Quantum ring growth simulation algorithm

March 31, 2025

1 Main equations

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

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

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

Parameters in detail

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

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

Arsenic desorption time:

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

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

1.1 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.

To do that, we consider a more simple model, where the calculation takes place for $r \in [R_d, R_\infty]$ and only Ga is present. This leads to the following equation:

$$\frac{\partial C_{Ga}}{\partial t} = D_{Ga}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C_{Ga}}{\partial r}\right) \tag{9}$$

$$C_{Ga}\left(R_d, t\right) = C_0$$

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

We can solve it through separation of variables, or simply consider a stationary state:

$$\frac{\partial C_{Ga}}{\partial t} = 0$$

Then we need to solve:

$$\frac{\partial}{\partial r}\left(r\frac{\partial C_{Ga}}{\partial r}\right) = 0 \tag{10}$$

We obtain the general solution:

$$C_{Ga} = A\ln\frac{r}{R_0} \tag{11}$$

Where A, R_0 are unknown constants. Substitution of the boundary conditions leads to:

$$A\ln\frac{R_d}{R_0} = C_0$$

$$A\ln\frac{R_\infty}{R_0} = 0$$

Thus we obtain:

$$R_0 = R_\infty$$

$$A = \frac{C_0}{\ln\frac{R_d}{R_\infty}} \tag{12}$$

Thus we obtain:

$$C_{Ga} = \frac{C_0}{\ln\frac{R_d}{R_\infty}}\ln\frac{r}{R_\infty} \tag{13}$$

Now we need to find the flux from the droplet boundary:

$$J_{Ga} = -D_{Ga}\frac{\partial C_{Ga}}{\partial r}\,(r = R_d) \tag{14}$$

$$\frac{\partial C_{Ga}}{\partial r}(r = R_d) = \frac{C_0}{R_d \ln \frac{R_d}{R_\infty}} \tag{15}$$

$$J_{Ga} = -\frac{D_{Ga} C_0}{R_d \ln \frac{R_d}{R_\infty}}$$

The flux is positive (because it should move from the droplet in the direction $r \to R_\infty$), so we rewrite it as:

$$J_{Ga} = \frac{D_{Ga} C_0}{R_d \ln \frac{R_\infty}{R_d}} > 0 \tag{16}$$

Change of Ga atom number then will be:

$$\frac{dN_{Ga}}{dt} = -2\pi R_d J_{Ga} = -2\pi \frac{D_{Ga} C_0}{\ln \frac{R_\infty}{R_d}} \tag{17}$$

On the other hand, in our original problem we need to have:

$$\frac{dN_{Ga}}{dt} = -2\pi \int_0^{R_\infty} F_{Ga} r dr \tag{18}$$

$$-\frac{dN_{Ga}}{dt} = 2\pi \frac{C_0}{\tau_{Ga}} \int_0^{R_\infty} \exp\left(-\frac{(r - R_d)^2}{w^2}\right) r dr =$$

$$u = r - R_d, \qquad r = u + R_d$$

$$= 2\pi \frac{C_0}{\tau_{Ga}} \int_{-R_d}^{R_\infty - R_d} \exp\left(-\frac{u^2}{w^2}\right) u du + 2\pi \frac{C_0 R_d}{\tau_{Ga}} \int_{-R_d}^{R_\infty - R_d} \exp\left(-\frac{u^2}{w^2}\right) du$$

Due to symmetry we have:

$$\int_{-R_d}^{R_d} \exp\left(-\frac{u^2}{w^2}\right) u du = 0$$

$$\int_{-R_d}^{R_\infty - R_d} \exp\left(-\frac{u^2}{w^2}\right) u du = \int_{R_d}^{R_\infty - R_d} \exp\left(-\frac{u^2}{w^2}\right) u du$$

$$-\frac{dN_{Ga}}{dt} = \pi \frac{C_0 w^2}{\tau_{Ga}} \int_{\frac{R_d^2}{w^2}}^{\frac{(R_\infty - R_d)^2}{w^2}} \exp(-v)\, dv + 2\pi \frac{C_0 w R_d}{\tau_{Ga}} \int_{-\frac{R_d}{w}}^{\frac{R_\infty - R_d}{w}} \exp\left(-v^2\right) dv$$

$$-\frac{dN_{Ga}}{dt} = \pi \frac{C_0 w^2}{\tau_{Ga}} \left[\exp\left(-\frac{R_d^2}{w^2}\right) - \exp\left(-\frac{(R_\infty - R_d)^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{\text{erf}\left(\frac{R_\infty - R_d}{w}\right) + \text{erf}\left(\frac{R_d}{w}\right)\right\}\right] \tag{19}$$

Comparing the two fluxes gives us the following equation:

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

$$\tau_{Ga} = \frac{w^2}{2D_{Ga}} \left[\exp\left(-\frac{R_d^2}{w^2}\right) - \exp\left(-\frac{(R_\infty - R_d)^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{\text{erf}\left(\frac{R_\infty - R_d}{w}\right) + \text{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln \frac{R_\infty}{R_d} \tag{20}$$

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

$$\tau_{Ga} = \frac{w^2}{2D_{Ga}} \left[\exp\left(-\frac{R_d^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{1 + \text{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln \frac{R_\infty}{R_d} \tag{21}$$

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

$$F_{Ga}(r,t) = \frac{2D_{Ga}C_0}{w^2} \frac{\exp\left(-\frac{(r - R_d(t))^2}{w^2}\right)}{\left[\exp\left(-\frac{R_d^2}{w^2}\right) - \exp\left(-\frac{(R_\infty - R_d)^2}{w^2}\right) + \sqrt{\pi}\frac{R_d}{w}\left\{\text{erf}\left(\frac{R_\infty - R_d}{w}\right) + \text{erf}\left(\frac{R_d}{w}\right)\right\}\right] \ln \frac{R_\infty}{R_d}} \tag{22}$$

Note that if we define the variables:

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

Then the function:

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

Can be approximated by a straight line:

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

Where:

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

$$a\left(p\right)\approx 3.545$$

So we obtain:

$$F_{Ga}\left(r,t\right)=\frac{2D_{Ga}C_{0}}{w^{2}}\frac{\exp\left(-\frac{\left(r-R_{d}\left(t\right)\right)^{2}}{w^{2}}\right)}{\left[3.545\frac{R_{d}}{w}+\frac{0.187w}{R_{\infty}-3.156w}\right]\ln\frac{R_{\infty}}{R_{d}}} \tag{23}$$

3 Droplet geometry

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

For Ga depletion rate we pick the easiest expression 17:

$$\frac{dN_{Ga}}{dt}=-2\pi\frac{D_{Ga}C_{0}}{\ln\frac{R_{\infty}}{R_{d}}}$$

Form-factor:

$$B\left(\theta\right)=\left(\frac{1}{\sin\theta}-\cot\theta\right)^{2}\left(\frac{2}{\sin\theta}+\cot\theta\right)=\frac{\left(1-\cos\theta\right)^{2}\left(2+\cos\theta\right)}{\sin^{3}\theta}=\frac{\left(1-2\cos\theta+\cos^{2}\theta\right)\left(2+\cos\theta\right)}{\sin^{3}\theta}=$$

$$=\frac{2-4\cos\theta+2\cos^{2}\theta+\cos\theta-2\cos^{2}\theta+\cos^{3}\theta}{\sin^{3}\theta}=\frac{2-3\cos\theta+\cos^{3}\theta}{\sin^{3}\theta}=\frac{2-3\cos\theta+\frac{3}{4}\cos\theta+\frac{1}{4}\cos 3\theta}{\frac{3}{4}\sin\theta-\frac{1}{4}\sin 3\theta}$$

$$B(\theta) = \frac{8 - 9\cos\theta + \cos 3\theta}{3\sin\theta - \sin 3\theta} \tag{24}$$

$$\frac{dR_d}{dt} = \frac{\Omega_{Ga}}{\pi B(\theta) R_d^2} \frac{dN_{Ga}}{dt} \tag{25}$$

$$\frac{d}{dt}\frac{R_d}{R_\infty} = -\frac{2\Omega_{Ga} D_{Ga} C_0}{B(\theta) R_\infty^3} \frac{R_\infty^2}{R_d^2 \ln \frac{R_\infty}{R_d}} \tag{26}$$

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

$$\frac{2\Omega_{Ga} D_{Ga} C_0}{B(\theta) R_\infty^3} = \sigma \tag{27}$$

$$\frac{d\rho}{dt} = \frac{\sigma}{\rho^2 \ln \rho} \tag{28}$$

But this ODE can be easily solved:

$$\rho^2 \ln \rho d\rho = \sigma dt \tag{29}$$

$$\frac{\rho^3}{9}\left(\ln \rho^3 - 1\right) = \sigma t + C \tag{30}$$

The initial state is a known value:

$$\rho(0) = \rho_0 < 1 \tag{31}$$

$$C = \frac{\rho_0^3}{9}\left(\ln \rho_0^3 - 1\right) \tag{32}$$

We obtain the following equation for droplet radius:

$$\rho^3\left(\ln \rho^3 - 1\right) = \rho_0^3\left(\ln \rho_0^3 - 1\right) + 9\sigma t \tag{33}$$

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

$$p\left(\rho\right)=\rho^{3}\left(3\ln\rho-1\right)-\rho_{0}^{3}\left(3\ln\rho_{0}-1\right)-9\sigma t \tag{34}$$

$$p'\left(\rho\right)=3\rho^{2}\left(\ln\rho^{3}-1\right)-3\rho^{2}=3\rho^{2}\left(3\ln\rho-2\right) \tag{35}$$

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:

$$\rho_{i+1}^{(k+1)}=\rho_{i+1}^{(k)}-\frac{\left(\rho_{i+1}^{(k)}\right)^{3}\left(3\ln\rho_{i+1}^{(k)}-1\right)-\rho_{0}^{3}\left(3\ln\rho_{0}-1\right)-9\sigma t_{i}}{3\left(\rho_{i+1}^{(k)}\right)^{2}\left(3\ln\rho_{i+1}^{(k)}-2\right)} \tag{36}$$

Note that we should always have:

$$0<\rho<1 \tag{37}$$

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 $R_{d}\left(T\right)\leq a$.

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 original equations:

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

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

And the boundary conditions:

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

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

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:

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

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

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

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

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

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

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

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

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

The equations become:

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

$$C_{Ga}^{i+1,0} = C_{Ga}^{i,0} + 4\alpha\left[C_{Ga}^{i,1} - C_{Ga}^{i,0}\right] + 2\alpha\eta f_{i,j} - \omega C_{Ga}^{i,0} C_{As}^{i,0}$$

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

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

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

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

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

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

5 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:

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

$$D_{Ga} = 1$$

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

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

6 Ring growth speed

Let's derive the expression for the QR height $h\left(r,t\right)$, 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:

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

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

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

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

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

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

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

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

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

Finally, we obtain:

$$h_{i+1,j} = h_{i,j} + \omega h_0 C_{Ga}^{i,j} C_{As}^{i,j} \tag{38}$$

The initial condition is clearly:

$$h_{0,j} = 0 \tag{39}$$

References

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