Modeling of Lateral Bulging of Diffusion Couples

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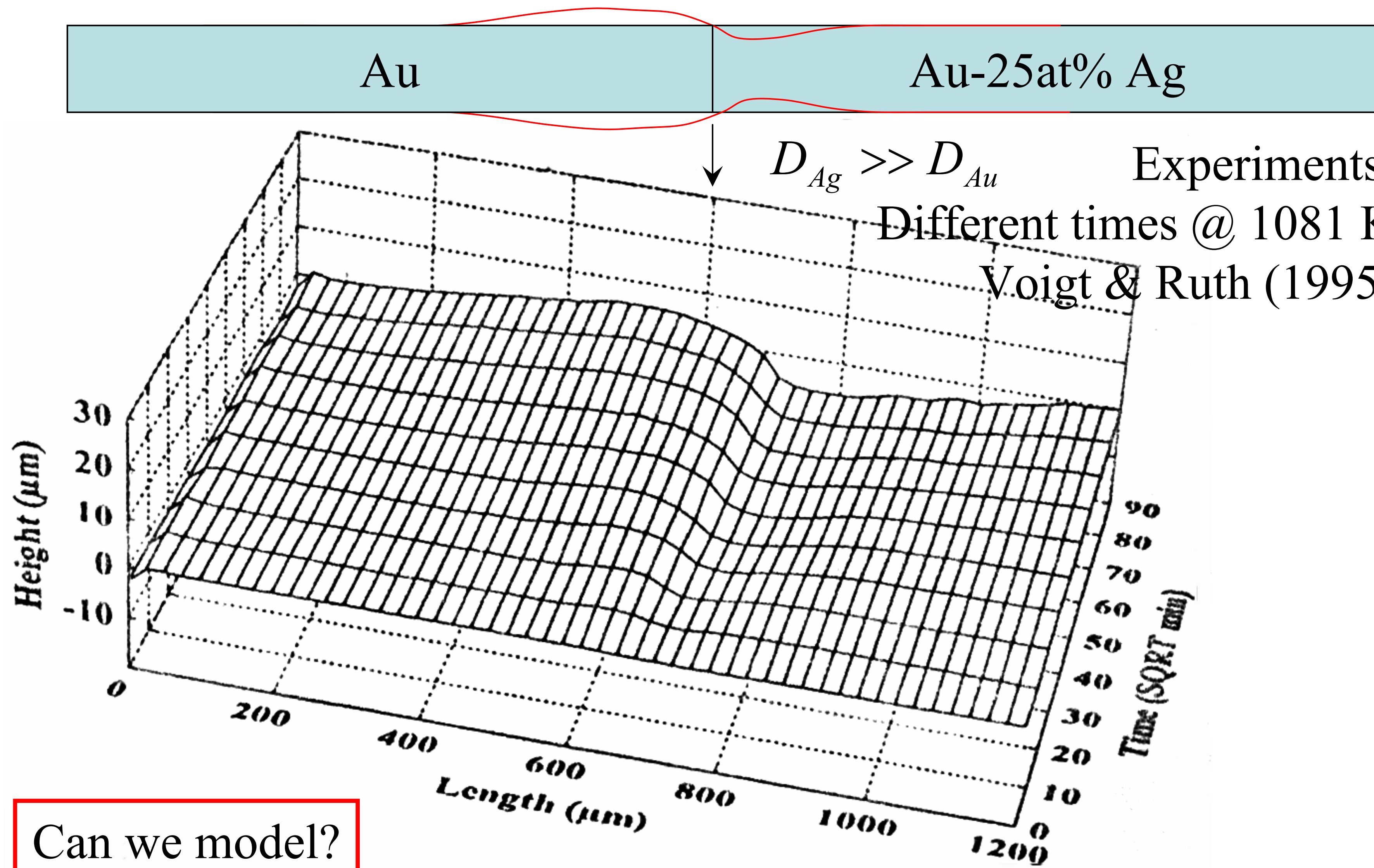


Experiments:
Different times @ 1081 K
Voigt & Ruth (1995)

Can we model?

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

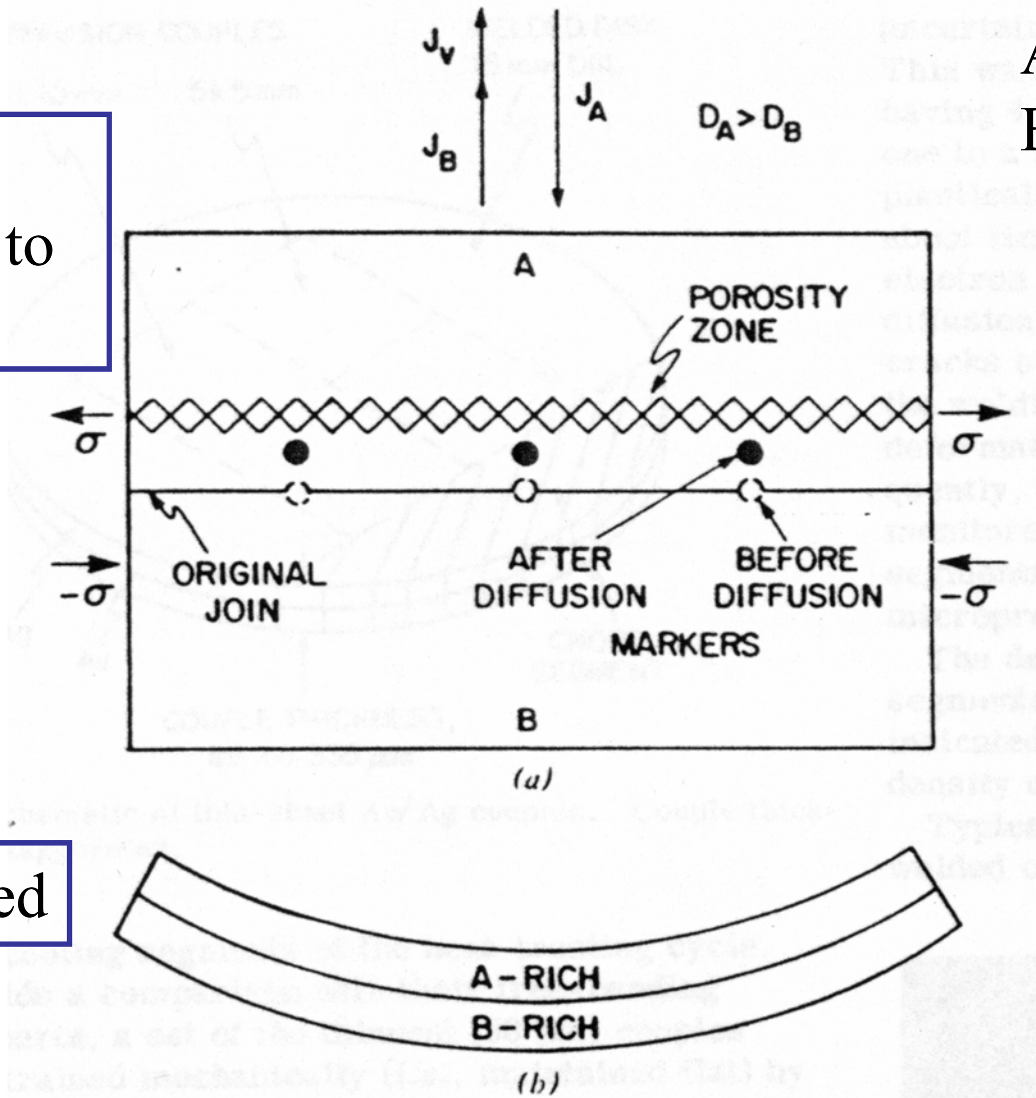
Stress if constrained to remain flat

Unconstrained

A=Ag
B=Au

Ag-rich

Au-rich



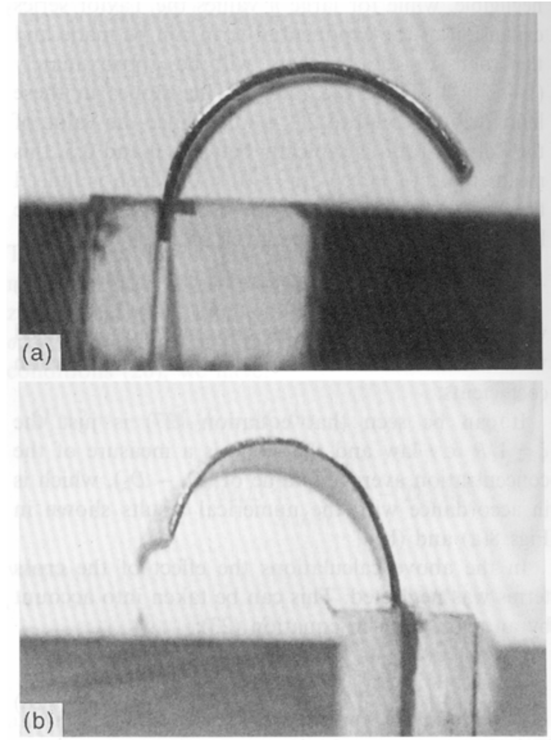
D. W. Stevens & G. W. Powell, Met. Trans 8A(1977) 1531

"Diffusion Induced Bending of Thins Sheet Couples: Theory and Experiment for Ti-Zr," I Daruka et al. Acta Mater. 44(1996), 4981-4993.

- 0.1 mm thick bonded Ti & Zr sheets, 2-5 hrs anneal @1183, 1233, 1273 K
- Radius as small as 4 mm

D_{Zr}-D_{Ti} = 5 x10^{-14} m^2/s

Dtilda = 1.7 x 10^{-13} m^2/s



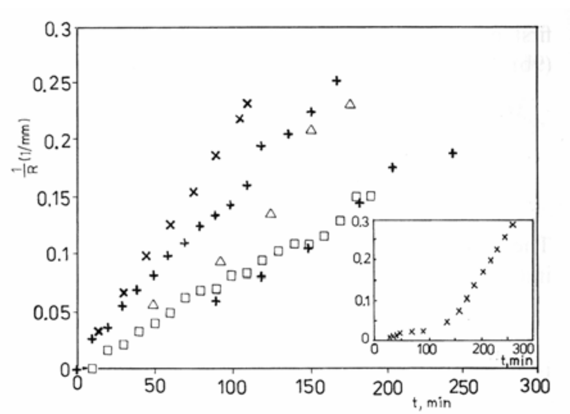


Fig. 10. Curvature (1/R) vs annealing time, t, functions for five samples at T = 1223 K (see also text). The insert shows the 1/R vs t function as measured, i.e. before the correction for the thermal expansion.

DARUKA et al.:

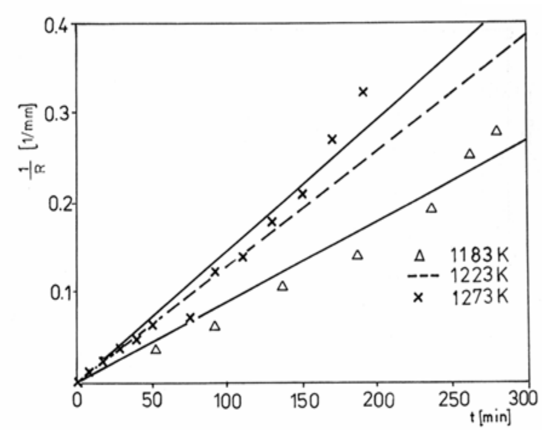
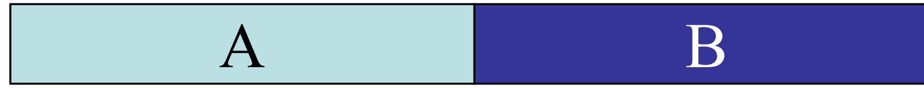


Fig. 12. 1/R vs t functions at three different temperatures (the average of the curves shown in Fig. 10 is illustrated by a dashed line).

Review of 1-D Kirkendall Effect



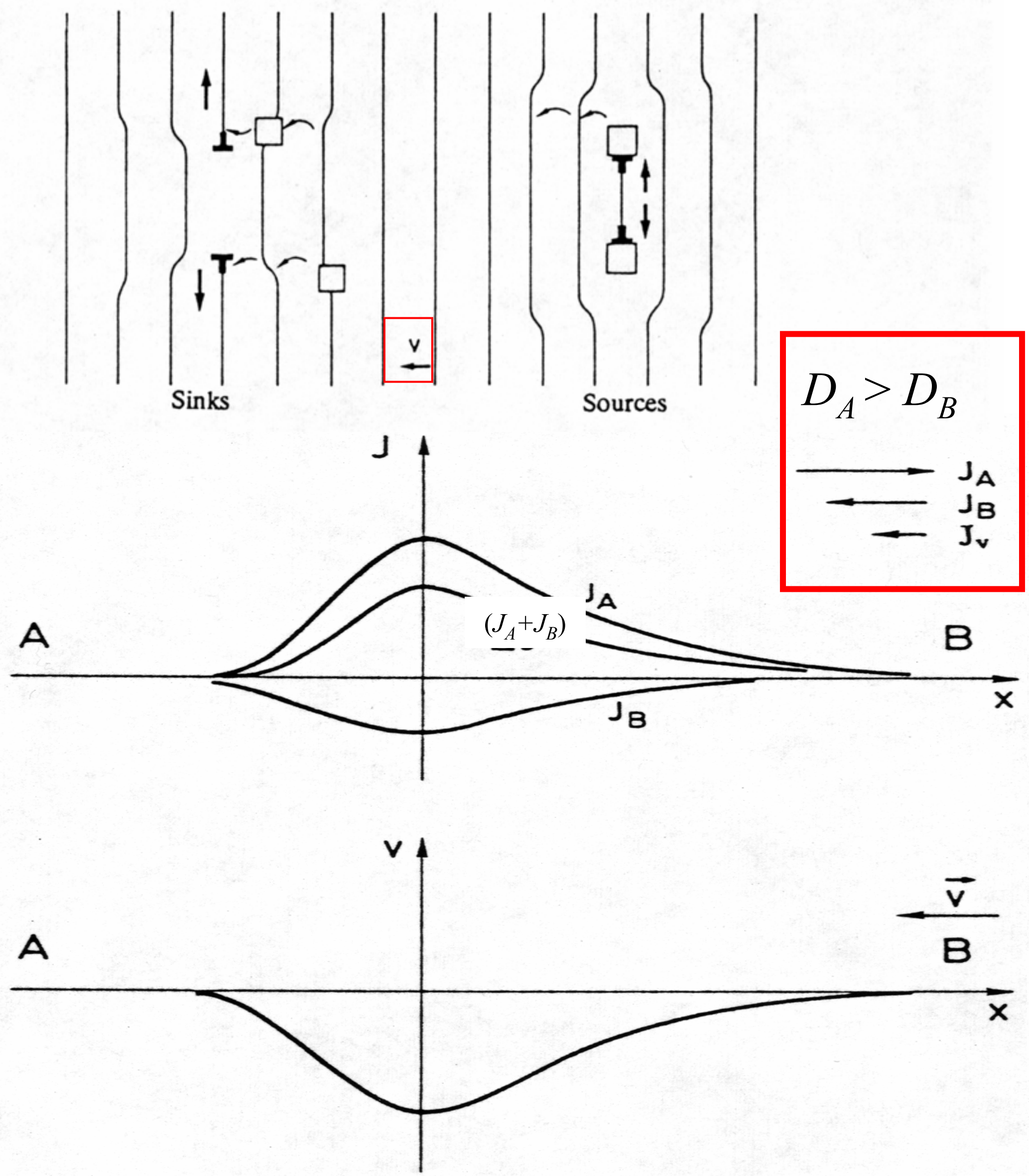
Simplest Form

- Diffusion by vacancy mechanism
- Separate fluxes for A & B wrt lattice
- $\bar{V}_A = \bar{V}_B = \bar{V}_V = \bar{V}$
- Vacancy creation/annihilation rate is precisely that to maintain equilibrium vacancy content

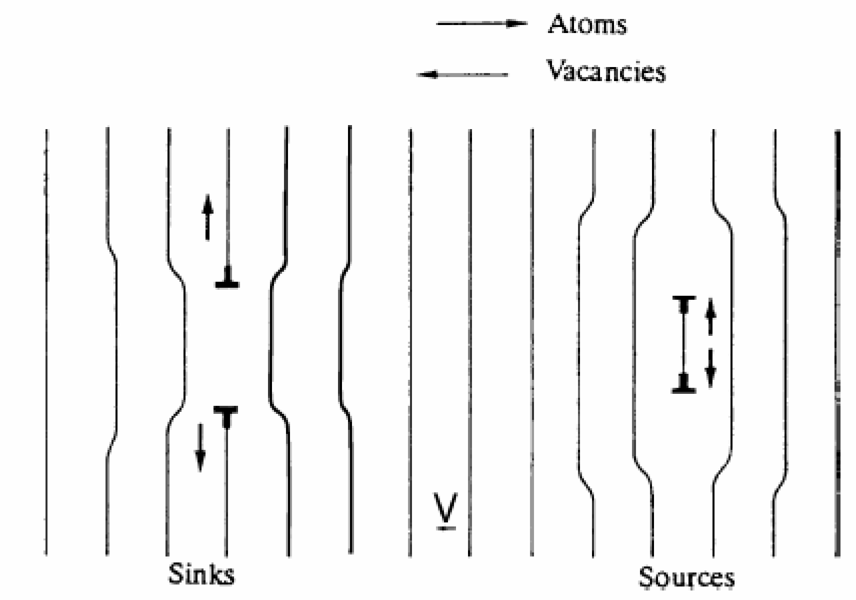
  i.e., $s_v = -\dfrac{\partial J_v}{\partial z}$
- No voids
- Solve diffusion equation with no advection for X_B in z-direction w/ $\tilde{D} = D_A X_B + D_B X_A$
- Lattice moves wrt observer in lattice outside diffusion zone with velocity

  $$v_z(z,t) = -\left(D_A - D_B\right)\frac{\partial X_B}{\partial z}$$

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



Stress-Free Strain Rate



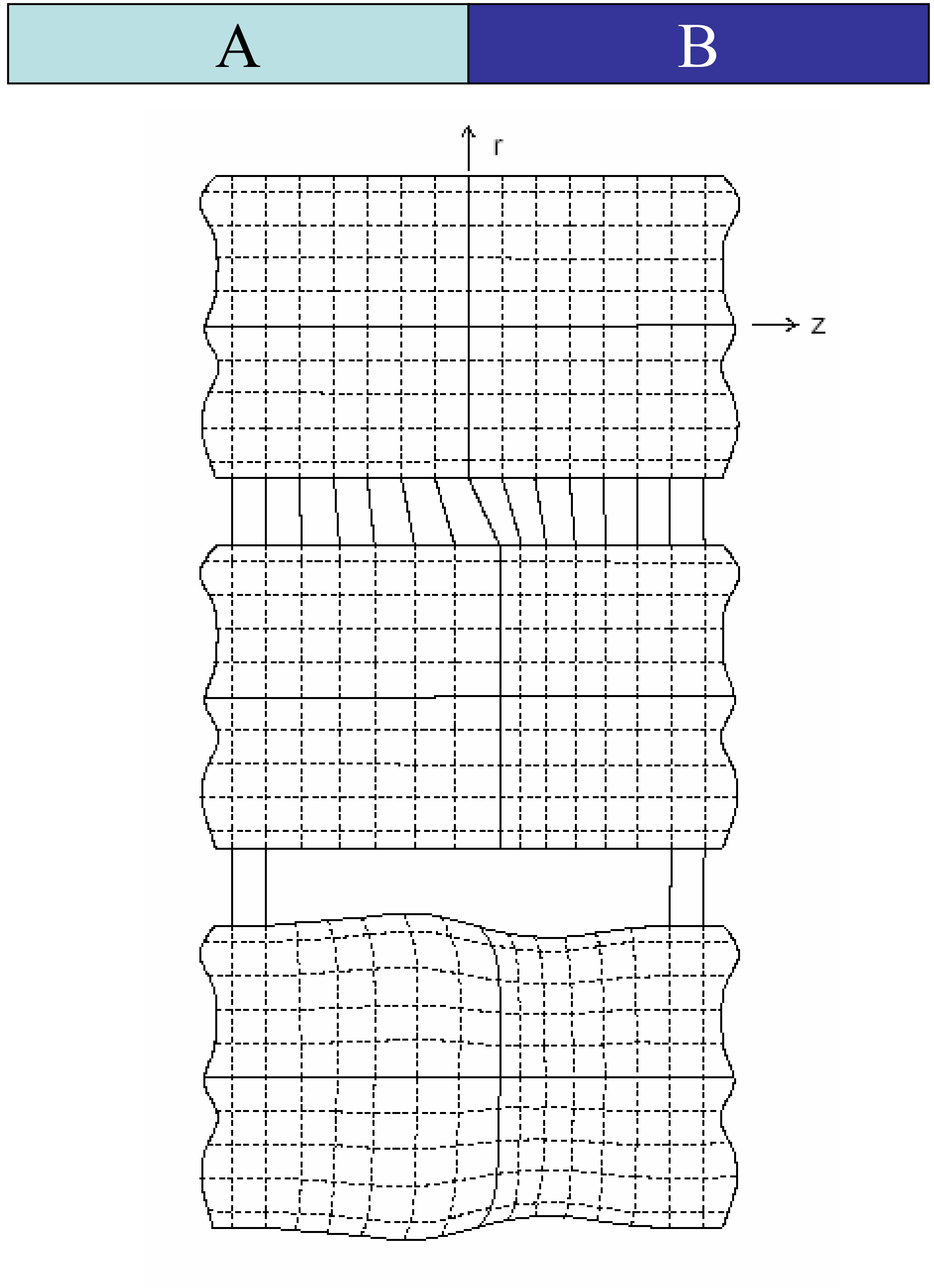
- Climb occurs perpendicular to Burger's vector <u>b</u> & line <u>l</u>
- Material may contain dislocations with a variety of <u>b</u>'s and <u>l</u>'s
- Material may also be polycrystalline
- If one averages over volumes containing many sources/sinks
- Can assume *stress-free* dilation rate due to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

$$\dot{\varepsilon}_{ij}^{0} = \frac{1}{3}\delta_{ij}\bar{V}\nabla \cdot J_v = -\frac{1}{3}\delta_{ij}\bar{V}\nabla \cdot (J_A + J_B)$$

Initial

1-D

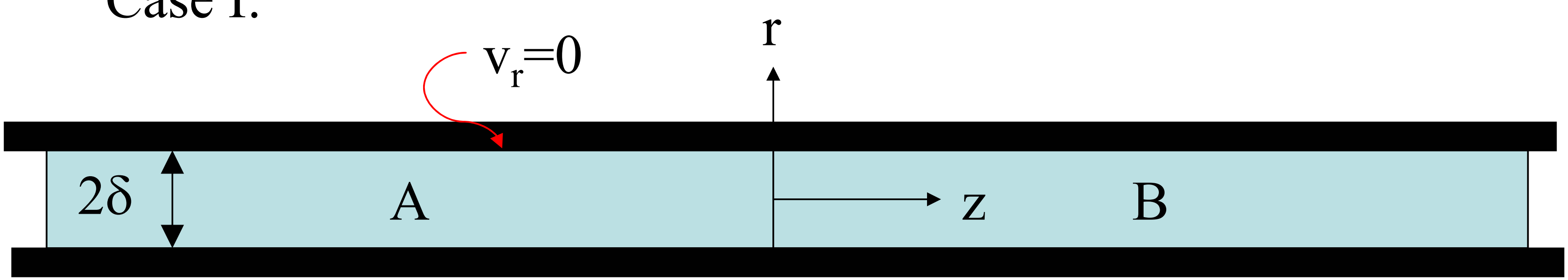
2-D or 3-D



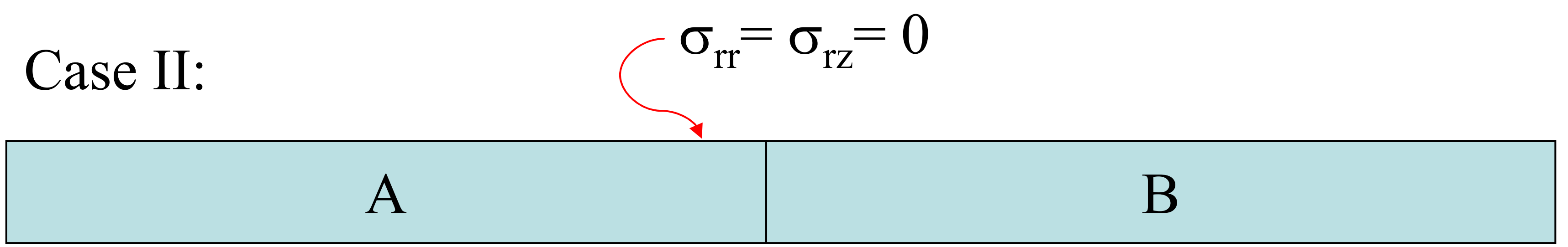
$D_A < D_B$

Assume radial symmetry

Case I:



Case II:



Analysis of Deformation; Small Strains

$$\dot{\varepsilon}_{ij}^{T} = \frac{1}{2}\left(\frac{\partial \mathrm{v}_i}{\partial x_j} + \frac{\partial \mathrm{v}_j}{\partial x_i}\right)$$ Definition

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

$$\dot{\varepsilon}_{ij}^{0} = \frac{1}{3}\delta_{ij}\bar{V}s_v$$ 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: $\dot{\varepsilon}_{kk}^{T} = \frac{\partial \mathrm{v}_k}{\partial x_k} = \nabla \cdot \mathrm{v}$

$$= \dot{\varepsilon}_{kk}^{0} = \bar{V}s_v$$

Analysis of Interdiffusion

- Decouples from Stress (for $\bar{V}_1 = \bar{V}_2 = \bar{V}_V = \bar{V}$)
- Still treat as 1-D for small deformation
  (advection normal to diffusion direction can be neglected)
- Diffusion Equation

$$\nabla \equiv \frac{\partial}{\partial z}$$

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

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

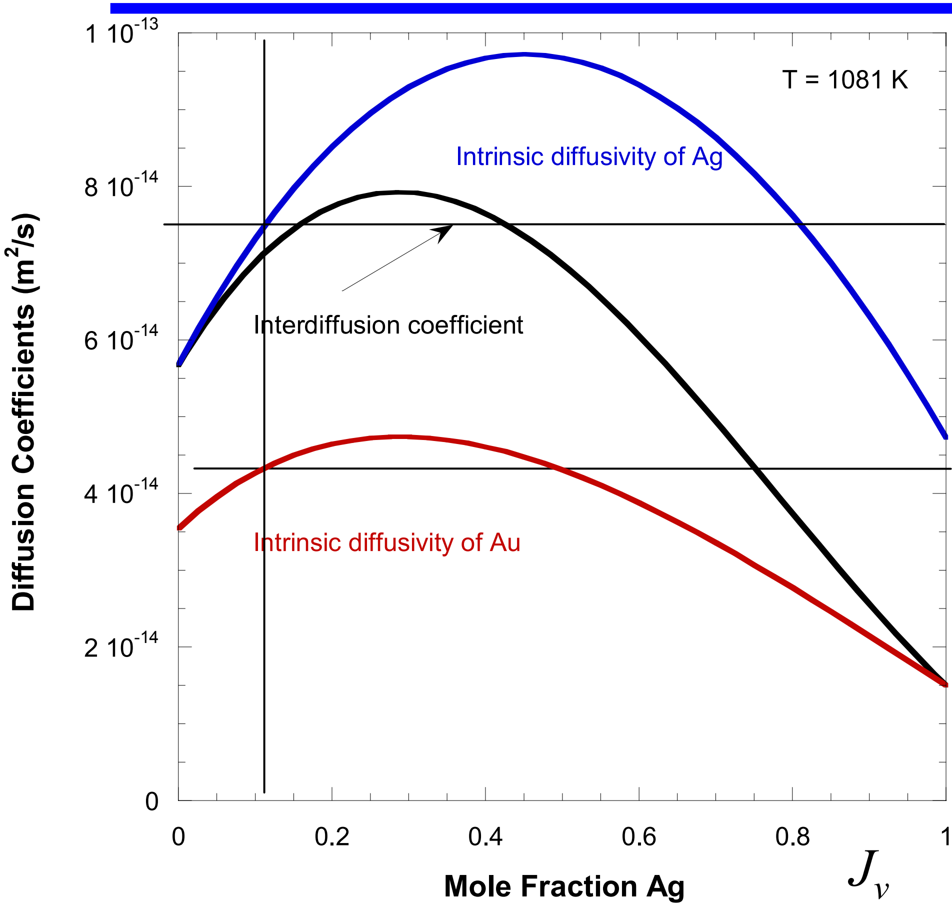
D_1 and D_2 are the intrinsic (lattice) diffusion coefficients

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

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

$$\Delta D = D_1 - D_2$$

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



Thermodynamics:
'S. Hassam et al.,Metall. Trans. 21A (1990) 1877-1884, Report at KTH; Ag-Au-Si'
Diffusivities:
Landholt-Börnstein, (1990); Self-diffusion of Ag in fcc Ag.'
Ch. Herzig and D. Wolter, Z. Metallkd. 65(1974)273.; Impurity diffusion of Ag in fcc Au.
W. C. Mallard et al., Phys. Rev. 129(1963)617.; Impurity diffusion of Au in fcc Ag.'
D. Duhl et al., Acta Met. 11(1963)1; self-diffusion of Au in fcc Au
Mobility assessment: ThermoCalc AB
Delivery to us: C.E. Campbell

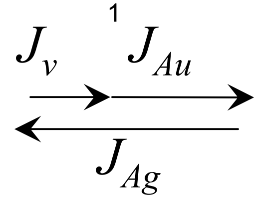
$$D_{Ag} = 8.96 \text{ x } 10^{-14} m^2 / s$$

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

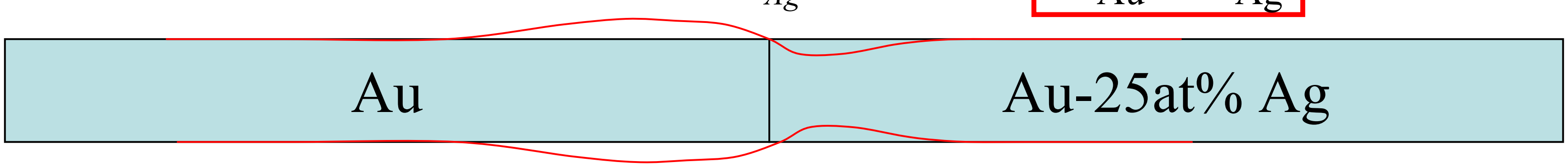
$$\tilde{D} = 7.9 \text{ x } 10^{-14} m^2 / s$$

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

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



$$D_{Au} < D_{Ag}$$



Simplification

If $\tilde{D}$ and ΔD can be treated as constants (small ΔX_2):

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

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

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

Case1: Displacement only in *axial* direction. Classical Kirkendall shift of markers on original (z=0) interface

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

$D_A < D_B$

$\Delta D = D_A - D_B < 0$

$\Delta X_B > 0$

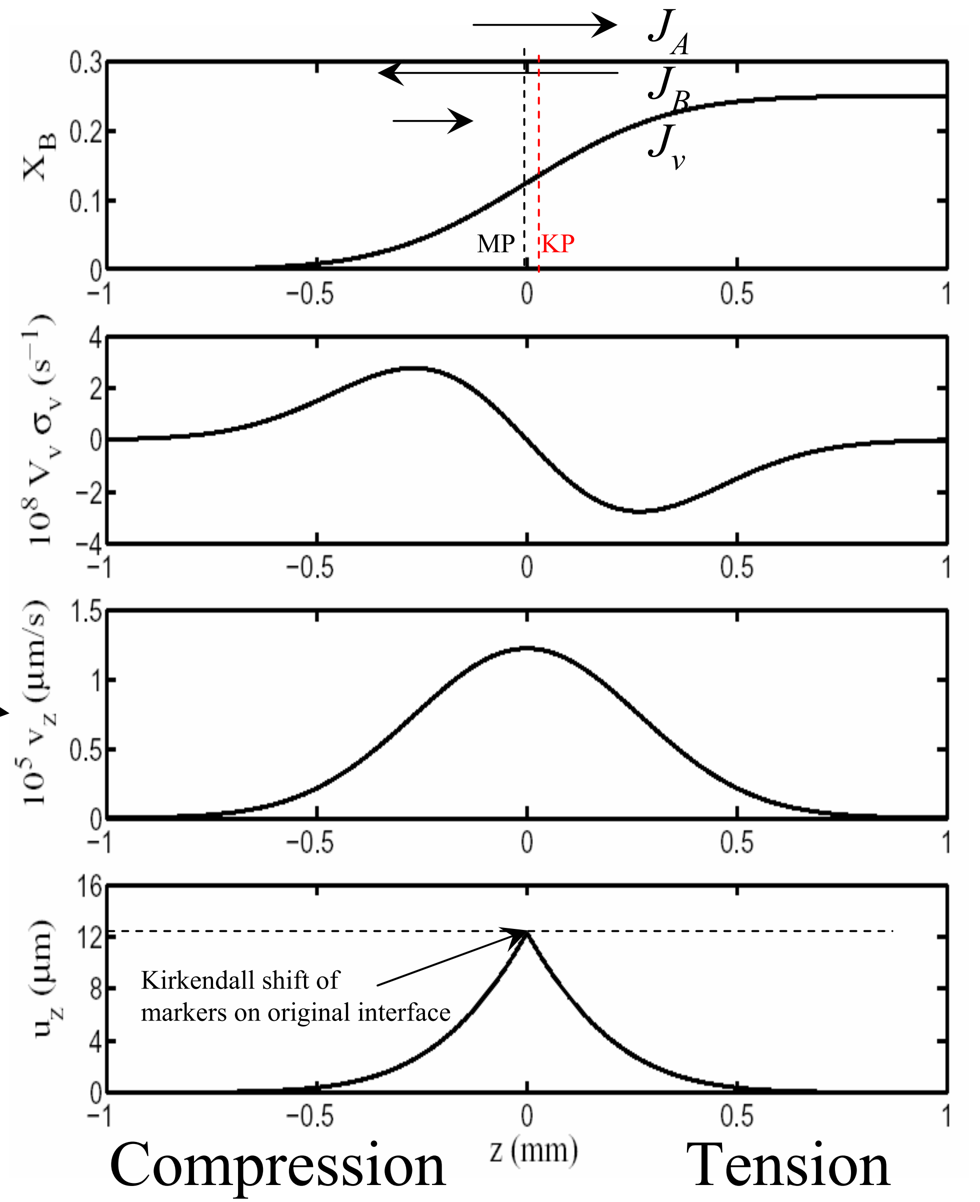
$$X_B(z,t) = \bar{X}_B + \frac{\Delta X_B}{2} erf[\eta]$$

$$\bar{V}_v s_V = -\Delta D \frac{\partial^2 X_B}{\partial z^2}$$

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

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

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



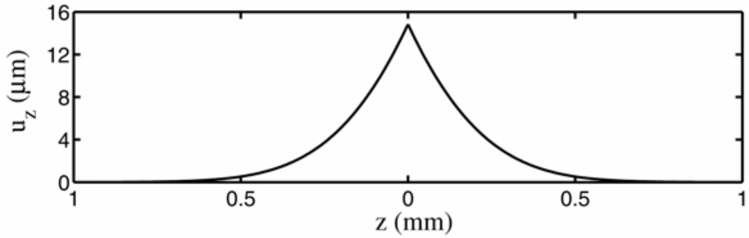
Case II – Displacement in *axial & lateral* directions

- Deformation is slaved to the diffusion… Independent of value of viscosity (and elastic modulus)

- Stress level depends on viscosity (and elastic modulus)

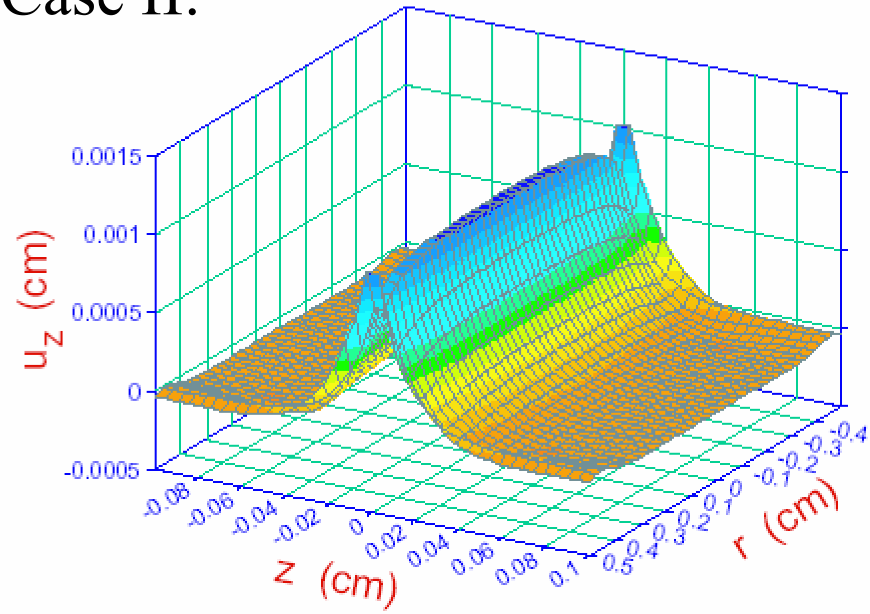
- Solution method
  - Erf diffusion solution written in terms if Fourier sine transform
  - Deformation obtained for individual sine components analytically
  - Full deformation obtained by Fourier Inversion
  - Some analytical results for limiting cases

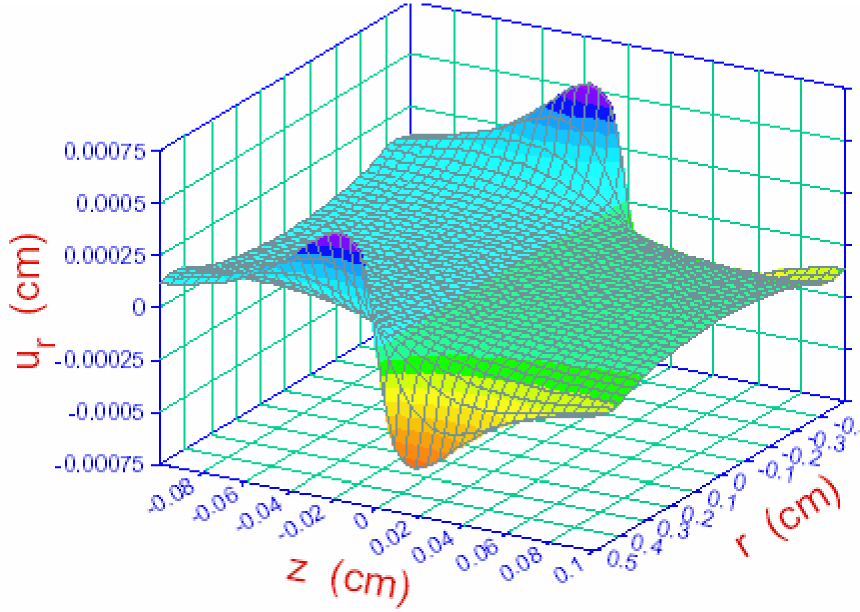
Case I:

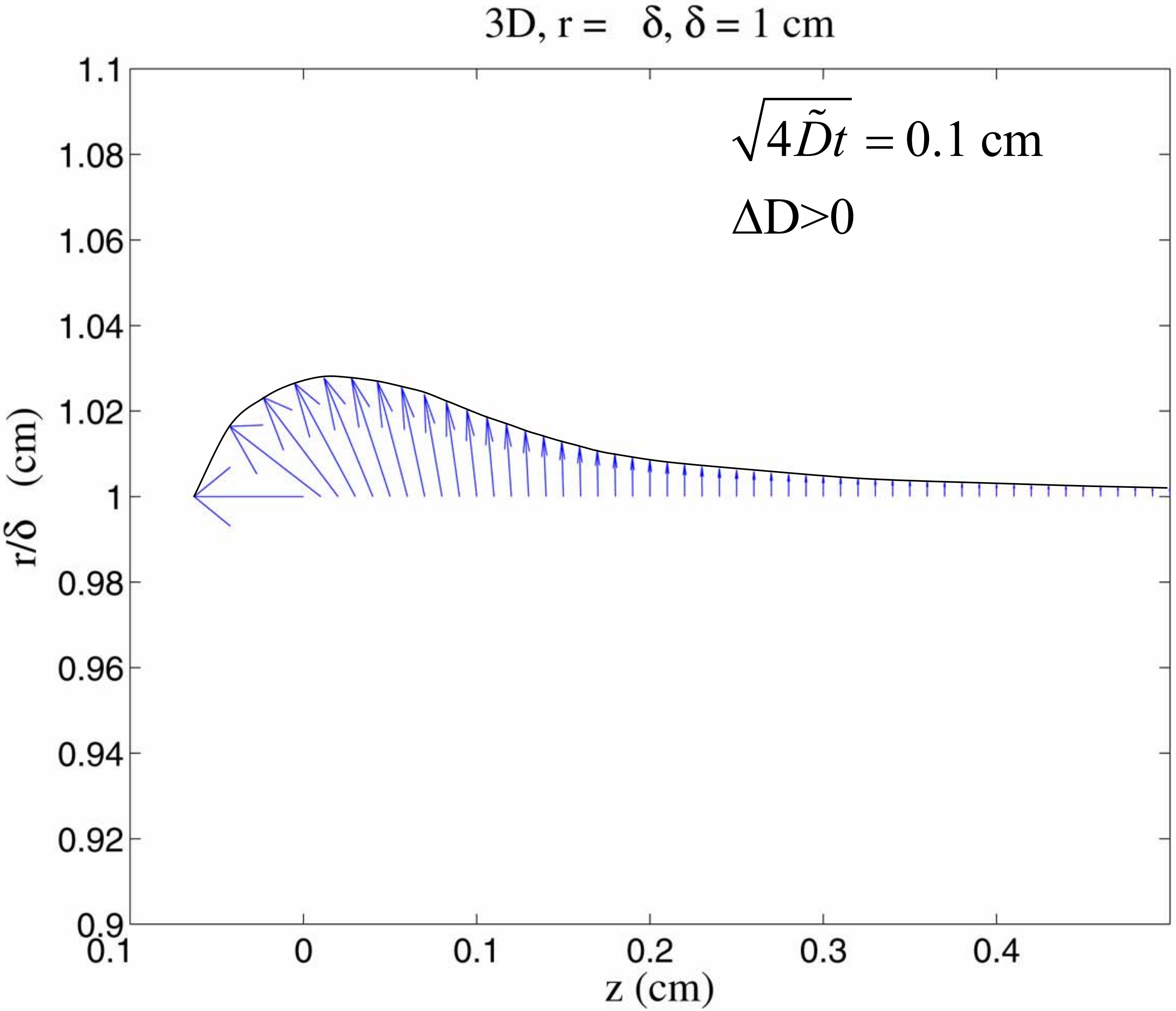


Case II:

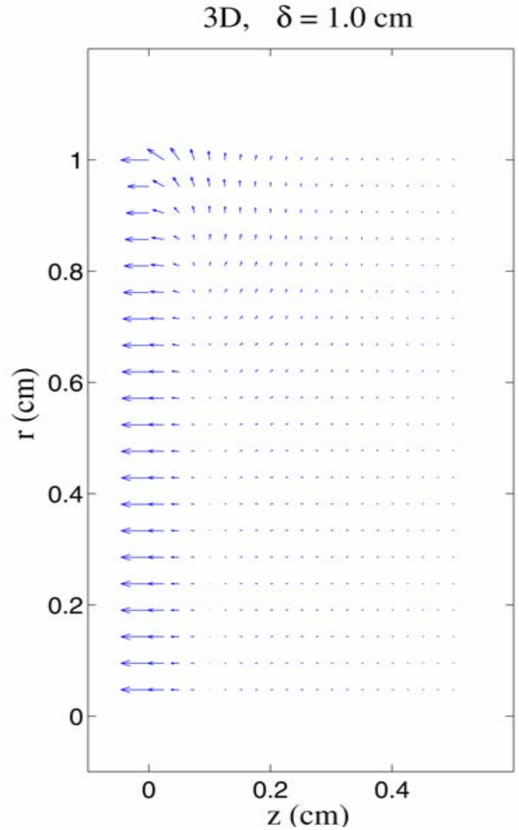
Cylinder, $\delta >> \sqrt{4\widetilde{D}t}$

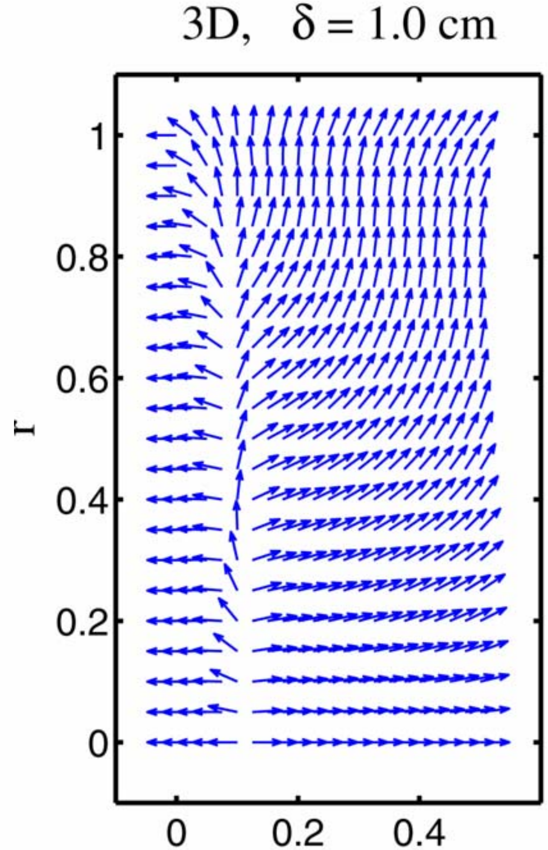


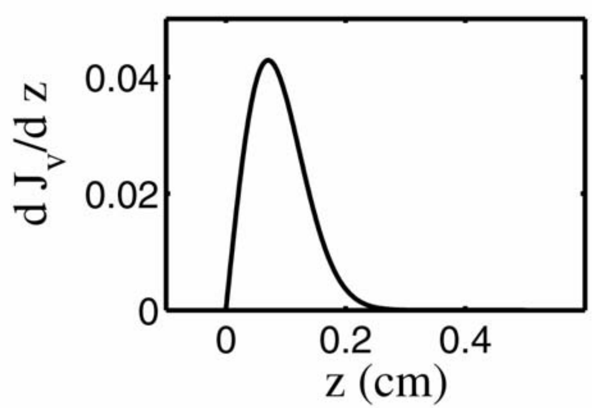




3D, r = δ, δ = 1 cm
$\sqrt{4\tilde{D}t} = 0.1$ cm
ΔD>0
r/δ (cm)
1.1
1.08
1.06
1.04
1.02
1
0.98
0.96
0.94
0.92
0.9
0.1
0
0.1
0.2
0.3
0.4
z (cm)







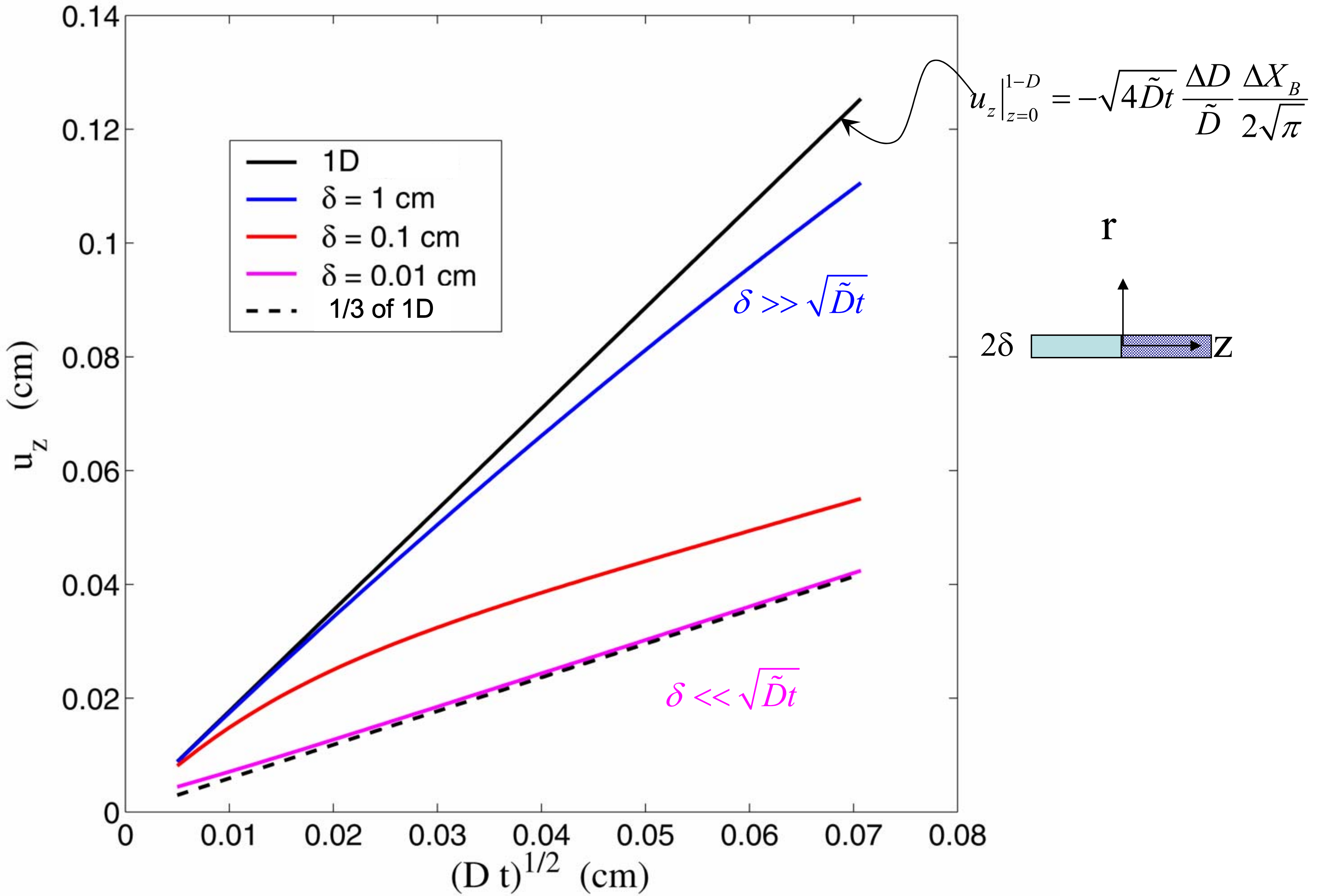
$\sqrt{4\tilde{D}t} = 0.1$ cm

$\Delta D > 0$

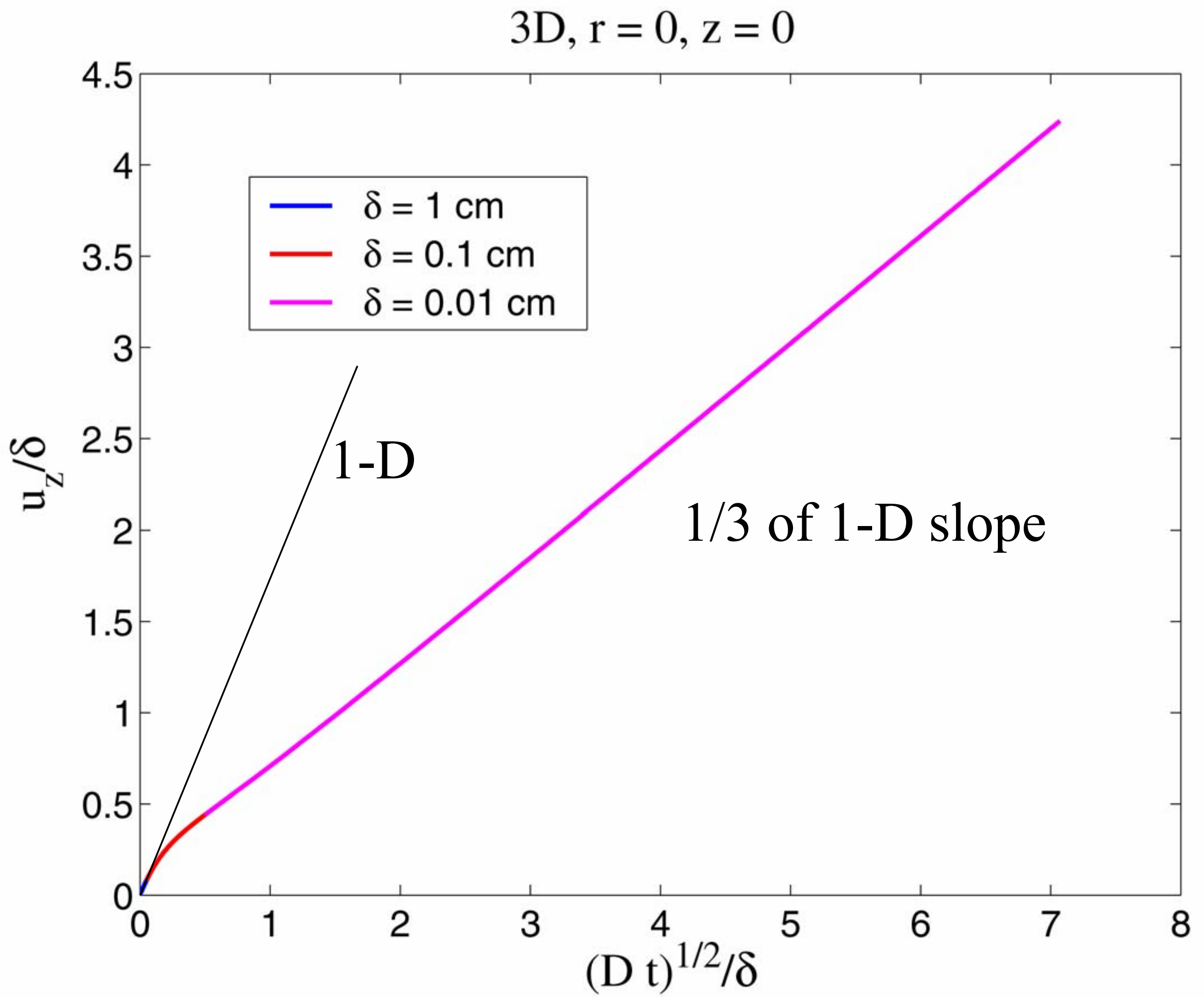
Arrows only indicate direction

Axial displacement of Kirkendall marker at rod center (ΔD<0)

3D, r = 0, z = 0



Collapse to single curve



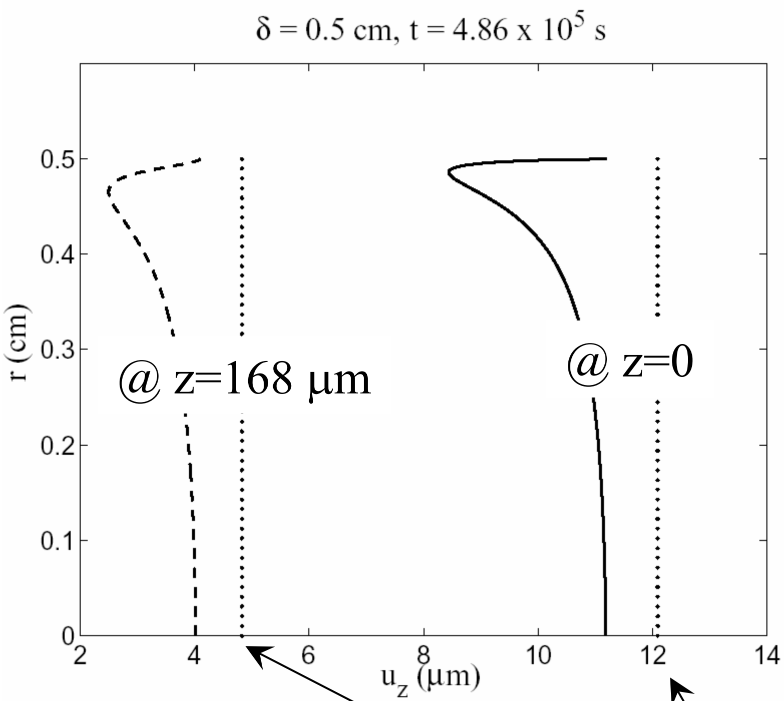


Figure 8: The dimensional horizontal displacement u_z at $t = 4.86(10^5)$ s and $\delta = 0.5$ cm for $0 < r < \delta$ at $z = 0$ (solid curve) and near the bulge minimum at $z = 168$ μm (dashed curve). The corresponding one-dimensional displacements are shown as dotted lines.

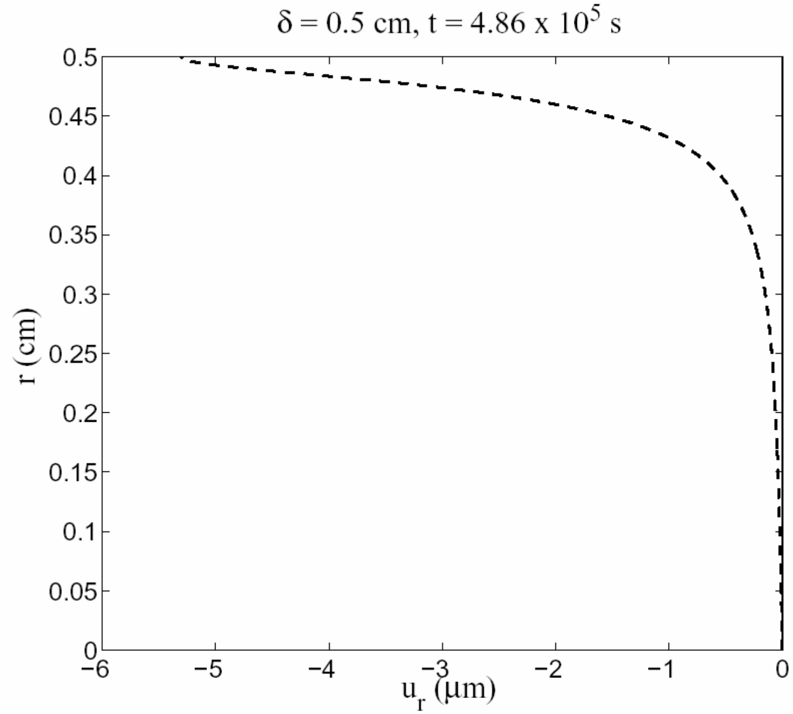
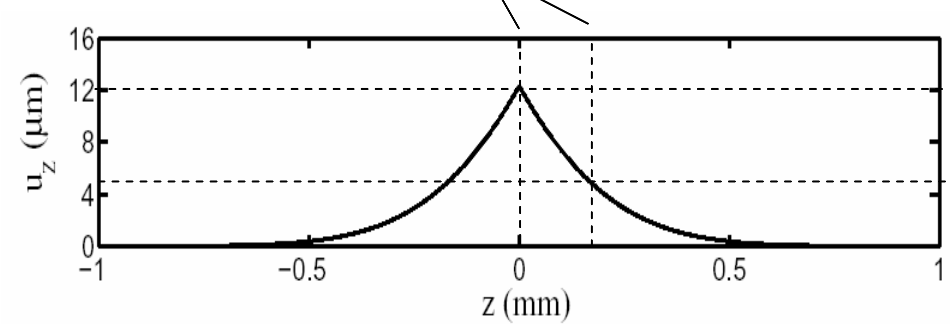


Figure 9: The dimensional vertical displacement u_r at $t = 4.86(10^5)$ s and $\delta = 0.5$ cm for $0 < r < \delta$ near the bulge minimum at $z = 168$ μm (dashed curve); the vertical displacement vanishes at $z = 0$.

1-D result



Radial displacement

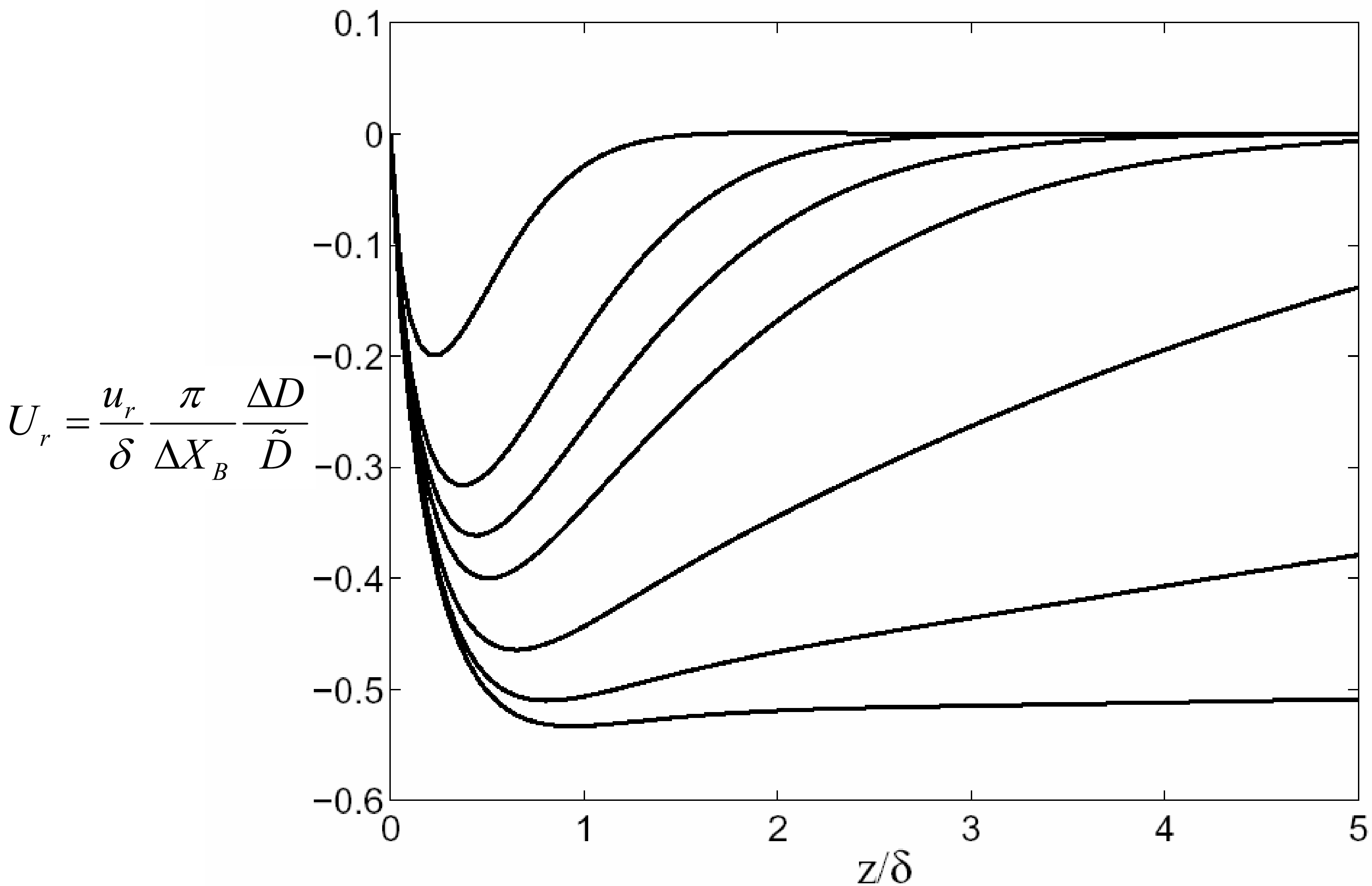


Figure 3: The dimensionless vertical displacement U_r at the outer surface for $0 < z/\delta < 5$ and (from top to bottom) $\tilde{D}t/\delta^2 = 0.1$, 0.5, 1.0, 2.0, 10.0, 100.0, and 10, 000.

Comparison of Calculation & Experiment

Au	Au-25at% Ag

$D_{Au} < D_{Ag}$

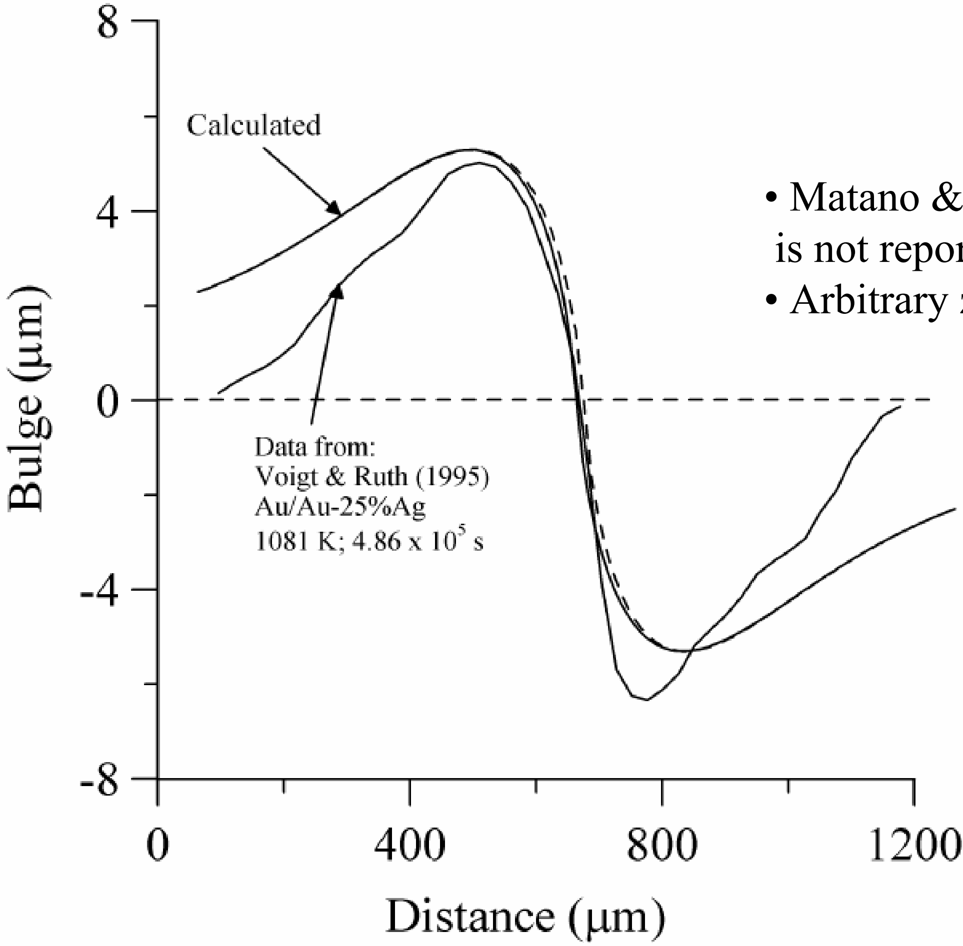


Figure 6: A comparison of the measured bulge and the calculated bulge, $u_r(\delta, z)$ [dashed], and $u_r(\delta, z + u_z)$ [solid].

- Matano &/or Kikendall interface position is not reported
- Arbitrary z axis shift of calculation

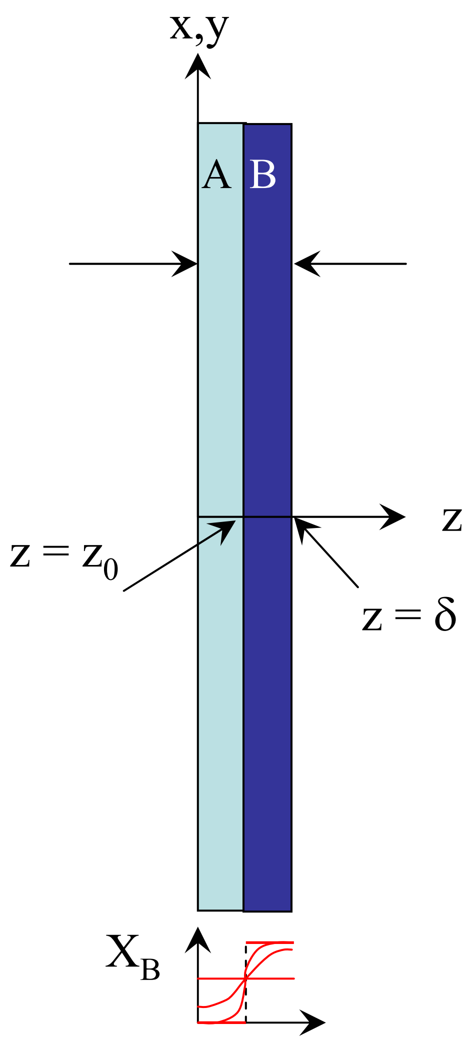
Simple free beam model

Curvature, $\kappa(t)$

Neutral axis position @ $z = c(t)$

$$\sigma_{zz} = 0$$

$$\varepsilon_{xx}^{T}(z,t) = \varepsilon_{yy}^{T}(z,t) = \kappa(t)\left(z - c(t)\right)$$



Balance of Forces, Balance of Moments

$$\begin{cases} 0 = \int_0^\delta \sigma_{xx} dz = \int_0^\delta E'\left(\varepsilon_{xx}^{T} - \varepsilon_{xx}^{0}\right) dz \\ 0 = \int_0^\delta z\sigma_{xx} dz = \int_0^\delta zE'\left(\varepsilon_{xx}^{T} - \varepsilon_{xx}^{0}\right) dz \end{cases}$$

Take $\dfrac{\partial}{\partial t}$

$$\begin{cases} 0 = \dfrac{\partial}{\partial t}\int_0^\delta \left[\left[\kappa(t)\left(z - c(t)\right)\right] dz\right] dz - \int_0^\delta \dot{\varepsilon}_{xx}^{0} dz \\ 0 = \dfrac{\partial}{\partial t}\int_0^\delta z\left[\kappa(t)\left(z - c(t)\right)\right] dz - \int_0^\delta z\dot{\varepsilon}_{xx}^{0} dz \end{cases}$$

But $\dot{\varepsilon}_{xx}^{0} = -\dfrac{\Delta D}{3}\dfrac{\partial^2 X_2}{\partial z^2} = -\dfrac{\Delta D}{3\tilde{D}}\dfrac{\partial X_2}{\partial t}$

Integrating both wrt to t

$$\begin{cases} 0 = \int_0^\delta \left[\kappa(t)\left(z - c(t)\right)\right] dz + \dfrac{\Delta D}{3\tilde{D}}\int_0^\delta \left[X_2(z,t) - X_2(z,0)\right] dz \quad (= 0) \\ 0 = \int_0^\delta z\left[\kappa(t)\left(z - c(t)\right)\right] dz + \dfrac{\Delta D}{3\tilde{D}}\int_0^\delta z\left[X_2(z,t) - X_2(z,0)\right] dz \end{cases}$$

Get

$$\begin{cases} c(t) = \delta / 2 \\ \dfrac{\delta^3}{12}\kappa(t) = -\dfrac{\Delta D}{3\tilde{D}}\int_0^\delta z\left[X_2(z,t) - X_2(z,0)\right] dz \end{cases}$$

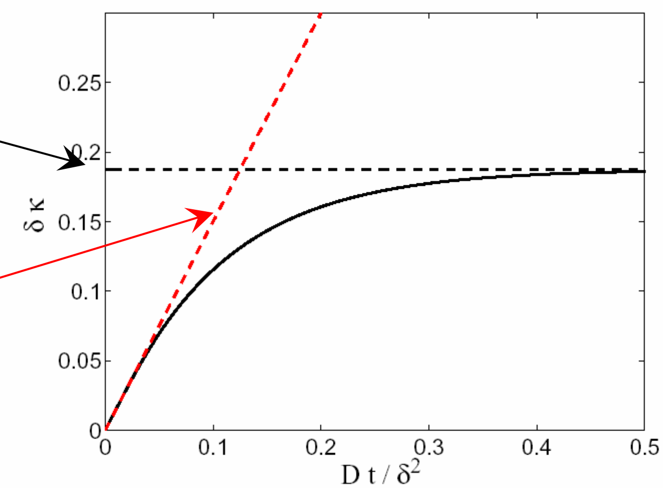
$$X_B(z,t) = \sum_{n=0}^{\infty} a_n \cos\frac{n\pi z}{\delta} \exp(-n^2\pi^2\widetilde{D}t/\delta^2), \qquad a_n = \frac{-2\left[X_B^R - X_B^L\right]}{n\pi} \sin\frac{n\pi z_0}{\delta}$$

$$\delta\,\kappa(t) = -16\frac{D_0}{\widetilde{D}}\frac{\left[X_B^R - X_B^L\right]}{\pi^3} \sum_{n\ \mathrm{odd}} \frac{1}{n^3} \sin\frac{n\pi z_0}{\delta} \left[\exp(-n^2\pi^2\widetilde{D}t/\delta^2) - 1\right]$$

$$\delta\,\kappa(t) \approx 2\frac{z_0(\delta - z_0)}{\delta^2}\frac{D_0}{\widetilde{D}}\left[X_B^R - X_B^L\right]$$

$D_0 = \Delta D$

$$\delta\,\frac{d\kappa}{dt} = 4\left[\frac{\widetilde{D}}{\delta^2}\right]\frac{D_0}{\widetilde{D}}\left[X_B^R - X_B^L\right]$$



- Experiments of Daruka et al. are for short times, $Dt/\delta^2 \sim 0.05$
- Linear form is appropriate
- Using their measured D values, we obtain a slope of 0.03 $m^{-1}s^{-1}$
- They measured a slope of 0.025 $m^{-1}s^{-1}$!

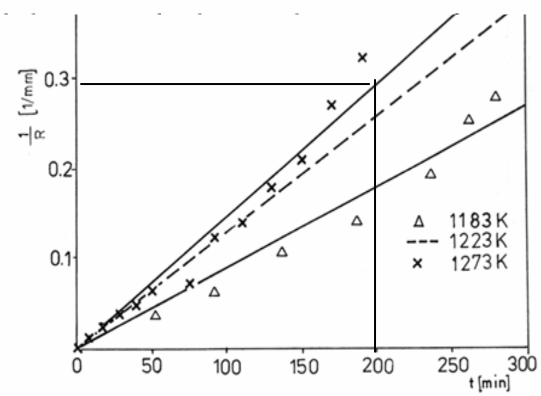


Fig. 12. $1/R$ vs t functions at three different temperatures (the average of the curves shown in Fig. 10 is illustrated by a dashed line).

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, $\delta \gg (Dt)^{1/2}$, marker motion at the sample center is the same as the 1-D for the 1-D Kirkendall effect.
- If $\delta \ll (Dt)^{1/2}$, 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.
- Method can be applied to beams with good result.