Modeling of Lateral Bulging of Diffusion Couples

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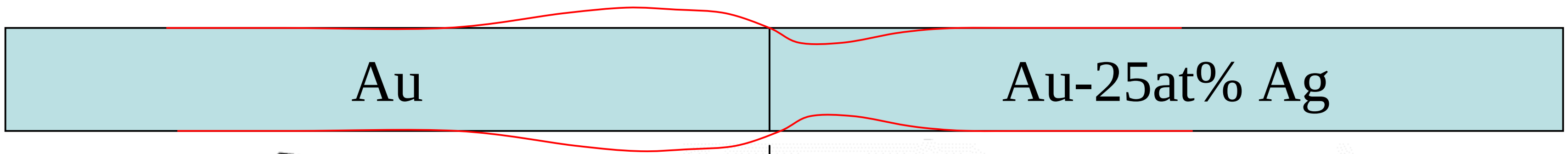
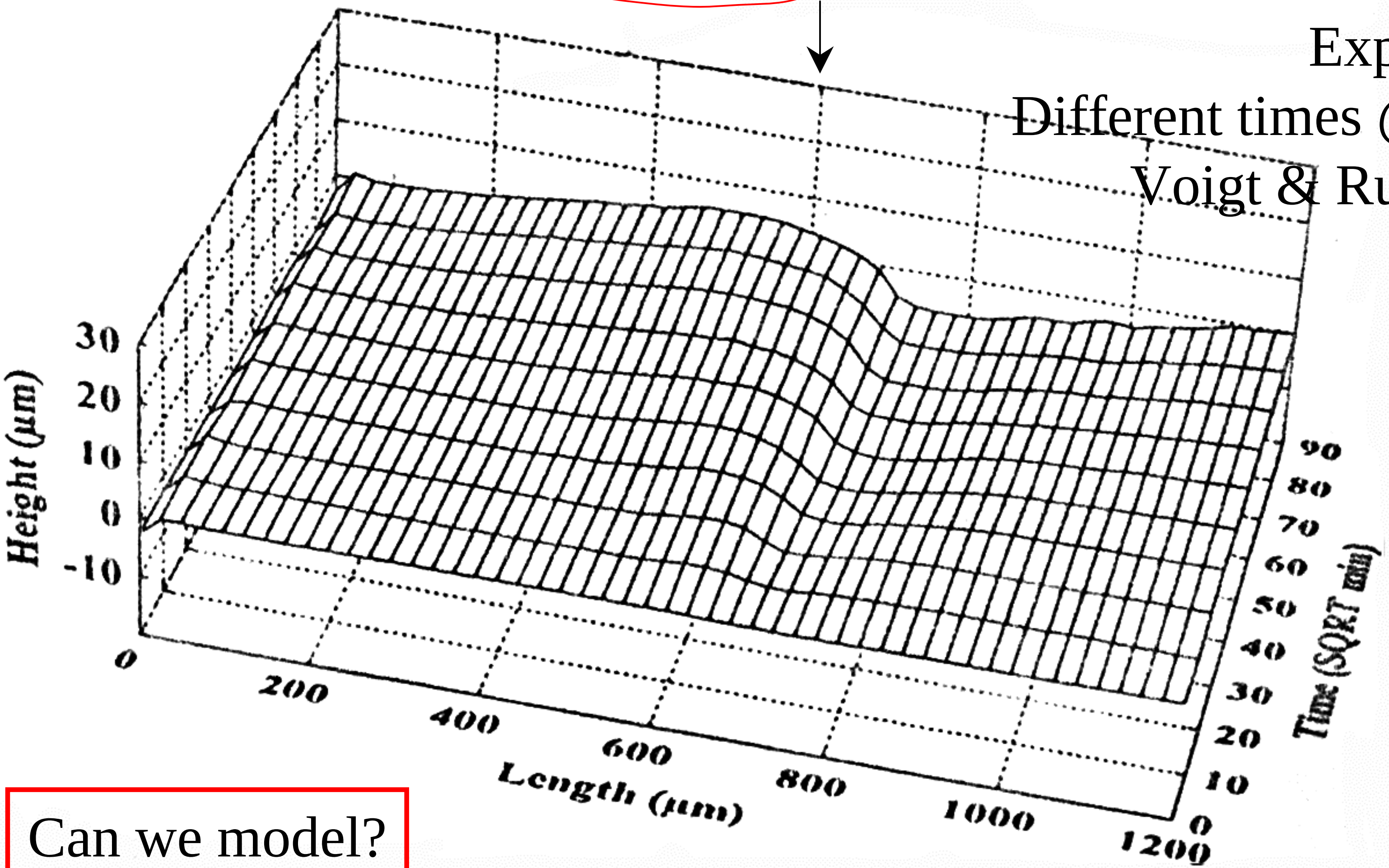




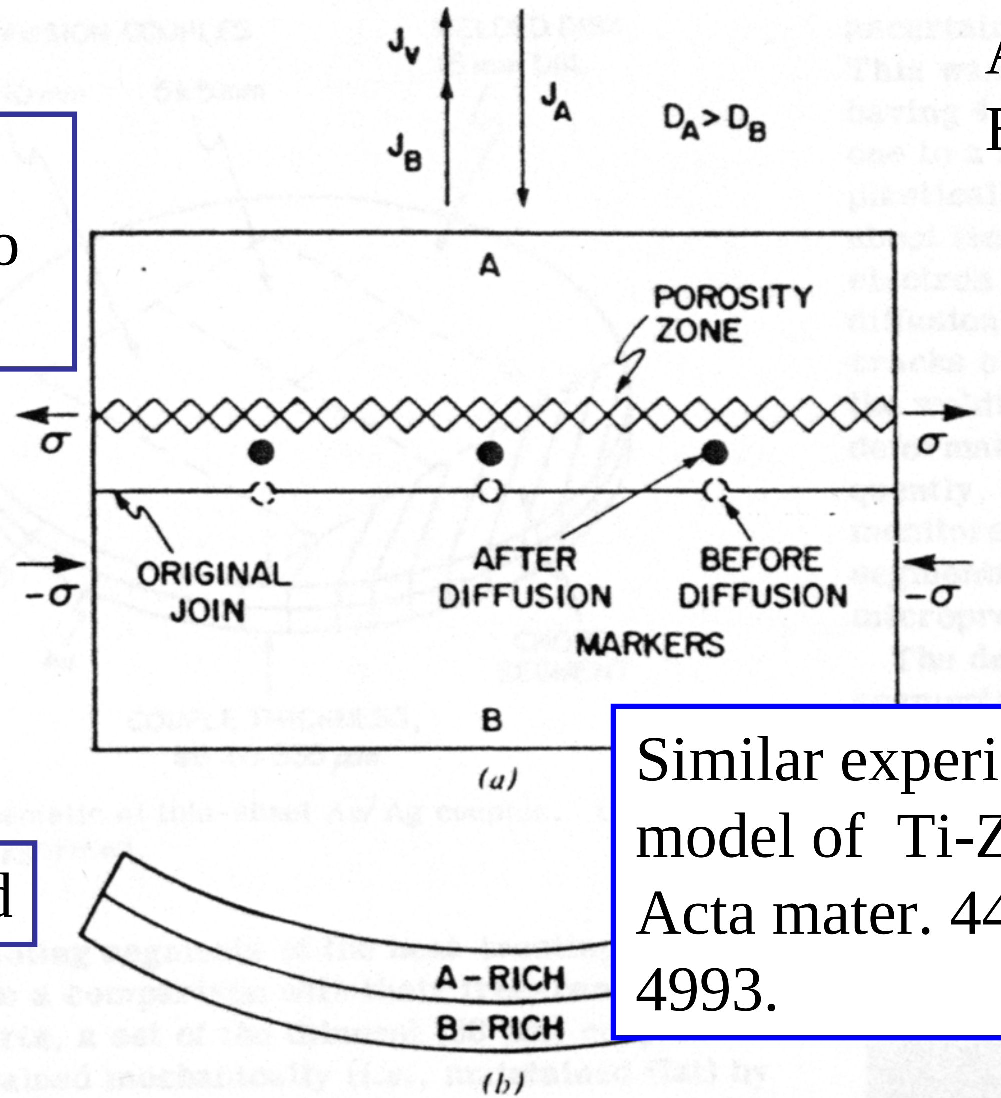
Experiments:
Different times @ 1081 K
Voigt & Ruth (1995)

Can we model?

Related phenomenon:
Bending of thin beams due to unequal diffusion rates

A=Ag
B=Au

Stress if constrained to remain flat



Ag-rich

Au-rich

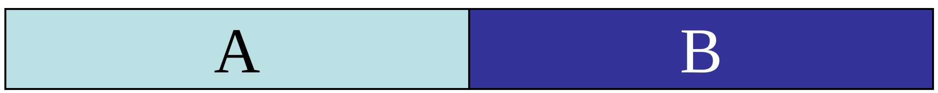
Unconstrained

Similar experiments and model of Ti-Zr, Daruka et al. Acta mater. 44(1996) 4981-4993.

D. W. Stevens & G. W. Powell, Met. Trans 8A(1977) 1531

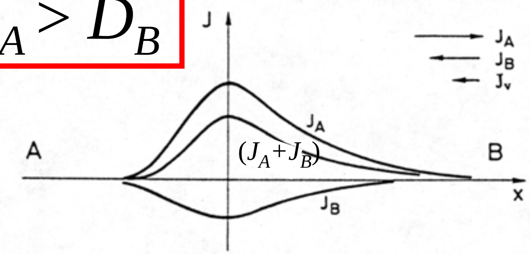
1-D Kirkendall Effect

Jean Philibert, *Atom Movements: Diffusion & Mass Transport in Solids*, 1991.



In 1-D, v is the velocity of the lattice planes relative to a lattice plane (observer) far from the diffusion zone

$D_A > D_B$



$$v = -\bar{V}\left(J_A + J_B\right)$$

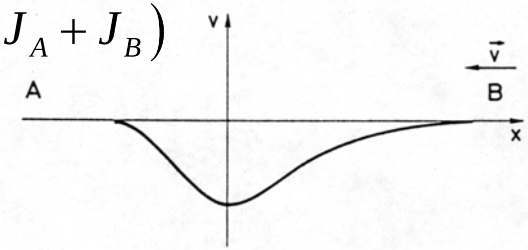


Fig. VI.1. — Interdiffusion of A and B, assuming $\tilde{D}_A > \tilde{D}_B$ everywhere.

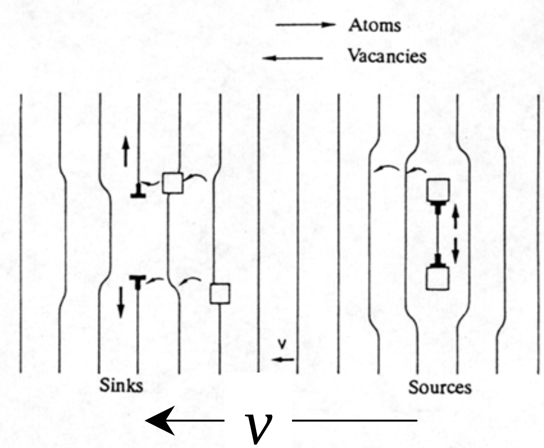


Fig VI.2 — Kirkendall effect: displacement of atomic planes resulting from a flux of vacancies. Vacancy sources are on the right, sinks on the left. The vertical arrows indicate dislocation climb.

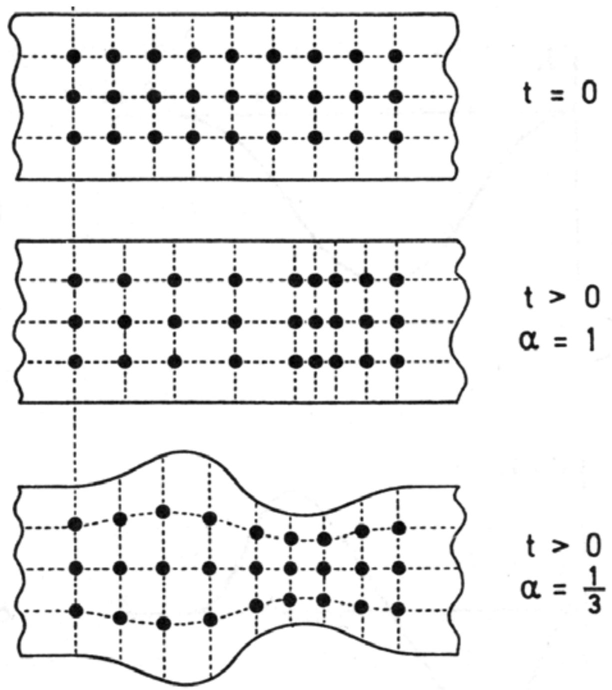


Fig. VIII.7. — Divergent flux of vacancies. Creation and annihilation of vacancies giving rise to purely axial deformation ($\alpha = 1$) or isotropic deformation ($\alpha = 1/3$). The points represent inert markers. After Monty (1972).

- α factor not necessary
- Stress boundary conditions determine amount of bulging

Stress-Free Strain Rate

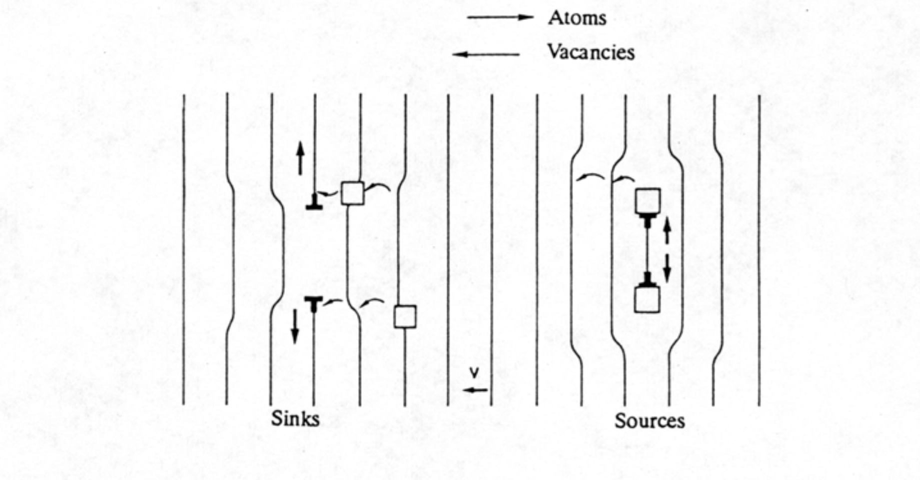


Fig VI.2 — Kirkendall effect: displacement of atomic planes resulting from a flux of vacancies. Vacancy sources are on the right, sinks on the left. The vertical arrows indicate dislocation climb.

But

- Climb of dislocations occurs ϒ to Burger's vector <u>b</u> & line <u>l</u>,
- Material may contain dislocations with a variety of <u>b</u>'s and <u>l</u>'s
- Material may also be polycrystalline
- If one averages over volumes containing many sources/sinks
- Can assume *stress-free* dilation rate due to vacancy creation/ annihilation is *isotropic*. G. B. Stephenson, Acta Met. 36 (1988) 2663.

$$\dot{\varepsilon}_{ij}^{0} = \frac{1}{3}\delta_{ij}\bar{V}\sigma_{v}$$

Analysis of Deformation; Small Strains

$$\dot{\varepsilon}_{ij}^{T} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}\right)$$ Definition

$$\dot{\varepsilon}_{ij}^{T} = \dot{\varepsilon}_{ij}^{o} + \dot{\varepsilon}_{ij}^{P}$$ Decomposition

$$\dot{\varepsilon}_{ij}^{0} = \frac{1}{3}\delta_{ij}\bar{V}\sigma_{v}$$ Stress-free strain rate

$$\dot{\varepsilon}_{ij}^{P} = \frac{1}{2\eta}\left[\sigma_{ij} - \frac{1}{3}\delta_{ij}\sigma_{kk}\right].$$ Plastic strain rate

$$\sigma_{ij,j} = 0$$ Stress equilibrium

Note: $\dot{\varepsilon}_{kk}^{T} = \nabla \cdot v = \dot{\varepsilon}_{kk}^{0} = \bar{V}\sigma_{v}$

Standard Analysis of Interdiffusion (for $\bar{V}_1 = \bar{V}_2 = \bar{V}_V = \bar{V}$)

- Decoupled from Stress
- Still treat as 1-D for small deformation
- Diffusion Equation

$$\frac{\partial X_2}{\partial t} = \nabla \cdot \left[\tilde{D} \nabla X_2 \right] \qquad \nabla \equiv \frac{\partial}{\partial z}$$

$$\tilde{D} = D_1 X_2 + D_2 (1 - X_2)$$

D_1 and D_2 are the intrinsic (lattice) diffusion coefficients

- Vacancy creation rate (required to maintain equilibrium vacancy content)

$$\bar{V} \sigma_v = \nabla \cdot J_v = -\nabla \cdot \left[J_1 + J_2 \right] = -\nabla \cdot \left[\left(D_1 - D_2 \right) \nabla X_2 \right]$$

$$\Delta D = D_1 - D_2$$

Simplification

If $\tilde{D}$ and ΔD can be treated as constants:

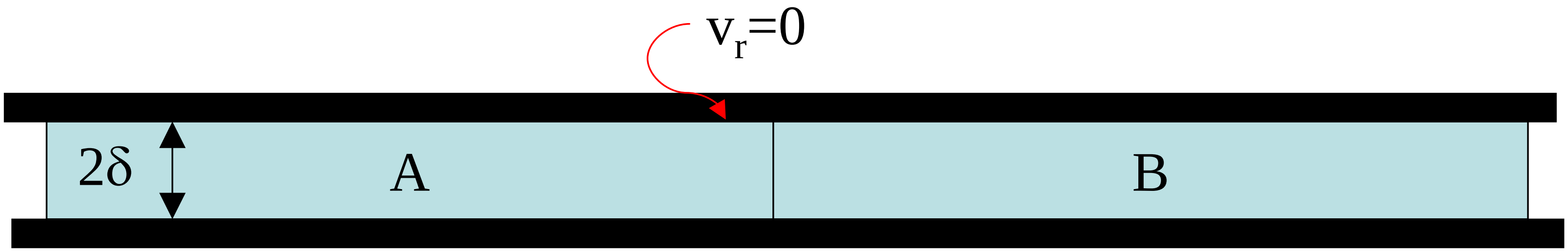
$$X_2(z,t) = \bar{X}_2 + \Delta X_2 \,\mathrm{erf}\left(\frac{z}{\sqrt{4\tilde{D}t}}\right)$$

$$\dot{\varepsilon}_{ij}^0 = -\frac{\Delta D}{3}\delta_{ij}\left(\frac{\partial^2 X_2}{\partial z^2}\right)$$

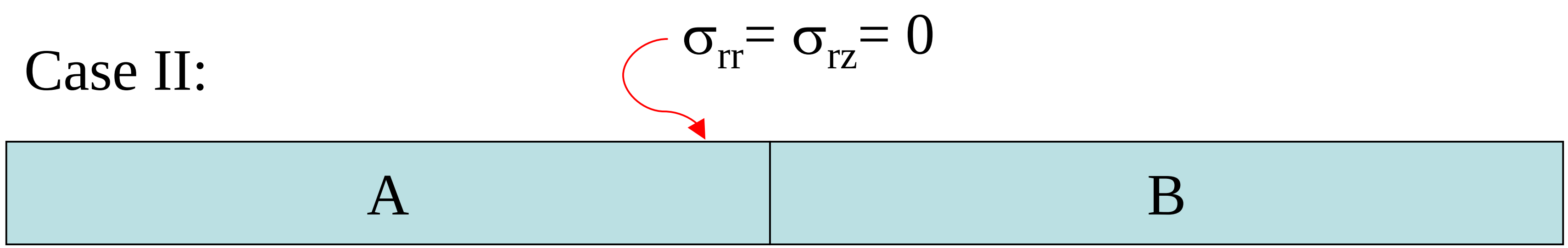
$$= \frac{2}{3}\delta_{ij}\left(\frac{\Delta D\,\Delta X_2}{\sqrt{\pi}\left(4\tilde{D}t\right)^{3/2}}\right) z \exp\left(-\frac{z^2}{4\tilde{D}t}\right)$$

Assume radial symmetry

Case I:



Case II:



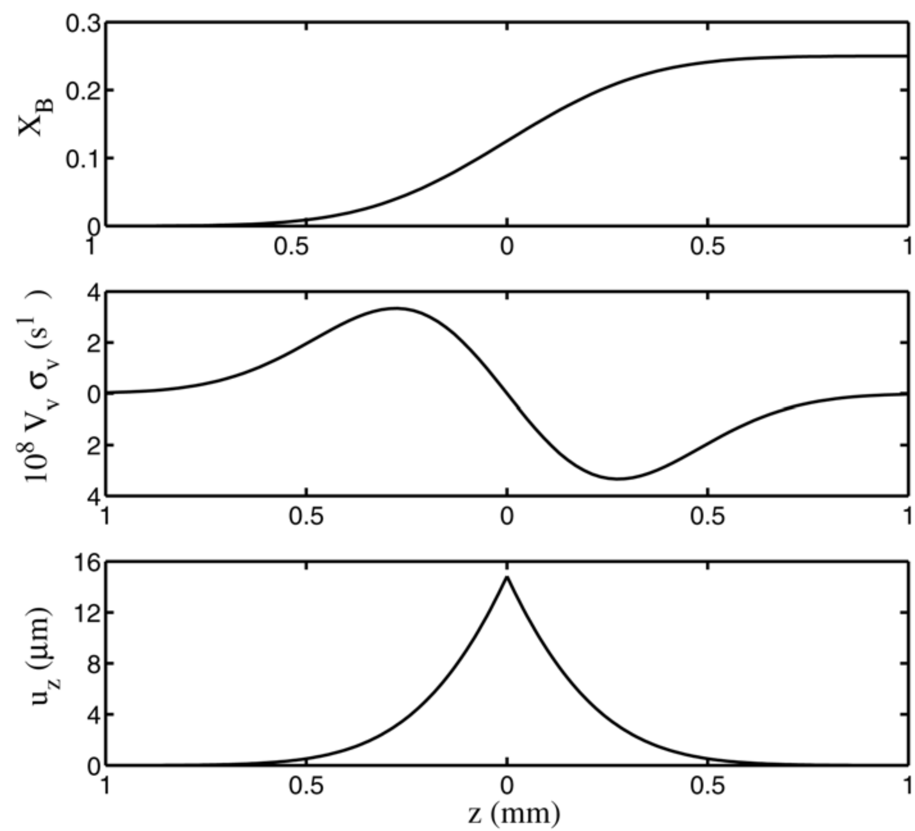
Case I – Displacement only in *axial* direction. Classical Kirkendall shift of markers on original (z=0) interface

$$\eta = \frac{z}{\sqrt{4\tilde{D}t}}$$

$$v_z(z,t) = -\Delta D \frac{\partial X_B}{\partial z} = \frac{\Delta D}{\sqrt{4\tilde{D}t}} \frac{\Delta X_B}{\sqrt{\pi}} \exp\left[-\eta^2\right]$$

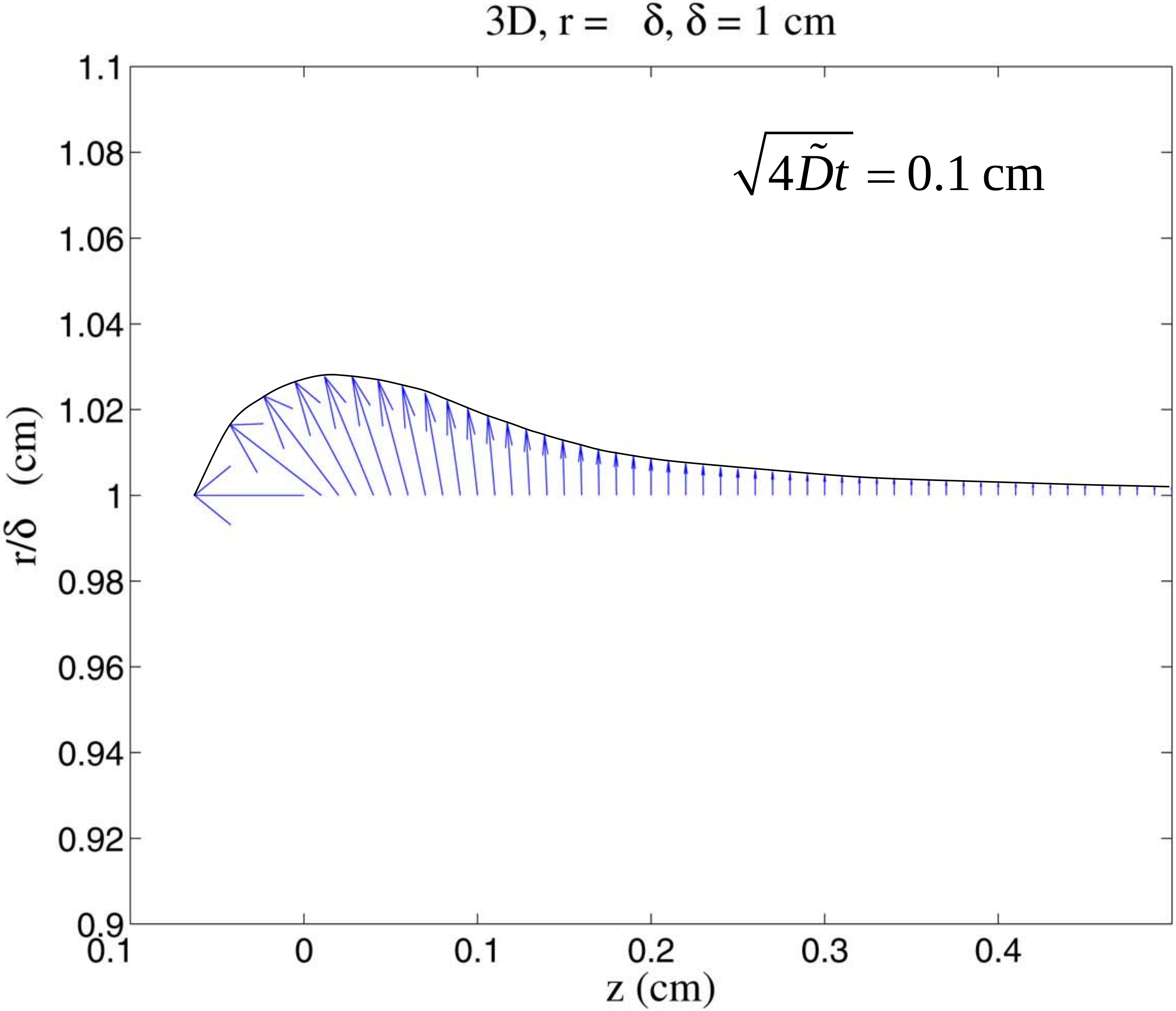
$$u_z(z,t) = \sqrt{4\tilde{D}t}\, \frac{\Delta D}{\tilde{D}} \frac{\Delta X_B}{2} \left[|\eta|\, \mathrm{erfc}|\eta| - \frac{1}{\sqrt{\pi}} \exp\left[-\eta^2\right] \right]$$

$$u_z(0,t) = \sqrt{4\tilde{D}t}\, \frac{\Delta D}{\tilde{D}} \frac{\Delta X_B}{2\sqrt{\pi}}$$

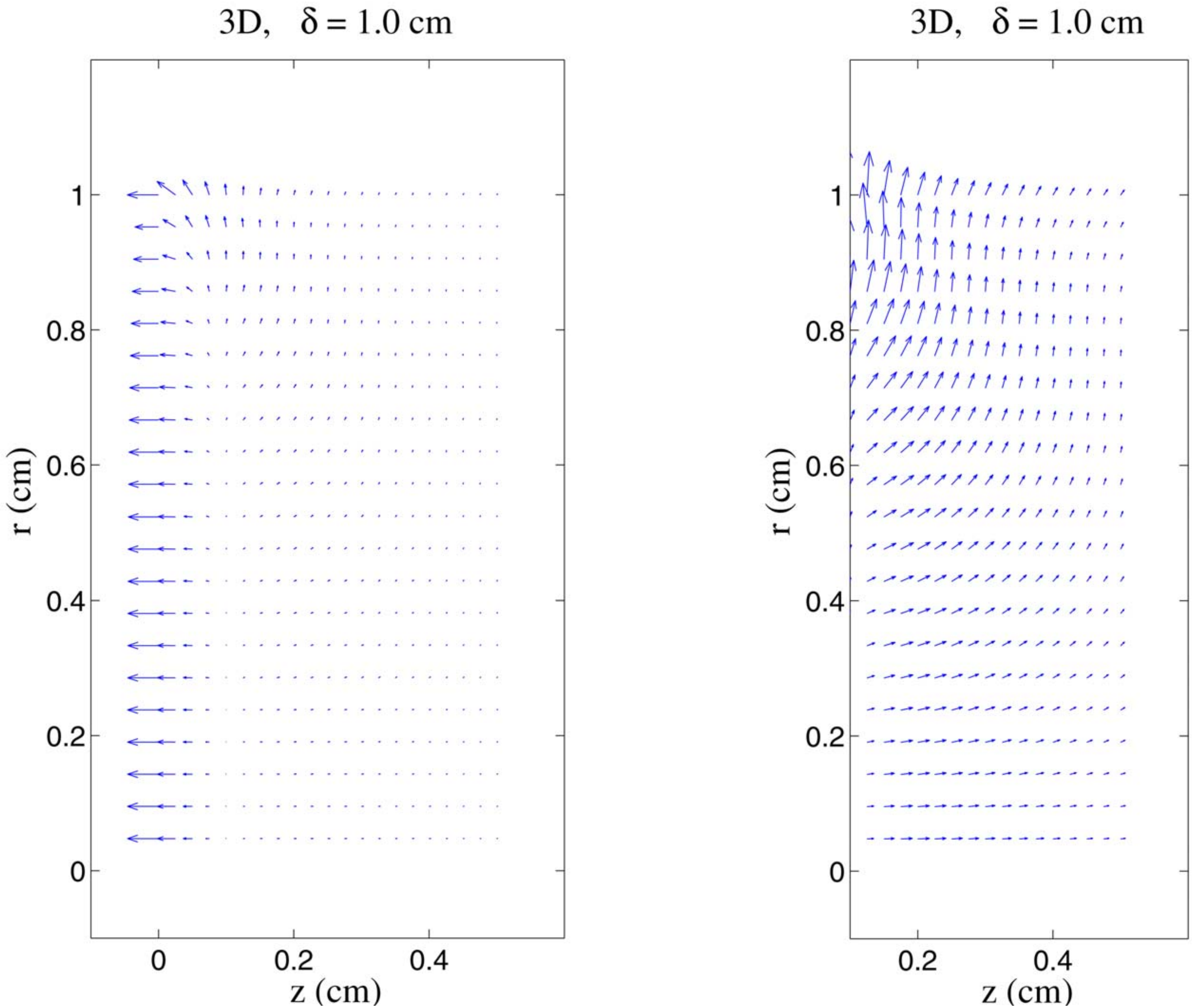


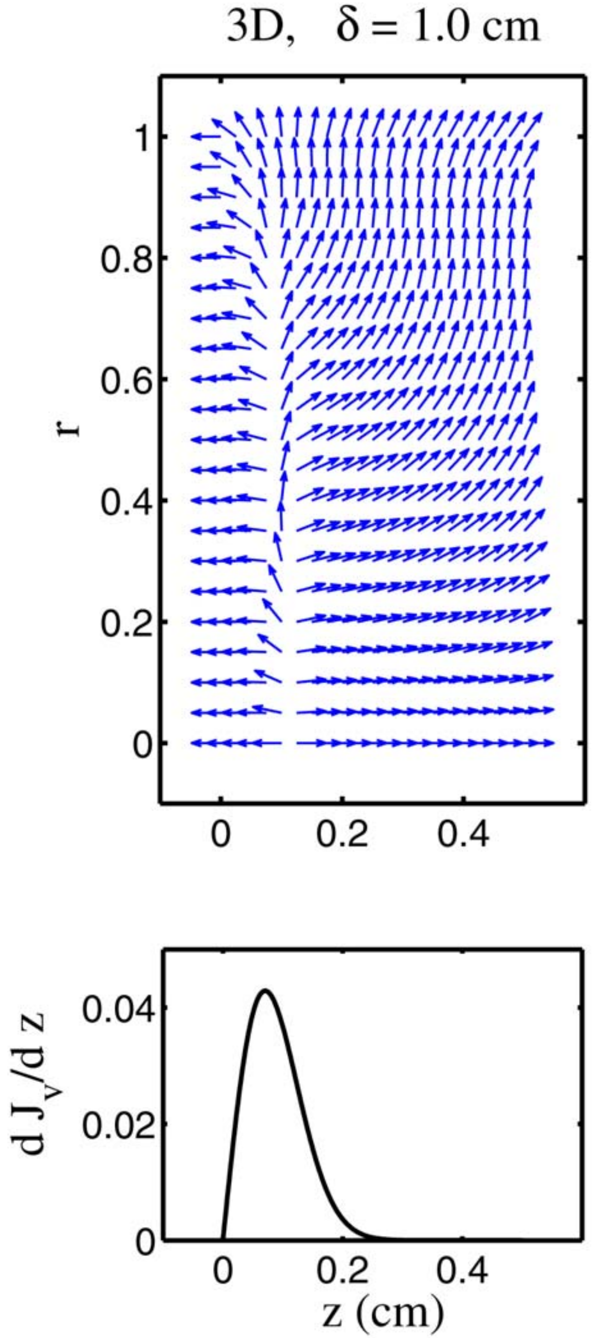
Case II – Displacement in *axial & lateral* directions

- Deformation is slaved to the diffusion… Independent of value of viscosity
- Stress level depends on viscosity
- Solution method
  - Erf diffusion solution written in terms if Fourier sine transform
  - Deformation obtained for individual sine components analytically
  - Full deformation obtained by Fourier Inversion
  - Some analytical results for limiting cases…still working



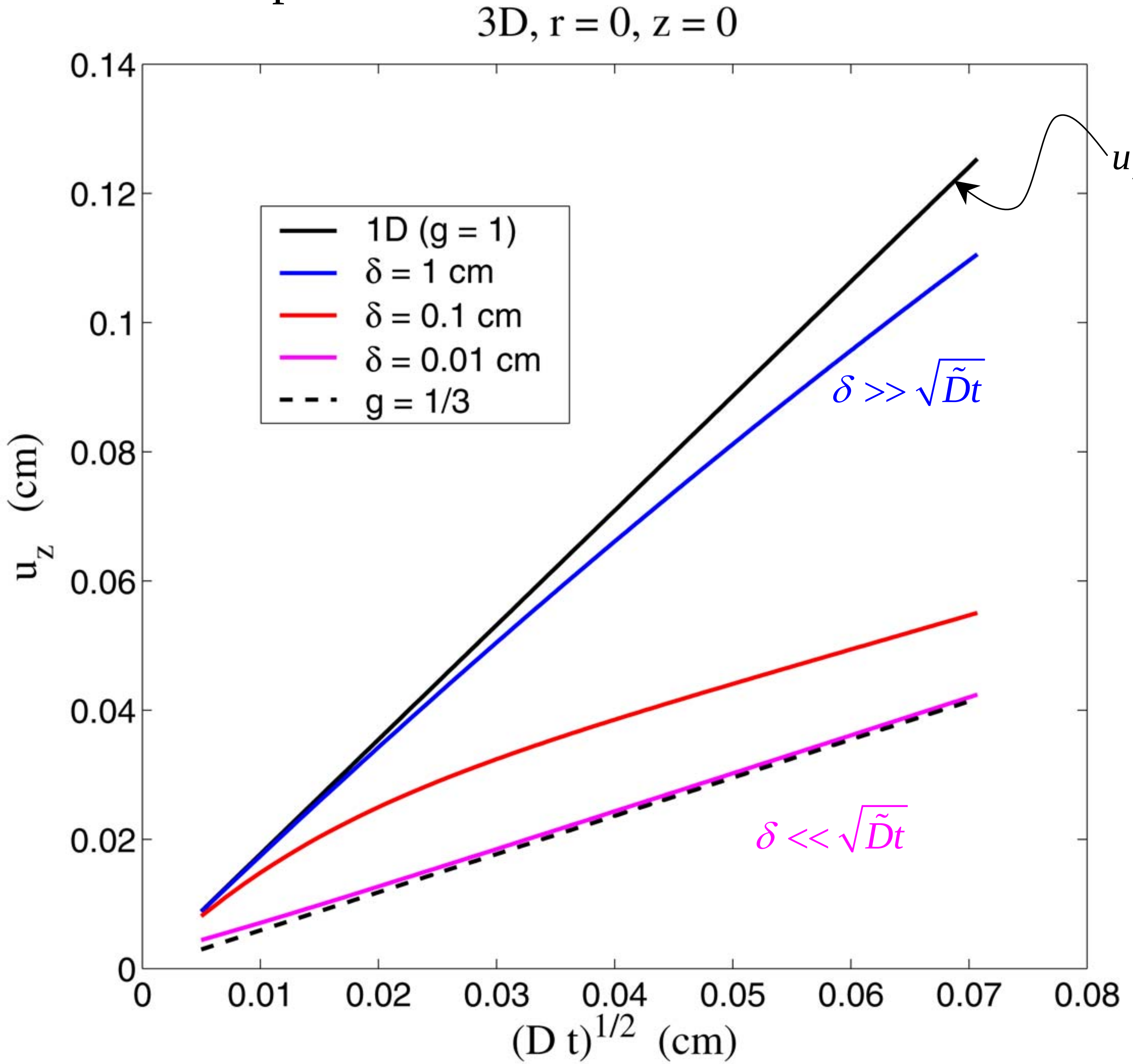
3D, r = δ, δ = 1 cm
$\sqrt{4\tilde{D}t} = 0.1$ cm
1.1
1.08
1.06
1.04
1.02
1
0.98
0.96
0.94
0.92
0.9
r/δ (cm)
0.1
0
0.1
0.2
0.3
0.4
z (cm)


3D, δ = 1.0 cm
3D, δ = 1.0 cm
r (cm)
z (cm)
0
0.2
0.4
0.6
0.8
1

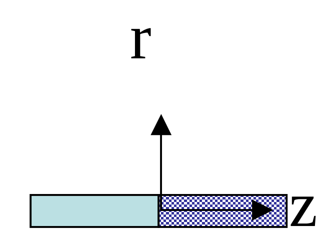


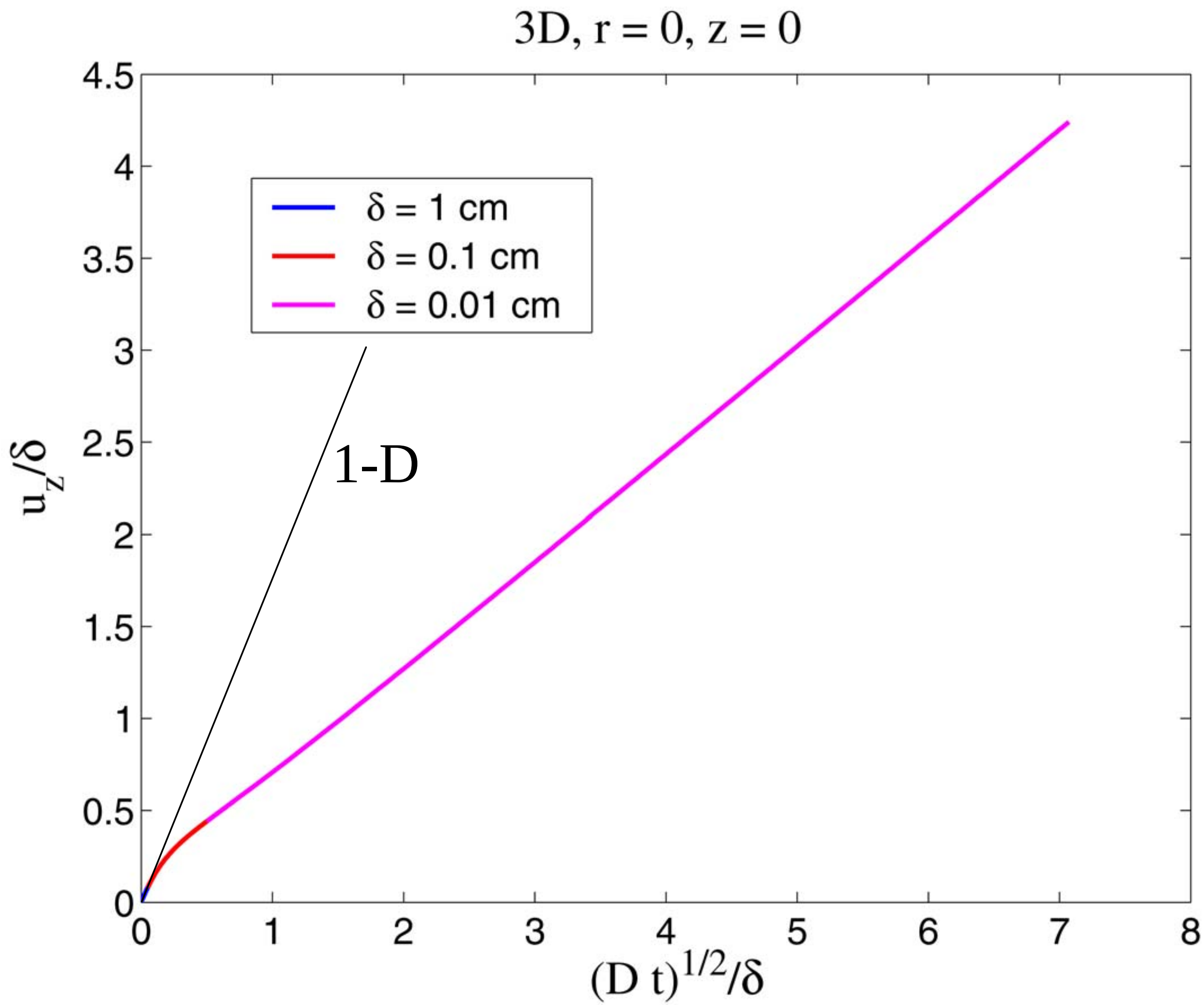
Arrows only indicate direction

Axial displacement of Kirkendall marker at rod center



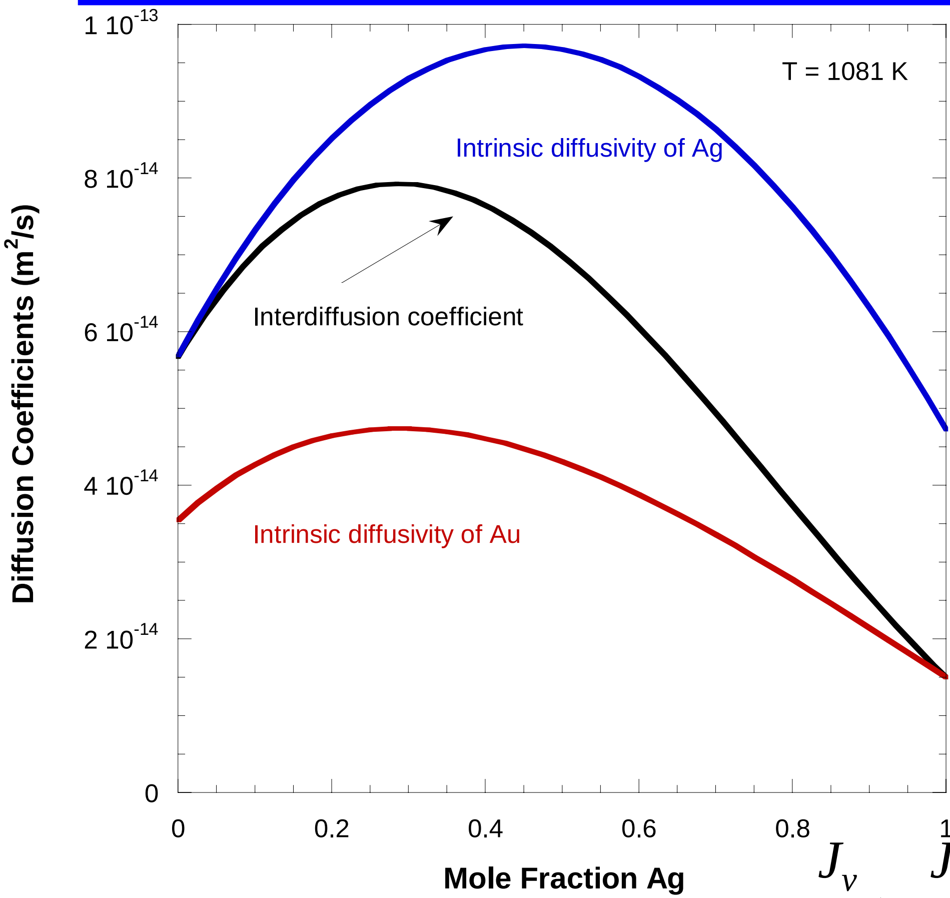
$$u_z\Big|_{z=0}^{1-D} = \sqrt{4\tilde{D}t}\,\frac{\Delta D}{\tilde{D}}\,\frac{\Delta X_B}{2\sqrt{\pi}}$$





3D, r = 0, z = 0
δ = 1 cm
δ = 0.1 cm
δ = 0.01 cm
1-D
u_z/δ
$(D\ t)^{1/2}/\delta$
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
0
1
2
3
4
5
6
7
8

Comparison of Calculation & Experiment: Need Ag-Au Diffusion Coefficients



$$D_{Ag} = 8.96 \text{ x } 10^{-14} m^2 / s$$

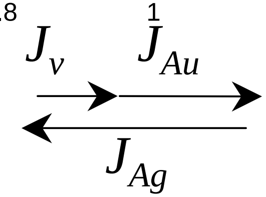
$$D_{Au} = 4.72 \text{ x } 10^{-14} m^2 / s$$

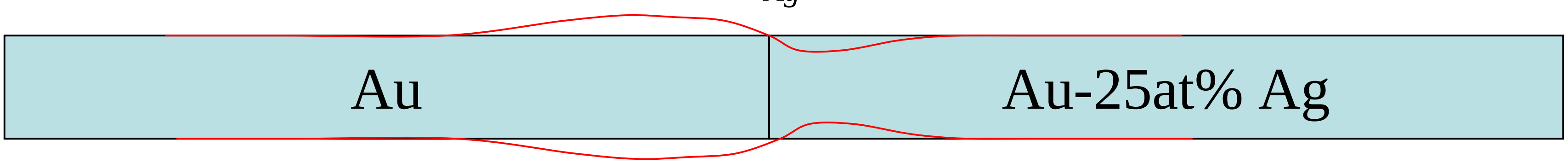
$$\tilde{D} = 7.9 \text{ x } 10^{-14} m^2 / s$$

$$\Delta D = D_{Au} - D_{Ag}$$

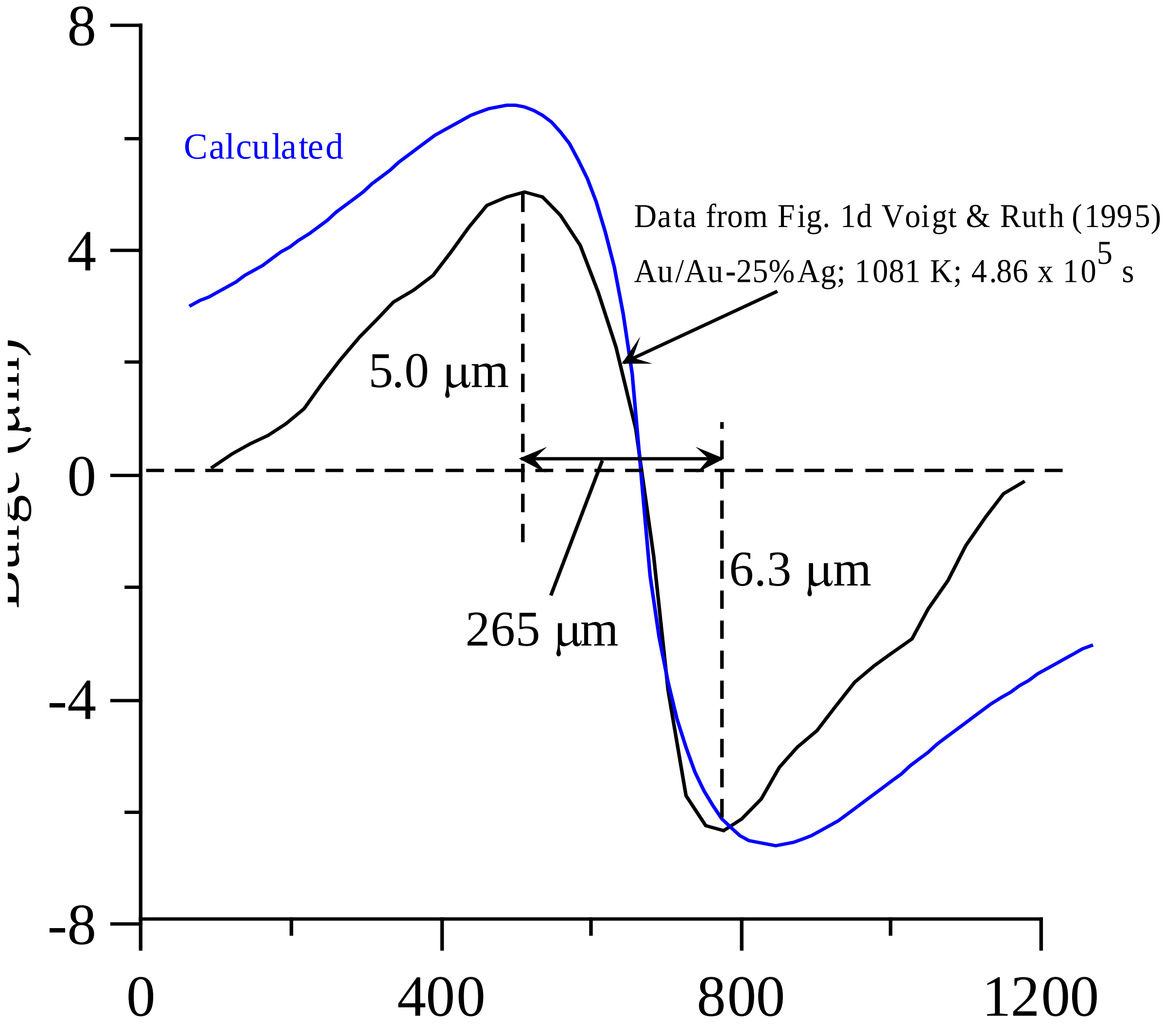
$$= -4.24 \text{ x } 10^{-14} m^2 / s$$

$\boxed{D_{Au} < D_{Ag}}$





Comparison of Calculation & Experiment



Conclusions

• A simple model is proposed to describe lateral bulging of diffusion couples that agrees approximately with experiment.

• Stress-free strain rate is assumed proportional to vacancy creation/annihilation rate

• Model recovers 1-D Kirkendall effect if displacement is constrained to be in the diffusion direction.

• More realistic zero surface traction BC leads naturally to bulging.

• If the lateral dimension of diffusion sample, $\delta >> (Dt)^{1/2}$, marker motion at the sample center is the same as the 1-D for the 1-DKirkendall effect.

• If $\delta << (Dt)^{1/2}$, marker motion at the sample center is ½ (slab geometry) or 1/3 (rod geometry) of the 1-D Kirkendall displacement.

• Full flow pattern of the sample is computed.