ln \rho - 1\right) - \rho_0^3 \left(3 \ln \rho_0 - 1\right) - 9\sigma t \tag{23}$$

$$p'\left(\rho\right) = 3\rho^2 \left(\ln \rho^3 - 1\right) - 3\rho^2 = 3\rho^2 \left(3 \ln \rho - 2\right) \tag{24}$$

We take the first guess for t_{i+1} as:

$$\rho_{i+1}^{(0)} = \rho_i$$

Then perform Newton iterations until the value stops changing:

$$\rho_{i+1}^{(k+1)} = \rho_{i+1}^{(k)} - \frac{\left(\rho_{i+1}^{(k)}\right)^3 \left(3\ln\rho_{i+1}^{(k)} - 1\right) - \rho_0^3 \left(3\ln\rho_0 - 1\right) - 9\sigma t_i}{3\left(\rho_{i+1}^{(k)}\right)^2 \left(3\ln\rho_{i+1}^{(k)} - 2\right)} \tag{25}$$

Note that we should always have:

$$0 < \rho < 1 \tag{26}$$

4 Finite difference scheme

We introduce grid in the following form:

$$r_j = j\Delta r, \quad j = 0, 1, \ldots, N_r, \quad N_r \Delta r = R_\infty$$

$$t_i = i\Delta t, \quad i = 0, 1, \ldots, N_t, \quad N_t \Delta t = T$$

Time limit T should be chosen from the condition of total droplet depletion $R_d\left(T\right) \leq a$.
Using the previous section:

$$T = \frac{\rho^3\left(\ln\rho^3 - 1\right) - \rho_0^3\left(\ln\rho_0^3 - 1\right)}{9\sigma} \tag{27}$$

4.1 Euler scheme

We can use Euler's method:

$$\frac{dx}{dt} = f \longrightarrow \frac{x_{i+1} - x_i}{\Delta t} = f_i \longrightarrow x_{i+1} = x_i + \Delta t f_i$$

The equations become:

$$\text{for } j = 0$$

$$C_{Ga}^{i+1,0} = C_{Ga}^{i,0} + \Delta t \left(\frac{D_{Ga}}{\Delta r^2} \left[4C_{Ga}^{i,1} - 4C_{Ga}^{i,0} \right] + F_{Ga}^{i,0} - k_r C_{Ga}^{i,0} C_{As}^{i,0} \right)$$

$$C_{As}^{i+1,0} = C_{As}^{i,0} + \Delta t \left(\frac{D_{As}}{\Delta r^2} \left[4C_{As}^{i,1} - 4C_{As}^{i,0} \right] - \frac{C_{As}^{i,0}}{\tau_{As}} + F_{As} - k_r C_{Ga}^{i,0} C_{As}^{i,0} \right)$$

$$\text{for } j \in [1, N_r]$$

$$C_{Ga}^{i+1,j} = C_{Ga}^{i,j} + \Delta t \left(\frac{D_{Ga}}{\Delta r^2} \left[\left(1 + \frac{1}{2j}\right) C_{Ga}^{i,j+1} - 2C_{Ga}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{Ga}^{i,j-1} \right] + F_{Ga}^{i,j} - k_r C_{Ga}^{i,j} C_{As}^{i,j} \right)$$

$$C_{As}^{i+1,j} = C_{As}^{i,j} + \Delta t \left(\frac{D_{As}}{\Delta r^2} \left[\left(1 + \frac{1}{2j}\right) C_{As}^{i,j+1} - 2C_{As}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{As}^{i,j-1} \right] - \frac{C_{As}^{i,j}}{\tau_{As}} + F_{As} - k_r C_{Ga}^{i,j} C_{As}^{i,j} \right)$$

$$\text{for } j = N_r - 1$$

$$C_{Ga}^{i,N_r} = 0, \qquad C_{As}^{i,N_r} = F_{As} \tau_{As}$$

We can divide all the equations by C_0 and further assume that we are measuring all concentrations in units of C_0.

$$C_{Ga}^{i+1,j} = C_{Ga}^{i,j} + \Delta t \left(\frac{D_{Ga}}{\Delta r^2} \left[\left(1 + \frac{1}{2j}\right) C_{Ga}^{i,j+1} - 2C_{Ga}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{Ga}^{i,j-1} \right] + \frac{F_{Ga}^{i,j}}{C_0} - C_0 k_r C_{Ga}^{i,j} C_{As}^{i,j} \right)$$

$$C_{As}^{i+1,j} = C_{As}^{i,j} + \Delta t \left(\frac{D_{As}}{\Delta r^2} \left[\left(1 + \frac{1}{2j}\right) C_{As}^{i,j+1} - 2C_{As}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{As}^{i,j-1} \right] - \frac{C_{As}^{i,j}}{\tau_{As}} + \frac{F_{As}}{C_0} - C_0 k_r C_{Ga}^{i,j} C_{As}^{i,j} \right)$$

Introducing parameters:

$$\frac{D_{Ga} \Delta t}{\Delta r^2} = \alpha$$

$$\frac{D_{As} \Delta t}{\Delta r^2} = \beta$$

$$C_0 k_r \Delta t = \omega$$

$$\frac{\Delta r^2}{w^2} = \eta$$

$$\frac{\Delta t F_{Ga}^{i,j}}{C_0} = \frac{2\Delta t D_{Ga}}{w^2} \frac{\exp\left(-\frac{\left(r-R_d^i\right)^2}{w^2}\right)}{\left[3.545\frac{R_d^i}{w} + \frac{0.187w}{R_\infty - 3.156w}\right] \ln\frac{R_\infty}{R_d^i}} = 2\alpha\eta f_{i,j} = \upsilon f_{i,j}$$

$$2\alpha\eta = \upsilon$$

$$f_{i,j} = \frac{\exp\left(-\frac{\left(r-R_d^i\right)^2}{w^2}\right)}{\left[3.545\frac{R_d^i}{w} + \frac{0.187w}{R_\infty - 3.156w}\right] \ln\frac{R_\infty}{R_d^i}}$$

$$\frac{\Delta t F_{As}}{C_0} = \kappa$$

$$\frac{\Delta t}{\tau_{As}} = \gamma$$

$$\frac{F_{As}\tau_{As}}{C_0} = \epsilon$$

The equations become:

$$\text{for } j = 0$$

$$C_{Ga}^{i+1,0} = C_{Ga}^{i,0} + 4\alpha\left[C_{Ga}^{i,1} - C_{Ga}^{i,0}\right] + \upsilon f_{i,j} - \omega C_{Ga}^{i,0} C_{As}^{i,0}$$

$$C_{As}^{i+1,0} = C_{As}^{i,0} + 4\beta\left[C_{As}^{i,1} - C_{As}^{i,0}\right] - \gamma C_{As}^{i,0} + \kappa - \omega C_{Ga}^{i,0} C_{As}^{i,0}$$

$$\text{for } j \in [1, N_r]$$

$$C_{Ga}^{i+1,j} = C_{Ga}^{i,j} + \alpha\left[\left(1 + \frac{1}{2j}\right) C_{Ga}^{i,j+1} - 2C_{Ga}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{Ga}^{i,j-1}\right] + \upsilon f_{i,j} - \omega C_{Ga}^{i,j} C_{As}^{i,j}$$

$$C_{As}^{i+1,j} = C_{As}^{i,j} + \beta \left[\left(1 + \frac{1}{2j}\right) C_{As}^{i,j+1} - 2C_{As}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{As}^{i,j-1} \right] - \gamma C_{As}^{i,j} + \kappa - \omega C_{Ga}^{i,j} C_{As}^{i,j}$$

$$\text{for } j = N_r - 1$$

$$C_{Ga}^{i,N_r} = 0, \qquad C_{As}^{i,N_r} = \epsilon$$

5 Ring growth speed

Let's derive the expression for the QR height $h(r,t)$, which defines the QR profile at each moment in time and at each distance from the droplet center.

Concentration of both types of atoms bound together and contributing to the crystal growth changes according to:

$$\frac{dC_{\text{bound}}}{dt} = k_r C_{Ga} C_{As}$$

The number of GaAs vertical crystal cells at each distance increases according to:

$$C_{\text{bound}} = N_{\text{cells}} C_0$$

$$\frac{dN_{\text{cells}}}{dt} = \frac{k_r}{C_0} C_{Ga} C_{As}$$

The height of the layer is connected to this number of cells:

$$h = h_0 N_{\text{cells}}$$

$$\frac{dh(r,t)}{dt} = \frac{h_0 k_r}{C_0} C_{Ga} C_{As}$$

Considering the scaling of the concentrations by C_0 and the introduced parameters, we write:

$$\frac{dh(r,t)}{dt} = h_0 C_0 k_r C_{Ga} C_{As}$$

$$h_{i+1,j} = h_{i,j} + \Delta t h_0 C_0 k_r C_{Ga}^{i,j} C_{As}^{i,j}$$

Finally, we obtain:

$$h_{i+1,j} = h_{i,j} + \omega h_0 C_{Ga}^{i,j} C_{As}^{i,j} \tag{28}$$

The initial condition is clearly:

$$h_{0,j} = 0 \tag{29}$$

6 Droplet radius change dependent on QR growth

We again use the concentration change:

$$\frac{dC_{\text{bound}}}{dt} = k_r C_{Ga} C_{As}$$

This should be the only way for Ga atoms to disappear (except for the ones going beyond the boundary, but we will discard them for now). Note that this surface concentration is a function of distance, so we should integrate to get the full number of Ga atoms disappearing:

$$\frac{dN_{Ga}}{dt} = -2\pi k_r C_0^2 \int_0^{R_\infty} r C_{Ga} C_{As} dr \tag{30}$$

Using the previous formula:

$$\frac{dR_d}{dt} = \frac{\Omega_{Ga} C_0^2}{\pi B(\theta) R_d^2} \frac{dN_{Ga}}{dt}$$

We obtain:

$$\frac{dR_d}{dt} = -\frac{2\Omega_{Ga} k_r C_0^2}{B(\theta) R_d^2} \int_0^{R_\infty} r C_{Ga} C_{As} dr$$

Using the finite difference values:

$$R_d^{i+1} = R_d^i - \frac{2\Omega_{Ga} k_r C_0^2 \Delta t \Delta r^2}{B(\theta) R_d^2} \sum_{j=1}^{N_r} j C_{Ga}^{i,j} C_{As}^{i,j}$$

Using an already introduced parameter (C_{Ga}, C_{As} are dimensionless, divided by C_0):

$$R_d^{i+1} = R_d^i - \frac{2\Omega_{Ga} \omega C_0 \Delta r^2}{B(\theta) R_d^2} \sum_{j=1}^{N_r} j C_{Ga}^{i,j} C_{As}^{i,j} \tag{31}$$

To get a crude estimate of Ga source at the droplet boundary, instead of using an additional flux, we can keep C_{Ga} near $r = R_d$ equal to C_0 at all time steps.

For example, introducing w again, we use the following scheme (without any Ga flux):

$$C_{Ga}^{i+1,j} = \begin{cases} C_{Ga}^{i,0} + 4\alpha \left[C_{Ga}^{i,1} - C_{Ga}^{i,0}\right] - \omega C_{Ga}^{i,0} C_{As}^{i,0}, & j = 0 \\ C_{Ga}^{i+1,j} = C_{Ga}^{i,j} + \alpha \left[\left(1 + \frac{1}{2j}\right) C_{Ga}^{i,j+1} - 2C_{Ga}^{i,j} + \left(1 - \frac{1}{2j}\right) C_{Ga}^{i,j-1}\right] - \omega C_{Ga}^{i,j} C_{As}^{i,j}, & 0 < j < j_a \text{ or } j_b < j \leq N_r \\ 1, & j_a \leq j \leq j_b \\ 0, & j = N_r + 1 \end{cases} \tag{32}$$

Where:

$$j_a = \left\lfloor \frac{R_d - w}{\Delta r} \right\rfloor, \qquad j_b = \left\lfloor \frac{R_d + w}{\Delta r} \right\rfloor \tag{33}$$

Remember that we measure the concentrations in units of C_0.

We end the calculation when $R_d \leq w - \Delta r$.

References

[1] Z. Y. Zhou, C. X. Zheng, W. X. Tang. Origin of Quantum Ring Formation During Droplet Epitaxy. PRL 111, 036102 (2013)