Predictive modeling of self-catalyzed GaAs nanowire growth

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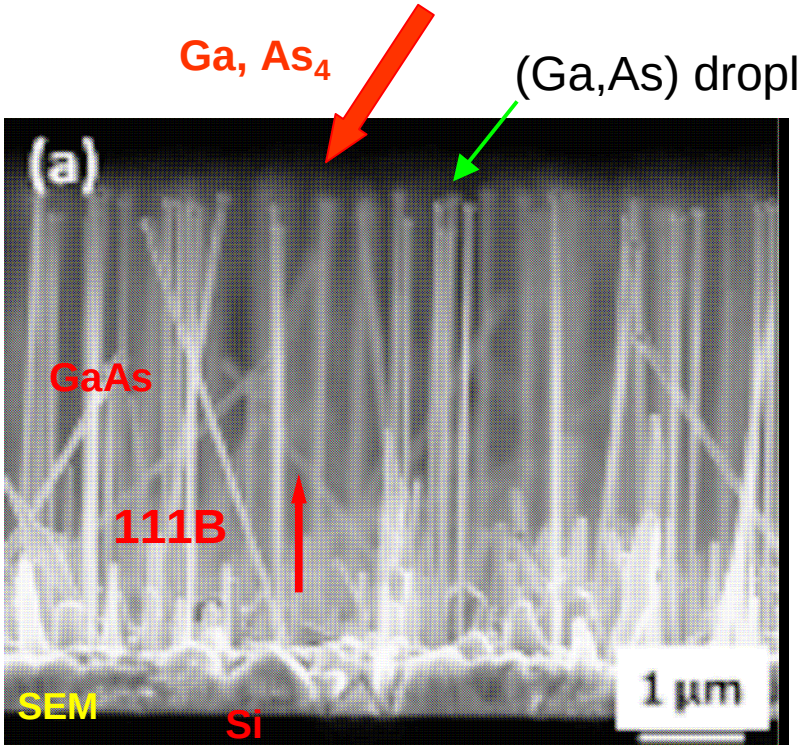
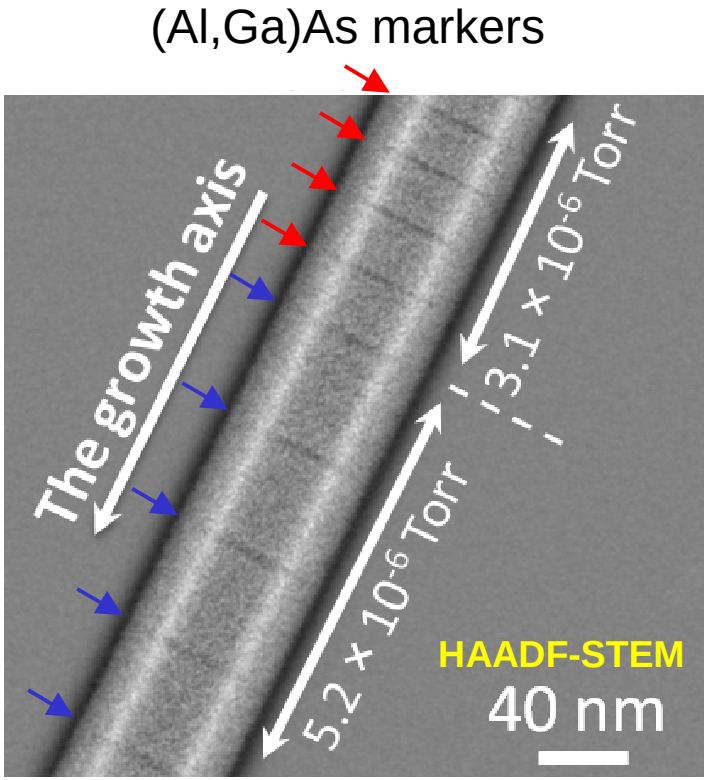
Chronology of self-catalyzed GaAs growth

GaAs NWs by VLS MBE





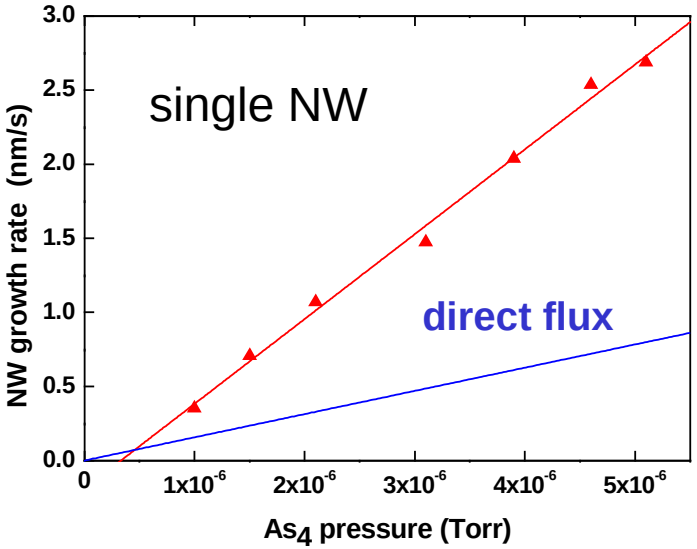
- Cubic ZB structure
- No radial growth

Ramdani, Harmand, Glas, Patriarche, Travers, NWG 2012, St Petersburg
Ramdani, Harmand, Glas, Patriarche, Travers, Cryst. Growth Des. 13, 91 (2013)

The mystery of the missing arsenic

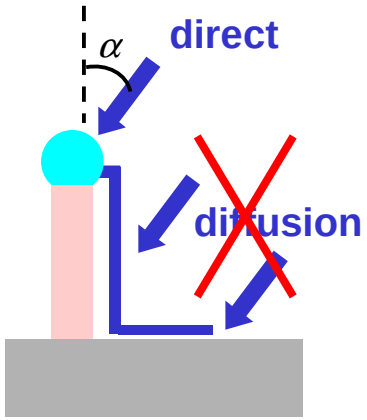
- **NW growth rate**
  - **is independent of Ga flux**
  - **varies linearly with As flux**

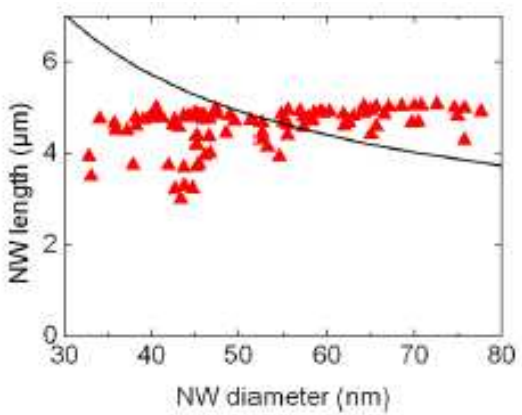
Colombo et al., Phys. Rev. B 77, 155326 (2008)



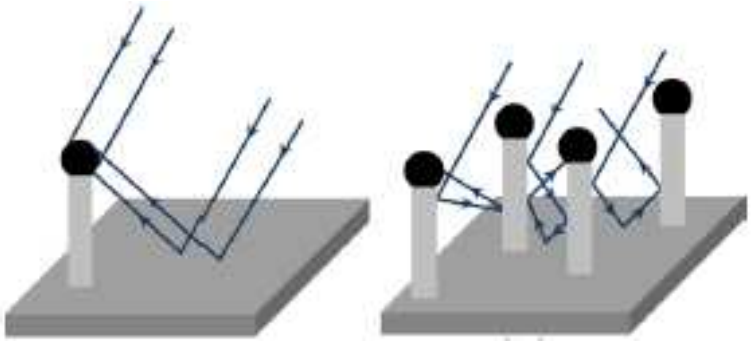
Ramdani et al., Cryst. Growth Des. 13, 91 (2013)

- **As diffusion is negligible**





- **Missing As: re-emission?**



- **Direct As flux is insufficient**

$$\phi_d = \eta \, p_{As_4}$$

As-only model

- Growth is governed by As
- Surface diffusion of As species can be neglected
- Droplet = (As,Ga) liquid
  Thermodynamics entirely determined by c_{As} $(c_{Ga} = 1 - c_{As})$

Find As concentration c_{As} in droplet

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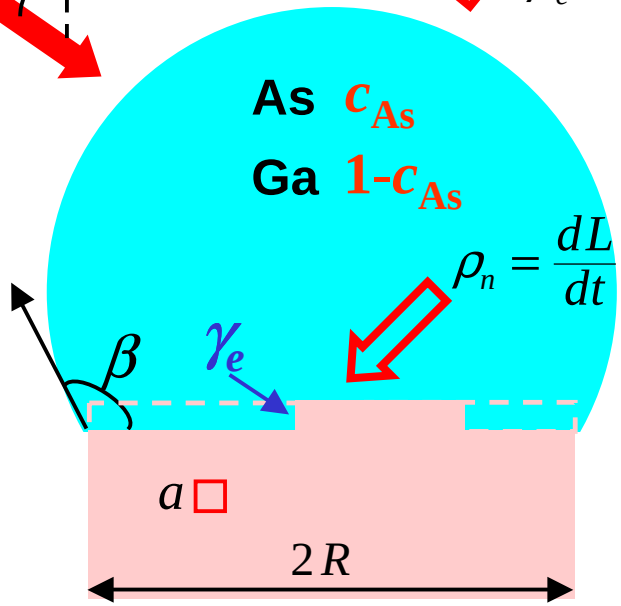
Model and experiments

$$\rho_d = \frac{a^3}{4\sin^2\beta}\,\eta\, p_{As_4}$$

$$\rho_e = k_e \frac{1-\cos\beta}{\sin^2\beta} \frac{a^3}{\sqrt{2\pi M k_B T}}\, p^{eq}_{As_2}(c_{As}, T)$$

$$\rho_r = \epsilon\, \rho_d$$

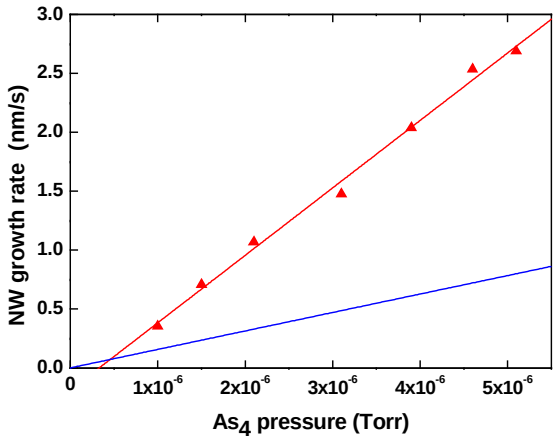


2D nucleation

$$\frac{dL}{dt} = \pi R^2 \frac{a}{\sqrt{3}} A(T)\, c_{As} \sqrt{\frac{\Delta\mu}{k_B T}} \exp\left[-\frac{\Delta G_c}{k_B T}\right]$$

$$\Delta G_c = \chi \frac{a^4}{4\sqrt{3}} \frac{\gamma_e^2}{\Delta\mu\,(c_{As}, T)}$$

2 model parameters

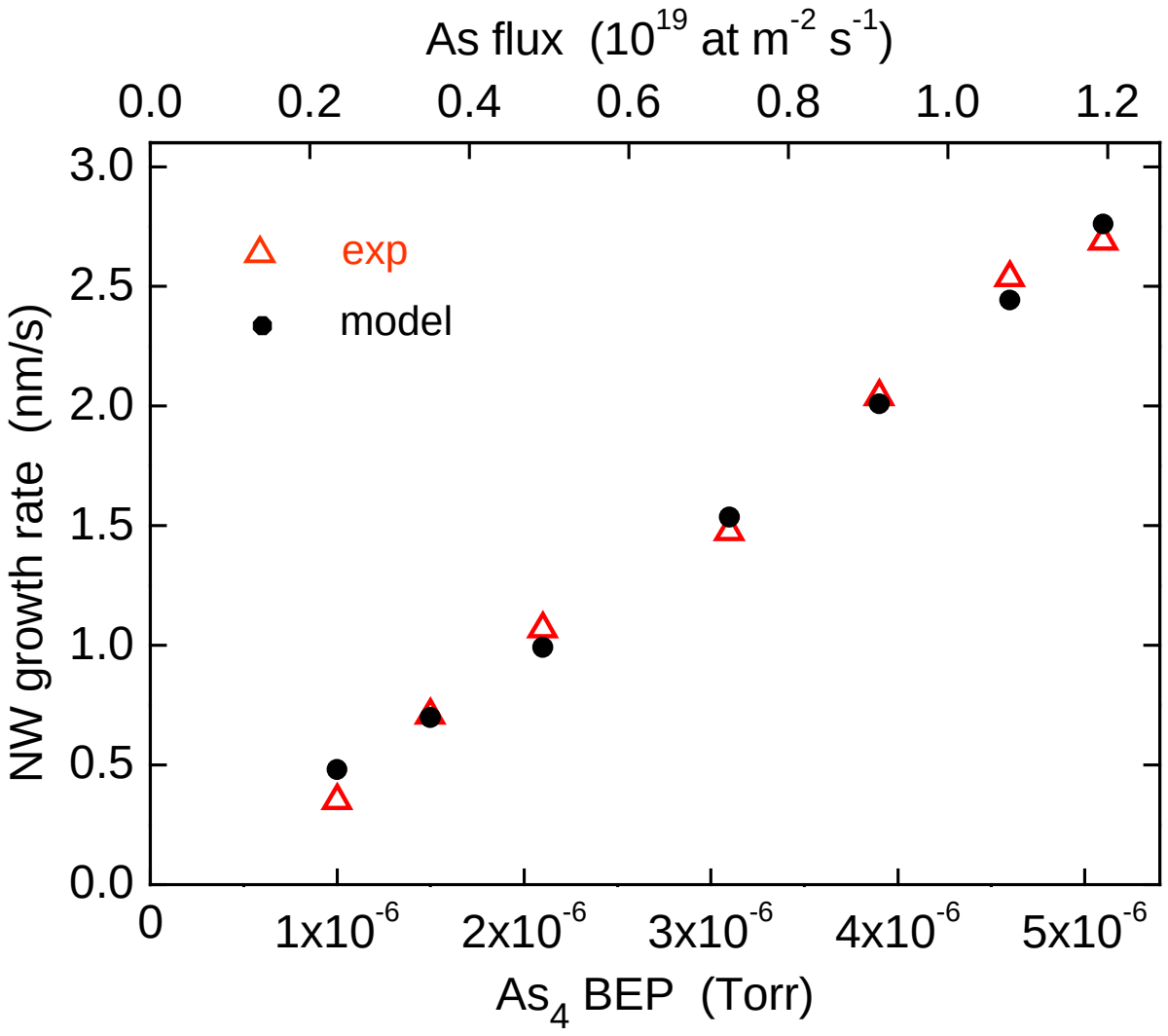


As material balance (drop) $\quad (1+\epsilon)\,\rho_d = \rho_e(c_{As}, T) + \rho_{exp}$

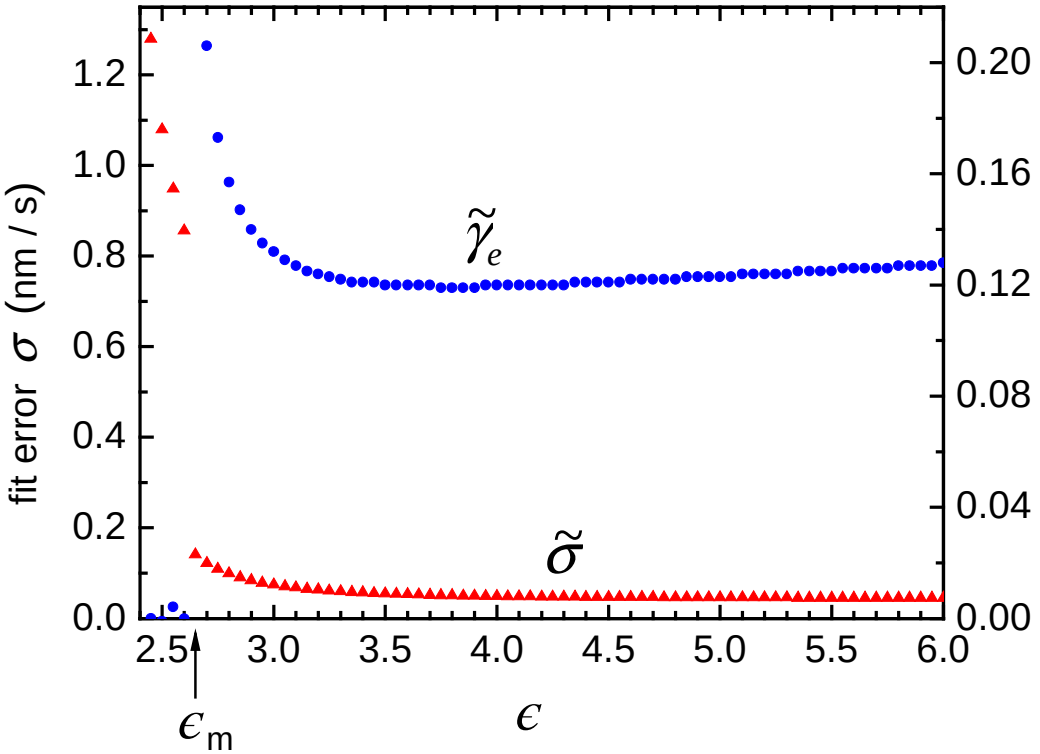
arbitrary ϵ → $\{c_{As}\}$ → $\{\rho_n\}$ → $\{\rho_n = \rho_{exp}\}$ → best model parameters **for this ϵ** $\tilde{A}$, $\tilde{\gamma}_e$

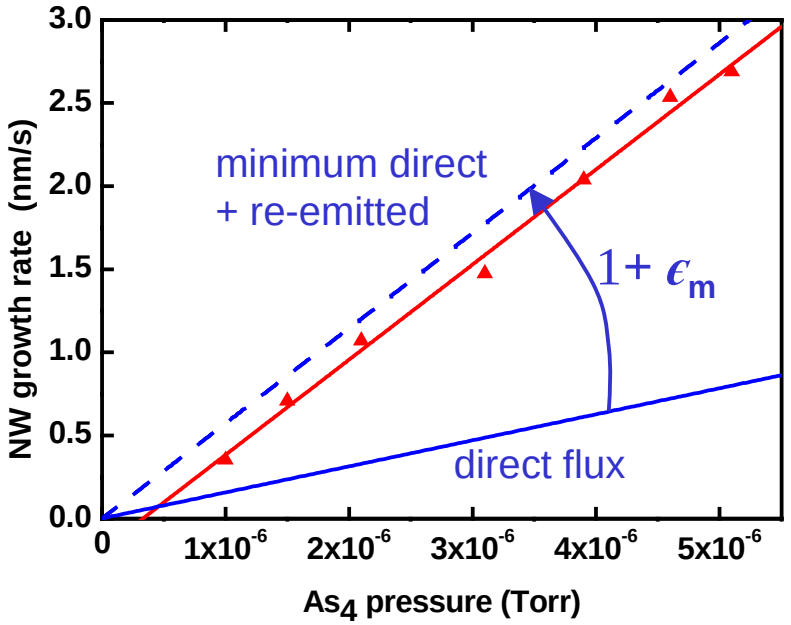
model parameters

Fitting pressure series

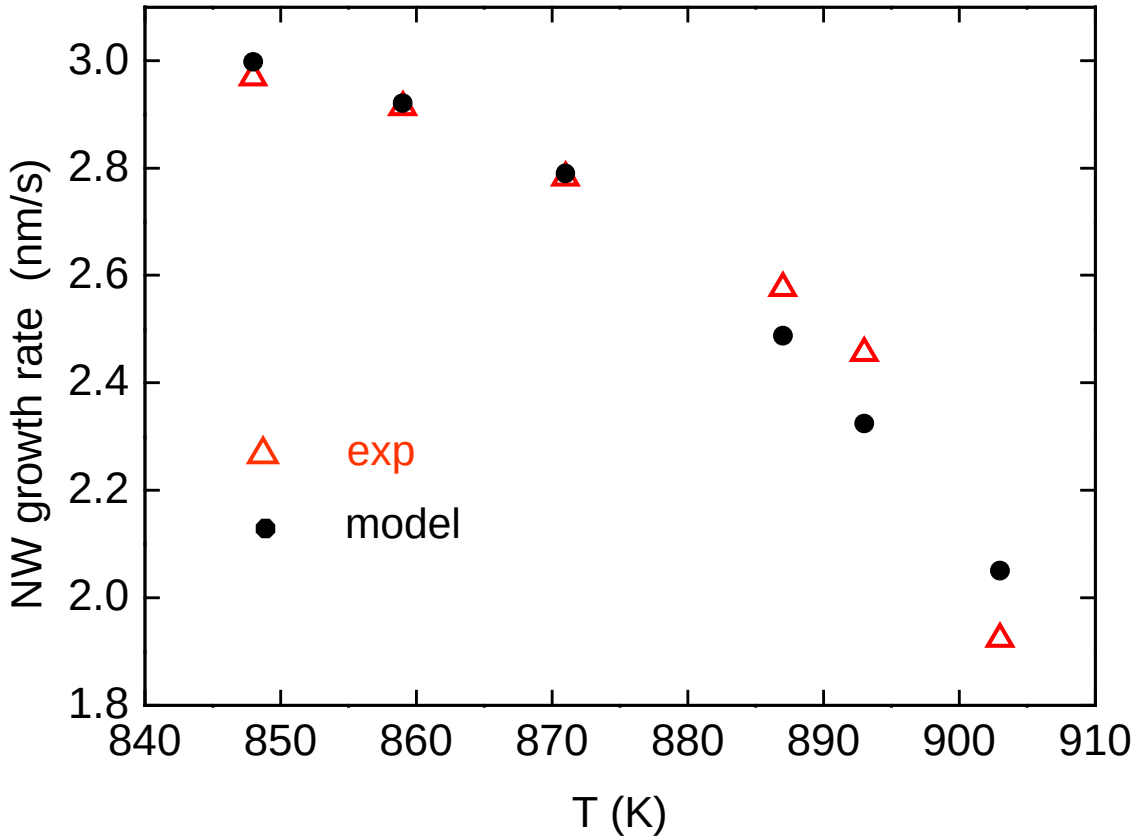


Fitting pressure series - Minimum ϵ



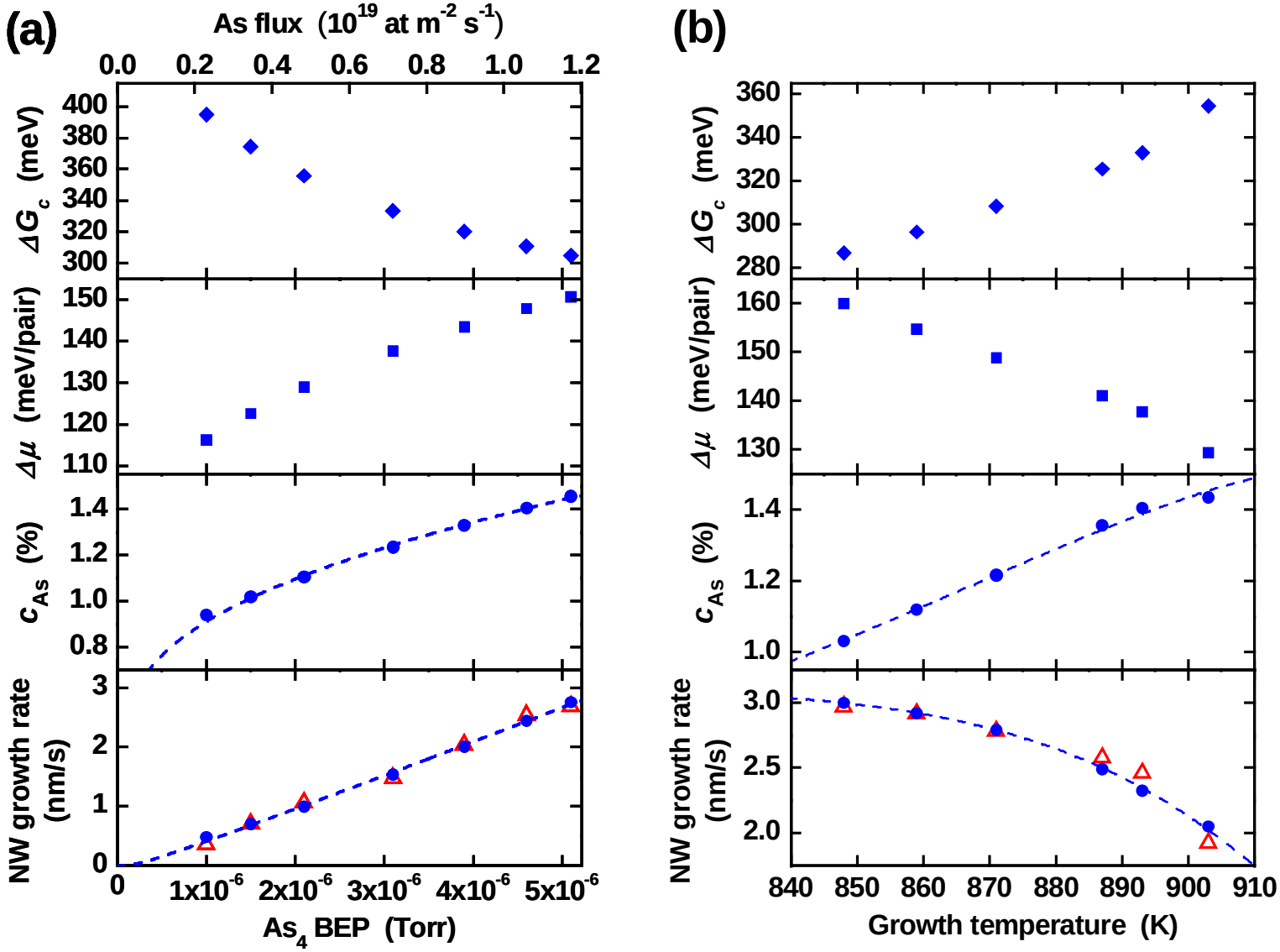


Fitting temperature series

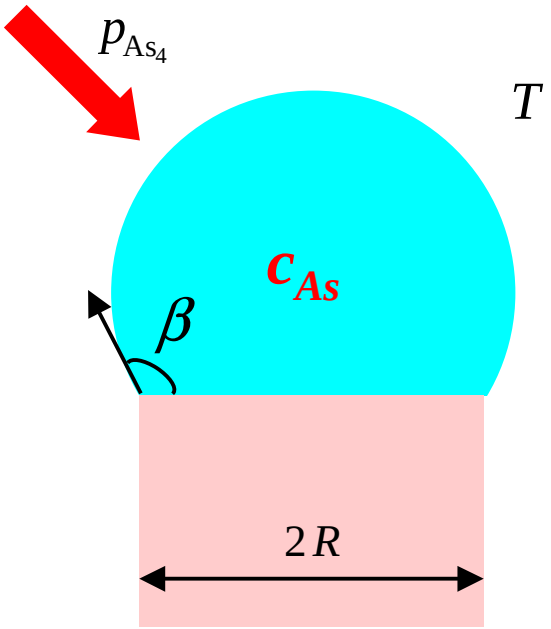


- Best values of model parameters $\gamma_e \approx 0.123\ \mathrm{J/m^2}$
- Re-emission coefficient ϵ for individual NWs

Non-measurable quantities



Predictive modeling



known

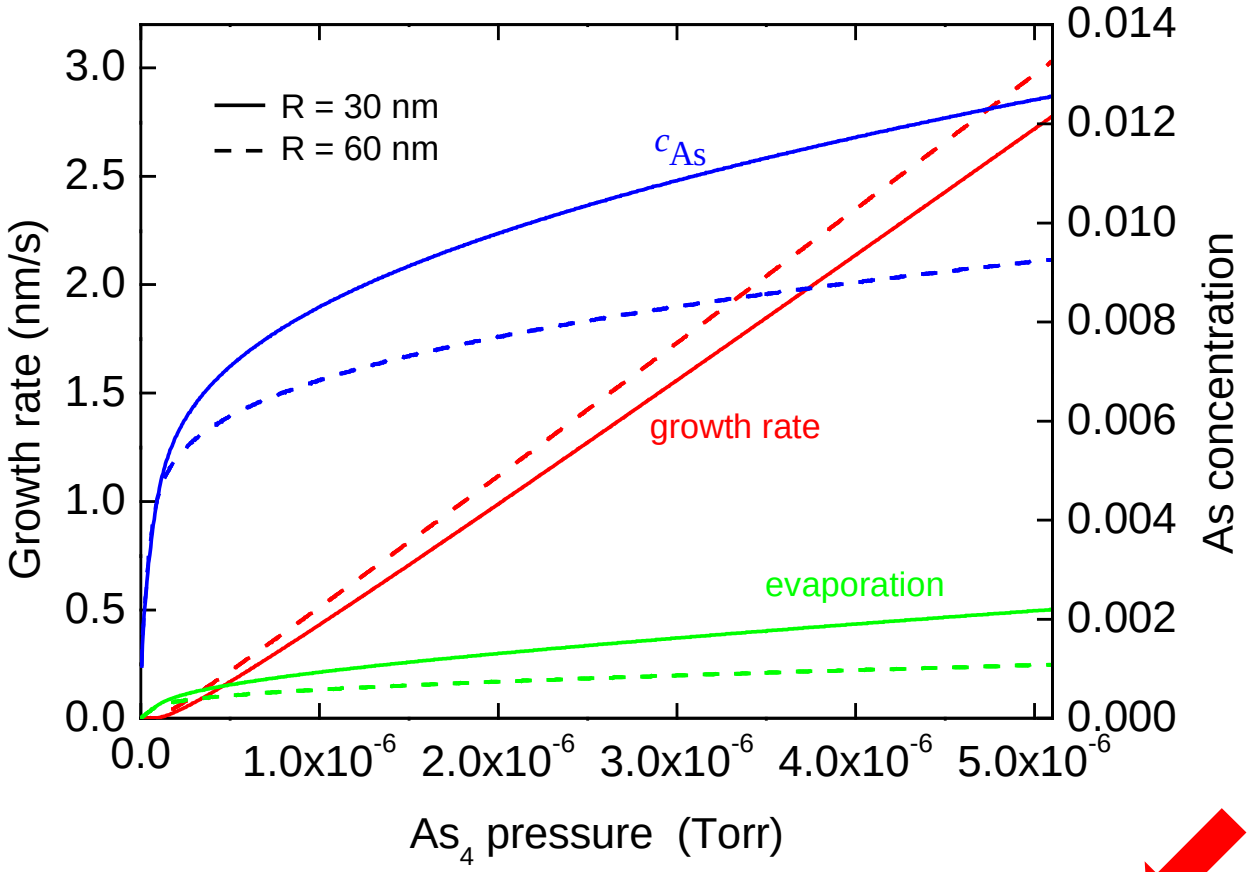
$$\rho_n = \pi R^2 \frac{a}{\sqrt{3}} A(T) c_{As} \sqrt{\frac{\Delta\mu}{k_B T}} \exp\left[-\chi \frac{a^3}{4\sqrt{3}} \frac{\gamma_e^2}{\Delta\mu k_B T} \right]$$

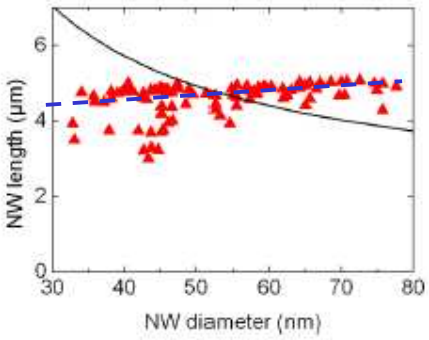
c_{As}?

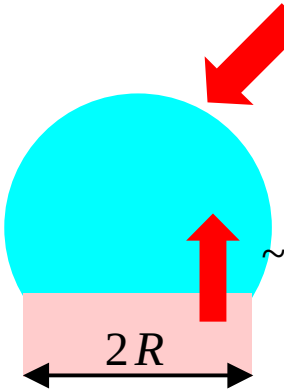
As material balance

$$(1+\epsilon)\,\rho_d = \rho_e(c_{As},T) + \rho_n(c_{As},T)$$

Simulated pressure series





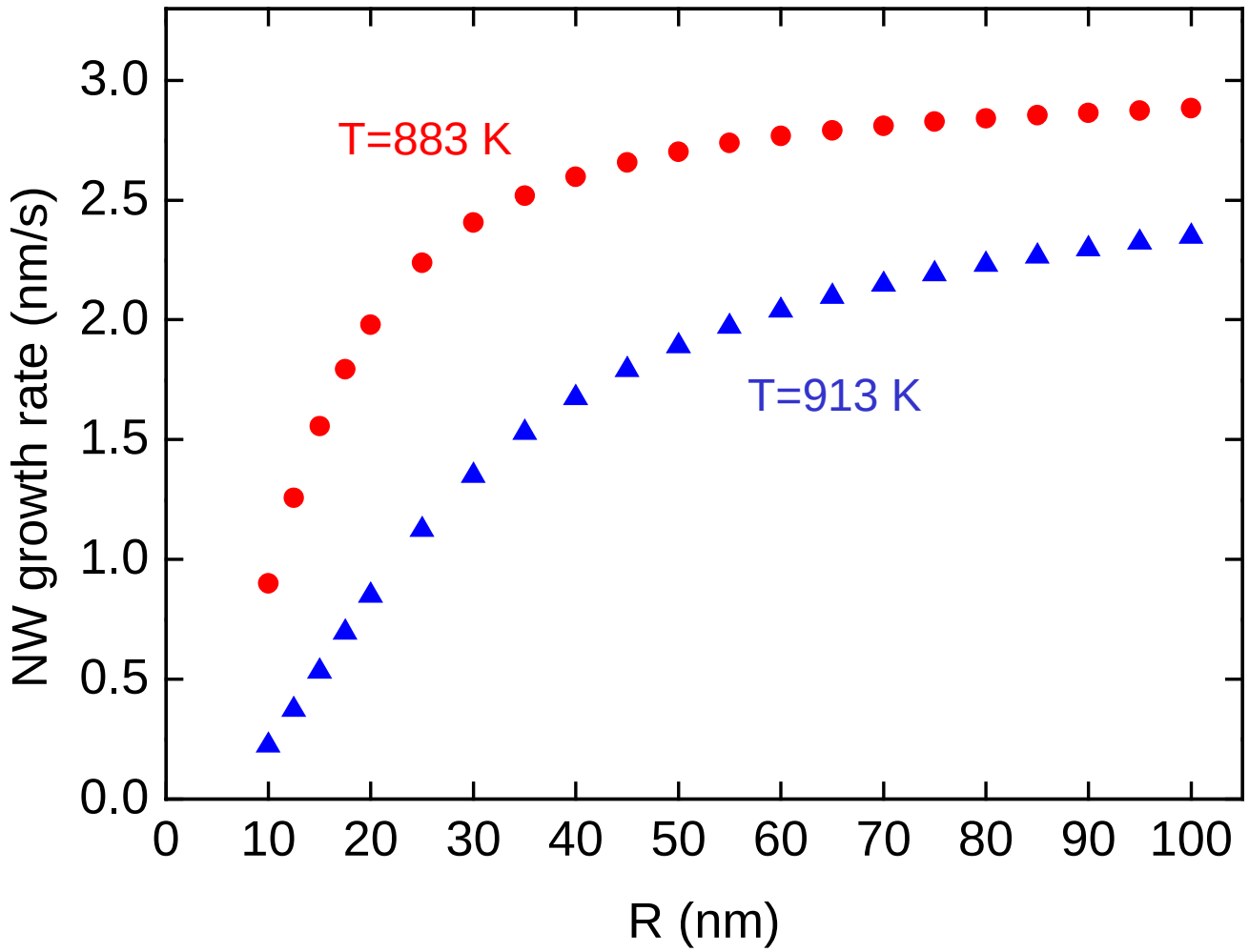


$\sim R^2$

$\sim R^2 \frac{dL}{dt} \sim R^2 \times R^2 \times \text{nucleation rate}$

$\rightarrow$ nucleation rate $\sim 1/R^2$

Effect of NW radius



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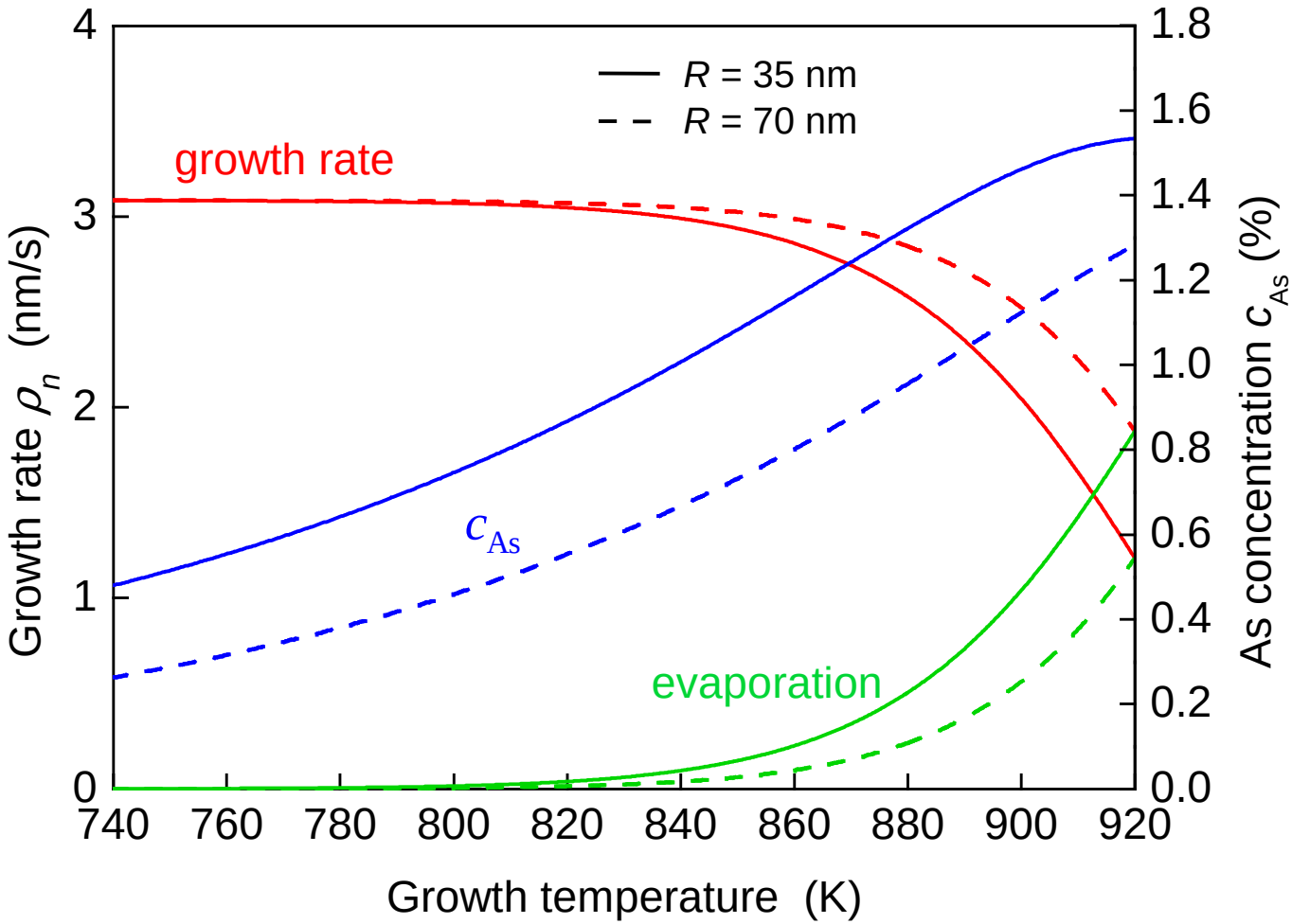
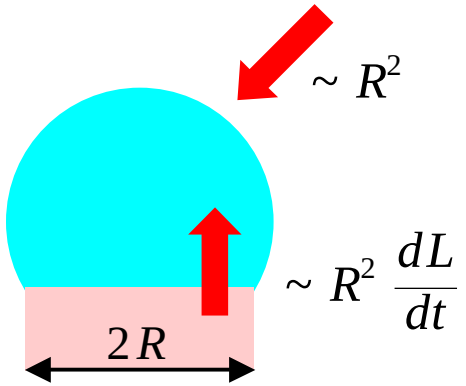
Simulated temperature series





Summary and conclusions

- As-only model for self-catalyzed GaAs NW growth
- Values of model parameters

$$\gamma_e \approx 0.123 \pm 0.02 \ \mathrm{J/m^2}$$

Hurle, J. Cryst. Growth 147, 239 (1995) $\gamma_e = 0.119 \ \mathrm{J/m^2}$ $\gamma_e = 0.064 \ \mathrm{J/m^2}$ (InP)

- Values of non-measurable quantities for single NWs

$$c_{\mathrm{As}} \sim 1\% \qquad \Delta\mu \qquad \Delta G_c \qquad \epsilon$$

- Model predicts NW growth rate and thermodynamic state of the droplet in given growth conditions (As flux, T, ϵ)

Glas, Ramdani, Patriarche, Harmand, Phys. Rev. B 88, 195304 (2013)