Diffusion and Kinetics

Lecture: Diffusion in Metals and Alloys

Nikolai V. Priezjev

<u>Textbook</u>: Phase transformations in metals and alloys (Third Edition), By: Porter, Easterling, and Sherif (CRC Press, 2009).

Diffusion in Metals and Alloys

- ► Diffusion mechanisms
 - (1) Vacancy diffusion (2) Interstitial diffusion (3) Impurities
- ► The mathematics of diffusion
 - (1) Steady-state diffusion (Fick's first law)
 - (2) Nonsteady-State Diffusion (Fick's second law)

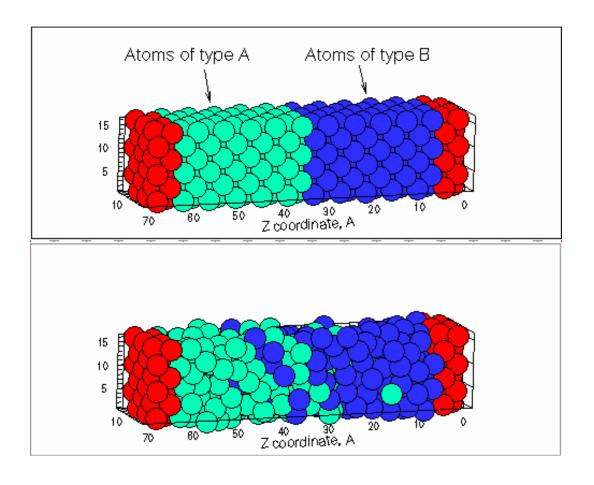
► Factors that influence diffusion

Diffusing species, Host solid, Temperature, Microstructure

Reading: Chapter 2 of Porter, Easterling, Sherif

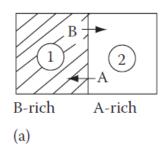
What is diffusion?

Diffusion is material transport by atomic motion.

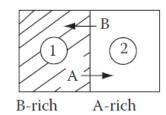


Inhomogeneous materials can become homogeneous by diffusion. For an active diffusion to occur, the temperature should be high enough to overcome energy barriers to atomic motion.

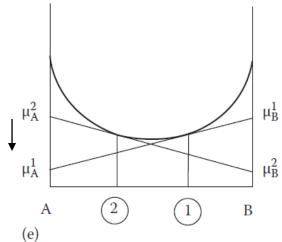
Miscible

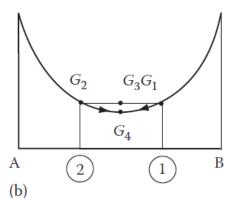


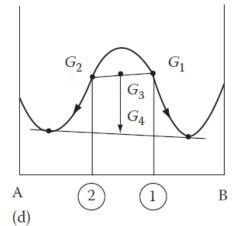
Oil/water

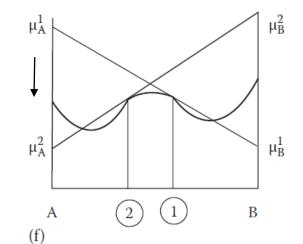


(c)









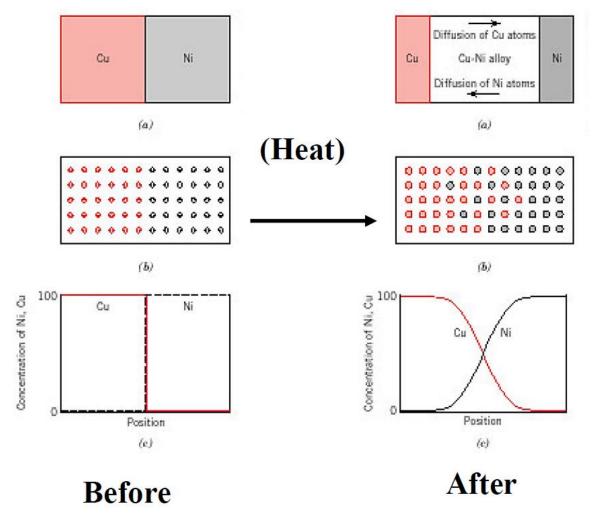
Why diffusion?

Free energy and chemical potential changes during diffusion

Interdiffusion and Self-diffusion

Interdiffusion (or impurity diffusion) occurs in response to a concentration

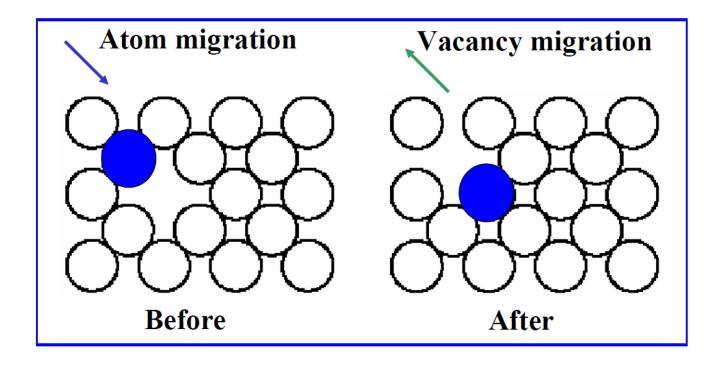
gradient.



Self-diffusion is diffusion in one-component material, when all atoms that exchange positions are of the same type.

Diffusion Mechanisms (I)

Vacancy diffusion mechanism

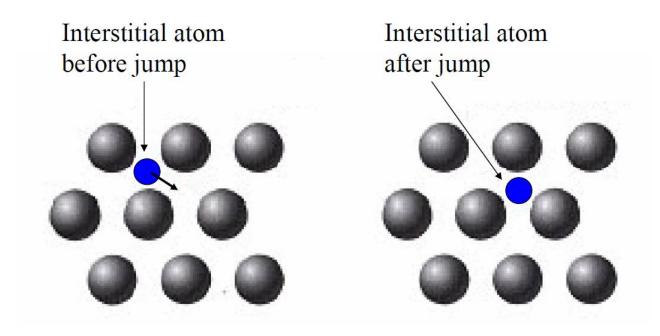


To jump from lattice site to lattice site, atoms need energy to break bonds with neighbors, and to cause the necessary lattice distortions during jump. This energy comes from the thermal energy of atomic vibrations ($E_{av} \sim k_B T$)

The direction of flow of atoms is opposite the vacancy flow direction.

Diffusion Mechanisms (II)

Interstitial diffusion mechanism



Interstitial diffusion is generally faster than vacancy diffusion because bonding of interstitials to the surrounding atoms is normally weaker and there are many more interstitial sites than vacancy sites to jump to.

Requires small impurity atoms (e.g. C, H, O) to fit into interstices in host.

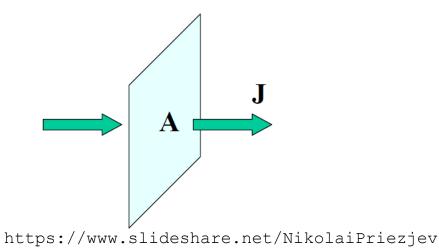
Diffusion Flux

The flux of diffusing atoms, J, is used to quantify how fast diffusion occurs. The flux is defined as either the number of atoms diffusing through unit area per unit time (atoms/m²-second) or the mass of atoms diffusing through unit area per unit time, (kg/m²-second).

For example, for the mass flux we can write

$$J = M / At \cong (1/A) (dM/dt) (Kg m-2 s-1)$$

where M is the mass of atoms diffusing through the area A during time t.



Steady-State Diffusion: Fick's first law

Steady-state diffusion: the diffusion flux does not change with time.

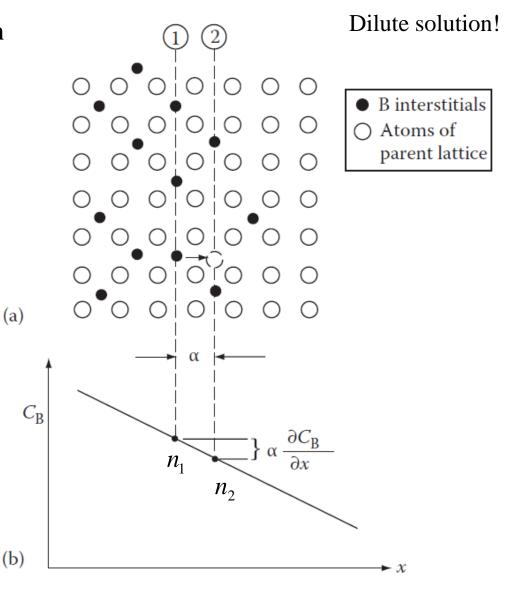
Concentration profile: concentration of atoms/molecules of interest as function of position in the sample.

 Γ_B = number of jumps per second

$$D_B = \frac{1}{6} \Gamma_B \alpha^2 \qquad J_B = -D_B \frac{\partial C_B}{\partial x}$$

$$D_B\left(\frac{\text{m}^2}{\text{s}}\right), J\left(\frac{\text{atoms}}{\text{m}^2\text{s}}\right), \frac{\partial C}{\partial x}\left(\frac{\text{atoms}}{\text{m}^4}\right)$$
 (a)

Fe/C example: γ – Fe at 1000° *C* lattice ~ 0.37nm; $\alpha = 0.37/\sqrt{2} = 0.26$ nm $D_C = 2.5 \cdot 10^{-11}$ m²/s $\Gamma_B = 2 \cdot 10^9$ jumps/s vibr freq ~ $10^{13} : 10^4$ attemps/jump



Random walk of a single atom

After n steps of length α , the net displacement is $\alpha \sqrt{n}$

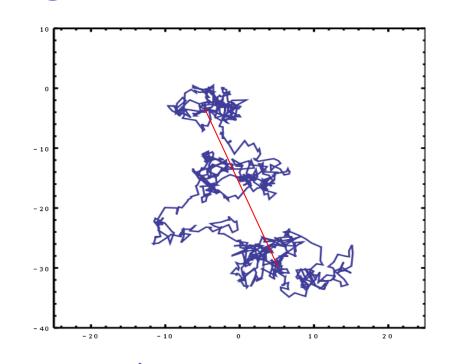
After time t, the average displacement of an atom:

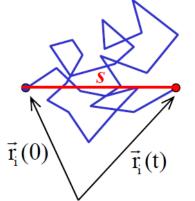
$$r = \alpha \sqrt{\Gamma t}$$

$$D_B = \frac{1}{6} \Gamma_B \alpha^2$$

$$r = 2.4\sqrt{Dt}$$

Fe/C example: γ – Fe at 1000° C Every 1 second C moves ~ 0.5m but net displacement is only ~ 10μ m





Einstein relation

$$<\Delta r(t)^2> = 2d Dt$$

Mean square displacement

$$MSD \equiv s^{2} \equiv \left\langle \Delta \vec{r}(t)^{2} \right\rangle_{i} \equiv \frac{1}{N} \sum_{i=1}^{N} (\vec{r}_{i}(t) - \vec{r}_{i}(0))^{2}$$

Random walk of a single atom

After n steps of length α , the net displacement is $\alpha \sqrt{n}$

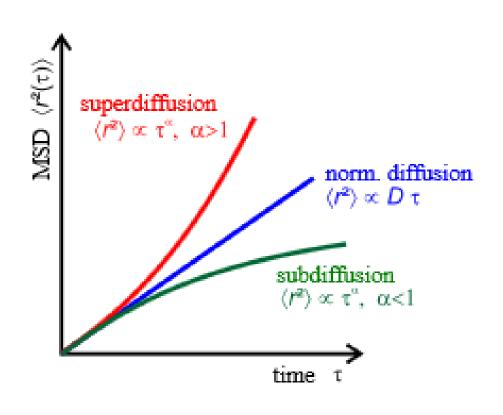
After time t, the average displacement of an atom:

$$r = \alpha \sqrt{\Gamma t}$$

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Anomalous diffusion

Effect of temperature – thermal activation (I)

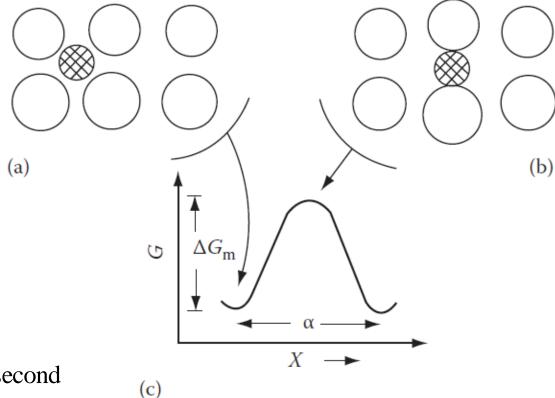
$$D_B = \frac{1}{6} \Gamma_B \alpha^2$$

$$\Gamma_B = z \, v \exp \frac{-\Delta G_{\rm m}}{RT}$$

v = vibrational frequency

z =coordination number

 Γ_B = number of jumps per second



Interstitial atom, (a) in equilibrium position, (b) at the position of maximum lattice distortion, (c) Variation of the free energy of the lattice as a function of the position of interstitial.

 $\Delta G_{\rm m}$ = activation energy

Effect of temperature – thermal activation (II)

$$D_B = \frac{1}{6} \Gamma_B \alpha^2$$

$$\Gamma_B = z \, v \exp \frac{-\Delta G_{\rm m}}{RT}$$

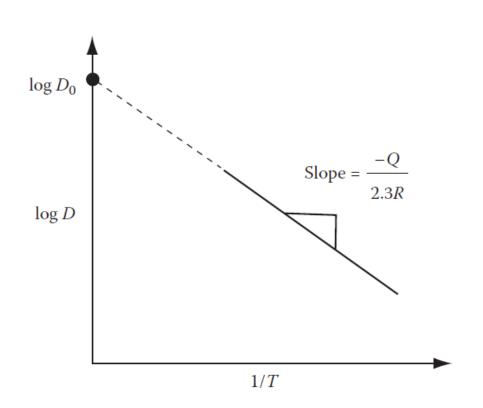
v = vibrational frequency

$$D_B = \left[\frac{1}{6}\alpha^2 zv \exp\frac{\Delta S_m}{R}\right] \exp\frac{-Q}{RT}$$

$$D_B = D_{B0} \exp \frac{-Q}{RT}$$

Q = activation enthalpy

$$\log D_B = \log D_{B0} - \frac{Q}{2.3R} \left(\frac{1}{T}\right)$$



Such plots are called Arrhenius plots.

Effect of temperature – thermal activation (III)

$$D_B = \frac{1}{6} \Gamma_B \alpha^2$$

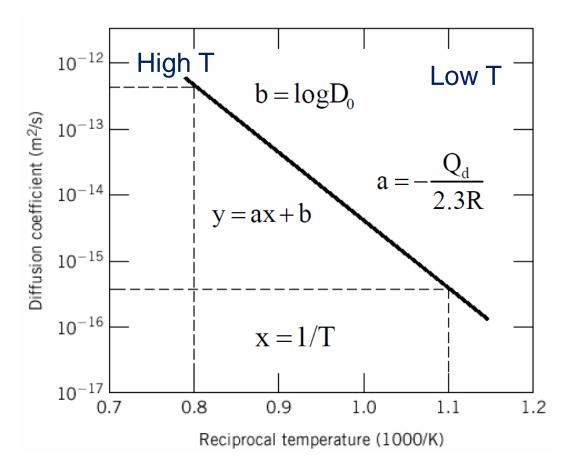
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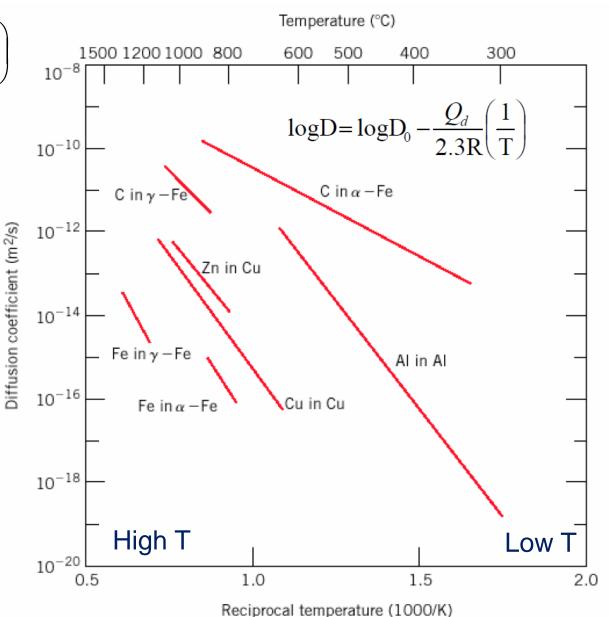
Graph of log D vs. 1/T has slop of $-Q_d/2.3R$, intercept of log D_o

$$Q_{d} = -2.3R \left[\frac{\log D_{1} - \log D_{2}}{1/T_{1} - 1/T_{2}} \right]$$

Effect of temperature – thermal activation (IV)

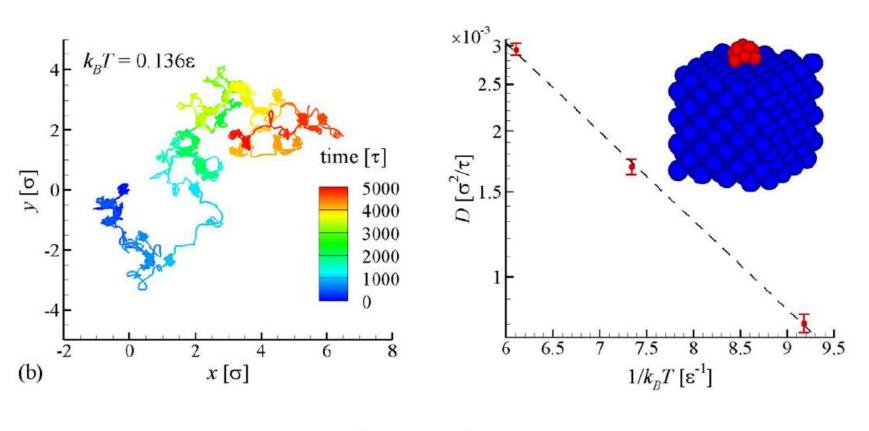
$$\log D_{\rm B} = \log D_{\rm B0} - \frac{Q}{2.3R} \left(\frac{1}{T}\right) \qquad {}_{10^{-8}} \frac{1500\ 1200\ 1000\ 800}{1}$$

Diffusion of interstitials is typically faster as compared to the vacancy diffusion mechanism (self-diffusion or diffusion of substitutional atoms).



Effect of temperature – thermal activation (V)

Diffusion of a cluster of 10 atoms on a substrate.



$$D(T) = D_0 \exp\left(-\frac{E_a}{k_B T}\right)$$
 $E_a = 0.421\varepsilon$

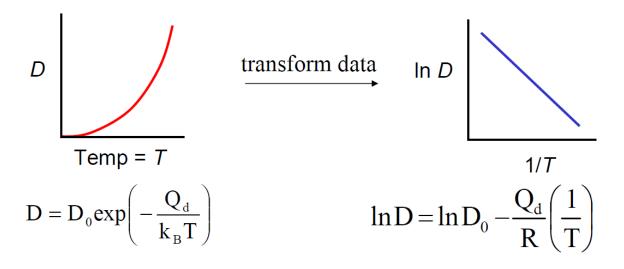
Example: Temperature dependence of D

At 300°C the diffusion coefficient and activation energy for Cu in Si are

$$D(300^{\circ}\text{C}) = 7.8 \text{ x } 10^{-11} \text{ m}^2/\text{s}$$

 $Q_d = 41.5 \text{ kJ/mol}$

What is the diffusion coefficient at 350°C?



$$\ln D_2 = \ln D_0 - \frac{Q_d}{R} \left(\frac{1}{T_2} \right) \quad \text{and} \quad \ln D_1 = \ln D_0 - \frac{Q_d}{R} \left(\frac{1}{T_1} \right)$$

$$\ln D_2 - \ln D_1 = \ln \frac{D_2}{D_1} = -\frac{Q_d}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right)$$

Example: Temperature dependence of D

$$\ln \frac{D_2}{D_1} = -\frac{Q_d}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right)$$

$$D_2 = D_1 \exp \left[-\frac{Q_d}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right) \right]$$

$$T_1 = 273 + 300 = 573 K$$

$$T_2 = 273 + 350 = 623 K$$

$$D_2 = (7.8 \times 10^{-11} \text{ m}^2/\text{s}) \exp \left[\frac{-41,500 \text{ J/mol}}{8.314 \text{ J/mol} - \text{K}} \left(\frac{1}{623 \text{ K}} - \frac{1}{573 \text{ K}} \right) \right]$$

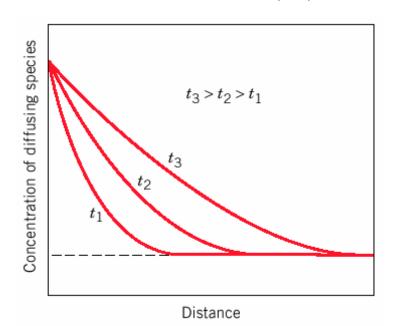
$$D_2 = 15.7 \times 10^{-11} \,\mathrm{m}^2/\mathrm{s}$$

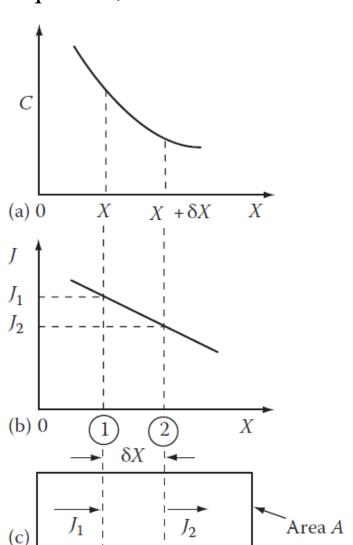
Nonsteady-State Diffusion: Fick's second law

In many real situations the concentration profile and the concentration gradient are changing with time. The changes of the concentration profile can be described in this case by a differential equation, Fick's second law.

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right) = D \frac{\partial^2 C}{\partial x^2}$$

Solution of this equation is concentration profile as function of time, C(x,t):

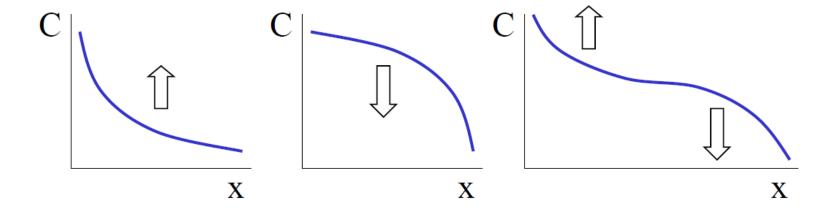




Nonsteady-State Diffusion: Fick's second law

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$

Fick's second law relates the rate of change of composition with time to the curvature of the concentration profile:



Concentration increases with time in those parts of the system where concentration profile has a positive curvature. And decreases where curvature is negative.

Nonsteady-State Diffusion: Homogenization

(Fick's second law)

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$

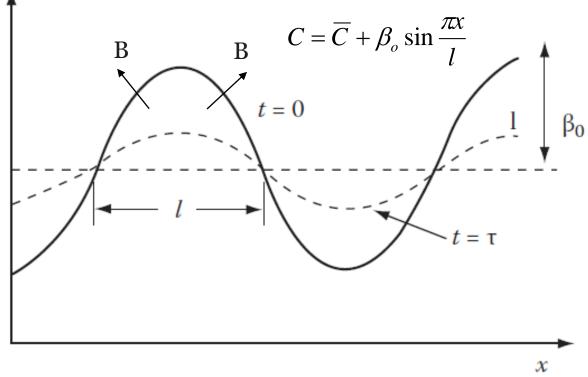
$$C = \overline{C} + \beta_o \sin \frac{\pi x}{l} \exp \frac{-t}{\tau}$$

$$\tau = \frac{l^2}{\pi^2 D_{\scriptscriptstyle R}}$$

Relaxation time

$$\beta = \beta_o \exp \frac{-t}{\tau}$$

Initial concentration profile:



The effect of diffusion on a sinusoidal variation of composition.

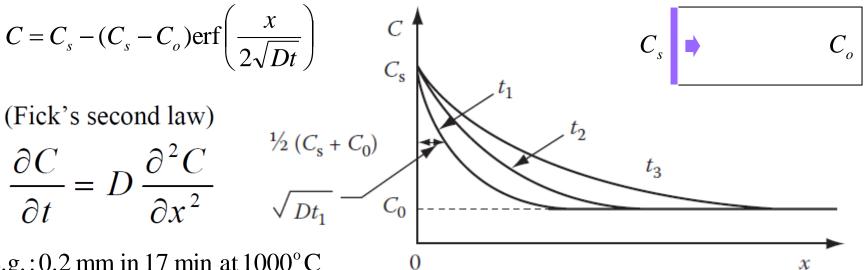
Nonsteady-State Diffusion: Carburization of Steel

Case Hardening: Hardening the surface of a metal by exposing it to impurities that diffuse into the surface region and increase surface hardness.

Common example of case hardening is carburization of steel. Diffusion of carbon atoms (interstitial mechanism) increases concentration of C atoms and makes iron (steel) harder.



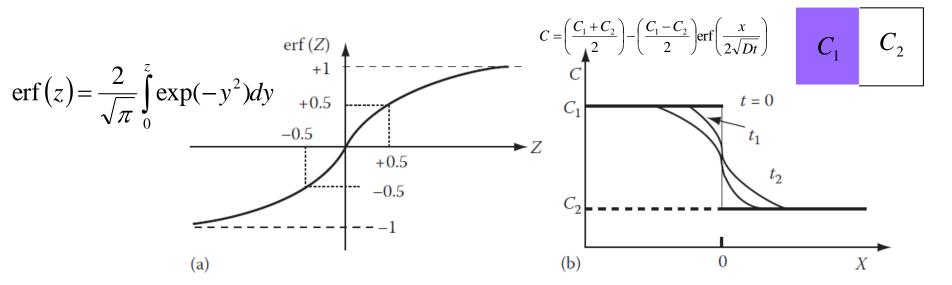
Nonsteady-State Diffusion: Carburization of Steel



E.g.: 0.2 mm in 17 min at 1000° C

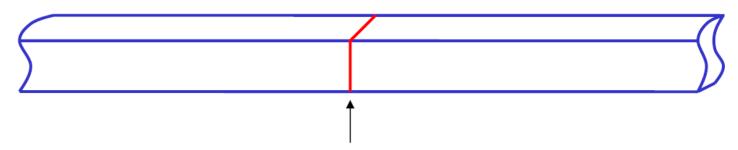
$$D \approx 4 \cdot 10^{-11} \,\mathrm{m}^2 \,\mathrm{/\,s}$$

Concentration profiles at successive times for diffusion into a semiinfinite bar when the surface concentration is maintained constant.



Nonsteady-State Diffusion: Analytical solution for plane source

Let's consider the simple case of solute atoms initially inserted into the middle of an infinite one-dimensional system (plane source).



All solute is in one plane initially

With time the solute atoms will diffuse from the plane. The redistribution of the atoms can be described by the following solution of the diffusion equation:

$$C(x, t) = \frac{A}{\sqrt{t}} \exp\left(-\frac{x^2}{4Dt}\right)$$
 (Fick's second law)
$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$

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Nonsteady-State Diffusion: Analytical solution for plane source

Coefficient A we can find from the condition that the total quantity of solute per unit cross-section area S is

$$M = S \int_{-\infty}^{+\infty} C(x, t) dx = S \int_{-\infty}^{+\infty} \frac{A}{\sqrt{t}} exp\left(-\frac{x^2}{4Dt}\right) dx$$

Using substitution of variable
$$y = \frac{x}{\sqrt{4Dt}}$$
 $dx = \sqrt{4Dt} dy$

$$M = \frac{AS \sqrt{4Dt}}{\sqrt{t}} \int_{-\infty}^{+\infty} \exp(-y^2) dy = AS \sqrt{4D} \times \sqrt{\pi}$$

$$A = \frac{M}{S\sqrt{4\pi D}}$$

(Fick's second law)

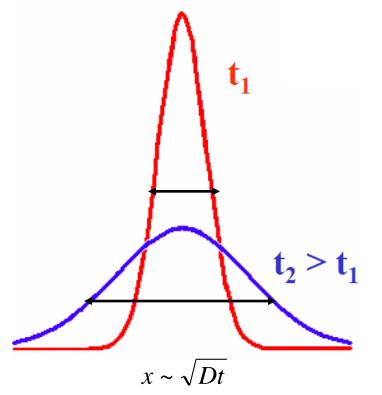
$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$

Nonsteady-State Diffusion: Analytical solution for plane source

$$C(x, t) = \frac{M}{S 2\sqrt{\pi Dt}} \exp\left(-\frac{x^2}{4Dt}\right) \qquad \frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}$$
(Field's second law)

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$

(Fick's second law)

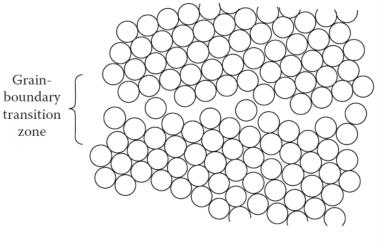


The solution is a Gaussian concentration profile whose width increases with time

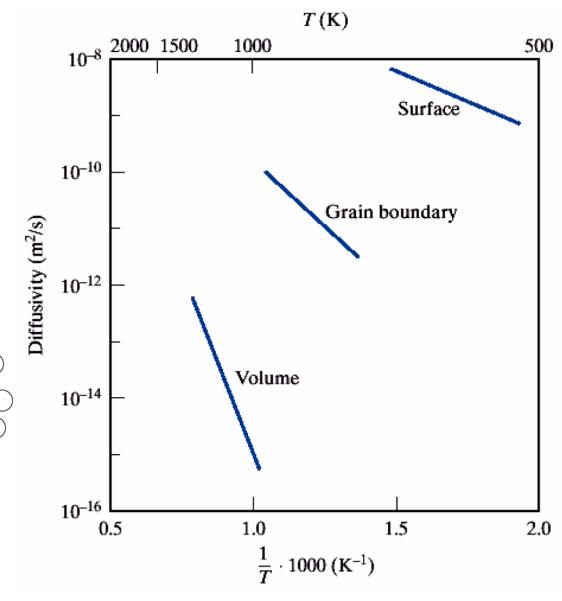


Diffusion: Role of the microstructure (I)

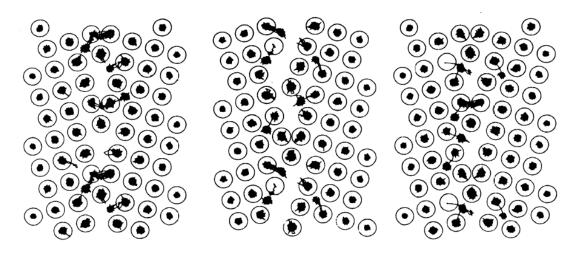
Self-diffusion coefficients for Ag depend on the diffusion path. In general the diffusivity if greater through less restrictive structural regions – grain boundaries, dislocation cores, external surfaces.



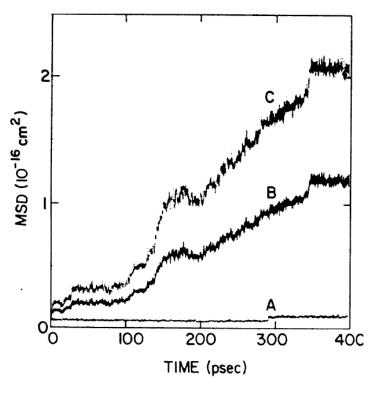
$$D^{G.B.} = D_0^{G.B.} \exp\left(-\frac{\varepsilon_{G.B.}^m}{k_B T}\right)$$



Diffusion: Role of the microstructure (II)



The plots are from the computer simulation by T. Kwok, P. S. Ho, and S. Yip. Initial atomic positions are shown by the circles, trajectories of atoms are shown by lines. We can see the difference between atomic mobility in the bulk crystal and in the grain boundary region.



Mean-square displacement of all atoms in the system (B), atoms in the grain boundary region (C), and bulk region of the system (A).

Mean square displacement: $MSD \equiv s^2 \equiv \left\langle \Delta \vec{r}(t)^2 \right\rangle_i \equiv \frac{1}{N} \sum_{i=1}^N (\vec{r}_i(t) - \vec{r}_i(0))^2$

2.3.1 Substitutional Diffusion: Self-Diffusion

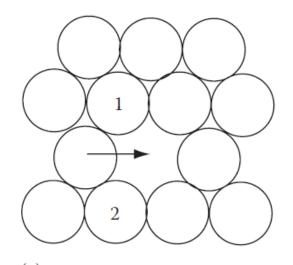
$$\Gamma = z v X_{v} \exp \frac{-\Delta G_{m}}{RT}$$

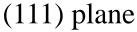
v = vibrational frequency

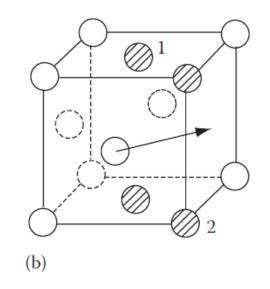
z = 12 in fcc metals

 X_{v}^{e} = vacancy concentration

$$X_{v}^{e} = \exp \frac{\Delta S_{v}}{R} \cdot \exp \frac{-\Delta H_{v}}{RT}$$







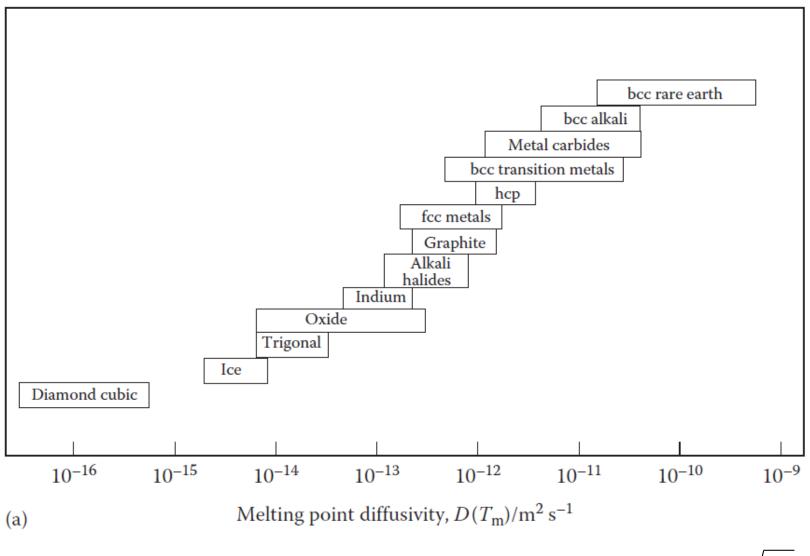
$$D = \frac{1}{6} \Gamma \alpha^{2} \qquad D = \frac{1}{6} \alpha^{2} zv \exp \frac{\Delta S_{m} + \Delta S_{v}}{R} \cdot \exp \left(-\frac{\Delta H_{m} + \Delta H_{v}}{RT}\right)$$

Note:
$$D_v = D / X_v^e$$

$$D = D_0 \exp \frac{-Q_{SD}}{RT} \qquad Q_{SD} = \Delta H_m + \Delta H_v$$

$$Q_{SD} = \Delta H_m + \Delta H_v$$

2.3.1 Substitutional Diffusion: Self-Diffusion



 $r = 2.4\sqrt{Dt}$

2.3.1 Substitutional Diffusion: Self-Diffusion

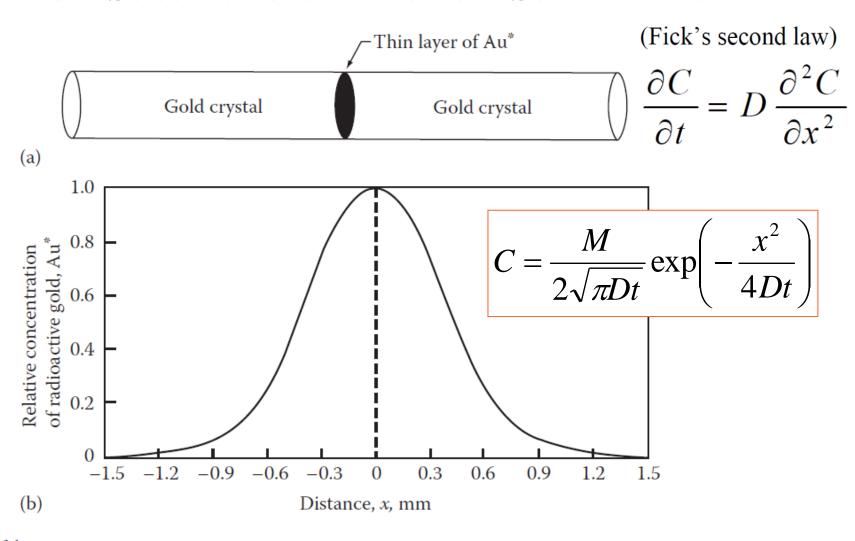
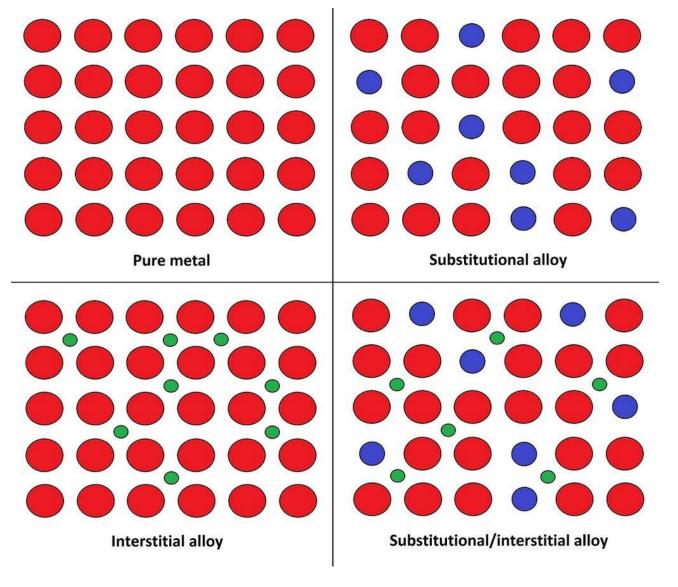


Figure 2.14 Illustration of the principle of tracer diffusion and of the planar source method for determining the self-diffusion coefficient of gold. (a) Initial diffusion couple with planar source of radioactive gold Au*. (b) Distribution of Au* after diffusion for 100 h at 920°C. (After A.G. Guy, *Introduction to Materials Science*, McGraw-Hill, New York, 1971.)

Substitutional and interstitial alloys



Different atomic mechanisms of alloy formation, showing pure metal, substitutional, interstitial, and a combination of the two.

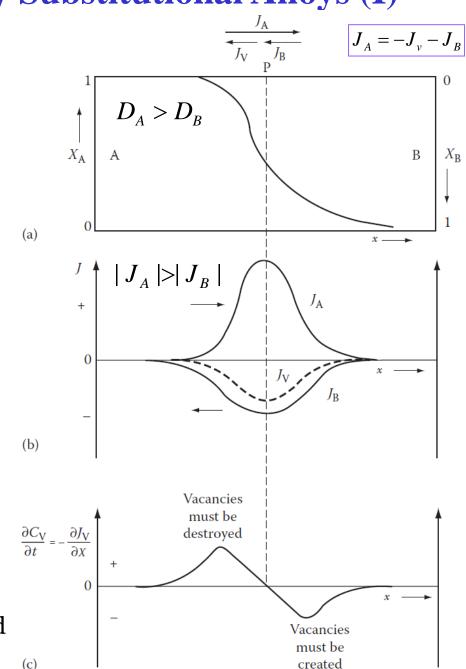
2.3.3 Diffusion in Binary Substitutional Alloys (I)

Diffusion relative to the lattice:

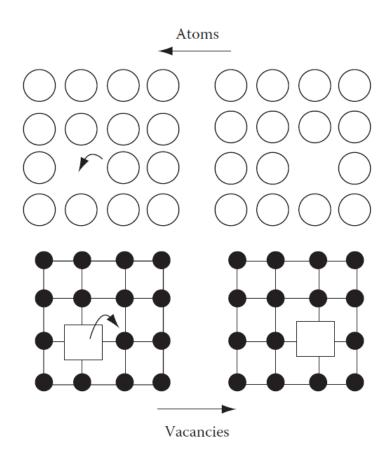
$$\boxed{C_0 = C_A + C_B} \qquad \boxed{\frac{\partial C_A}{\partial x} = -\frac{\partial C_B}{\partial x}}$$

Rewrite:

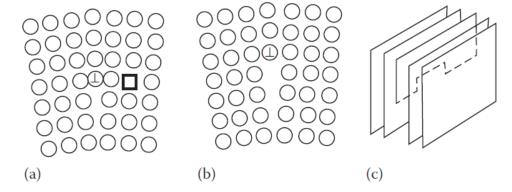
Interdiffusion and vacancy flow. (a) Composition profile after interdiffusion of A and B. (b) The corresponding fluxes of atoms and vacancies as a function of position x. (c) The rate at which the vacancy concentration created or destroyed by dislocation climb.



2.3.3 Diffusion in Binary Substitutional Alloys (II)



The jumping of atoms in one direction can be considered as the jumping of vacancies in the other direction.



(a) before, (b) after: a vacancy is absorbed at a jog on an edge dislocation (positive climb), (b) before, (a) after: a vacancy is created by negative climb of an edge dislocation, (c) Perspective drawing of a jogged edge dislocation.

2.3.3 Diffusion in Binary Substitutional Alloys (III)

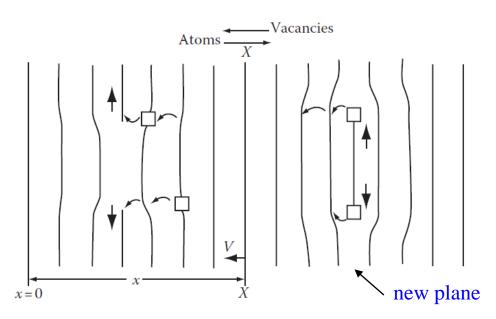
Vacancy net flux:

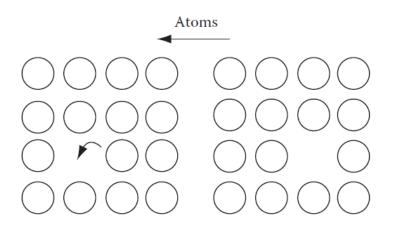
$$\boldsymbol{J}_{\boldsymbol{A}} = -\boldsymbol{J}_{\boldsymbol{v}} - \boldsymbol{J}_{\boldsymbol{B}}$$

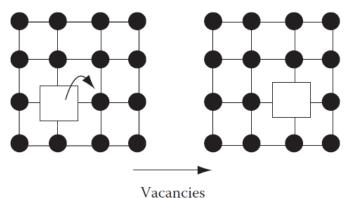
$$J_{v} = -J_{A} - J_{B}$$

$$\boxed{J_{v} = -J_{A} - J_{B}} \qquad J_{v} = (D_{A} - D_{B}) \frac{\partial C_{A}}{\partial x}$$

To keep C_v constant, vacancies should be created on B-rich side and destroyed on A-rich side.







The jumping of atoms in one direction can be considered as the jumping of vacancies in the other direction.

$$\frac{\partial C_{v}}{\partial t} = -\frac{\partial J_{v}}{\partial x}$$

A flux of vacancies causes the atomic planes to move through the specimen.

2.3.3 Diffusion in Binary Substitutional Alloys (IV)

Velocity of movement of lattice planes v:

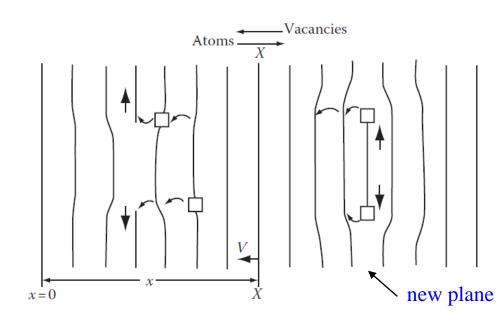
$$J_{v} A \cdot \delta t = A \cdot v \cdot \delta t \cdot C_{0}$$

atoms is removed by vacancies crossing plane A # atoms in the volume, which is swept by plane A during time dt

$$J_{v} = \mathbf{v} \cdot C_{0}$$

$$J_{v} = (D_{A} - D_{B}) \frac{\partial C_{A}}{\partial x}$$

$$X_A = C_A / C_0$$



A flux of vacancies causes the atomic planes to move through the specimen.

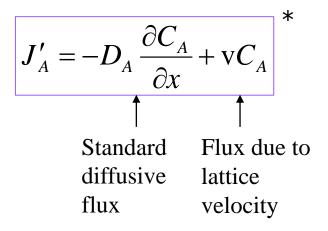
$$\mathbf{v} = (D_A - D_B) \frac{\partial \mathbf{X}_A}{\partial x}$$

But is does not tell us how fast A and B actually mix.

2.3.3 Diffusion in Binary Substitutional Alloys (V)

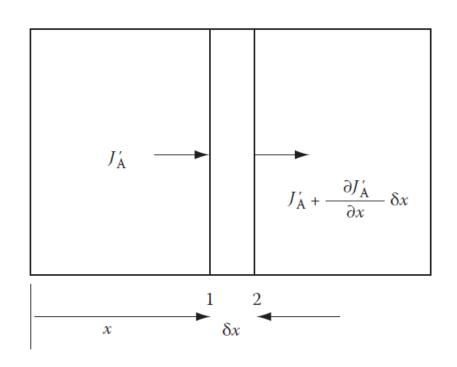
$$\frac{\partial C_A}{\partial t} = -\frac{\partial J_A'}{\partial x}$$

The total flux of atoms A:



$$\mathbf{v} = (D_A - D_B) \frac{\partial \mathbf{X}_A}{\partial x}$$
 $\mathbf{X}_A = C_A / C_0$

$$J_A' = -(X_B D_A + X_A D_B) \frac{\partial C_A}{\partial x}$$



<u>Interdiffusion coefficient:</u>

$$\widetilde{D} = X_B D_A + X_A D_B \qquad J_A' = -J_B'$$

$$J_A' = -\widetilde{D} \frac{\partial C_A}{\partial x} \qquad J_B' = -\widetilde{D} \frac{\partial C_B}{\partial x}$$

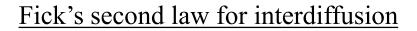
Fick's first law for interdiffusion

2.3.3 Diffusion in Binary Substitutional Alloys (VI)

$$\frac{\partial C_A}{\partial t} = -\frac{\partial J_A'}{\partial x} +$$

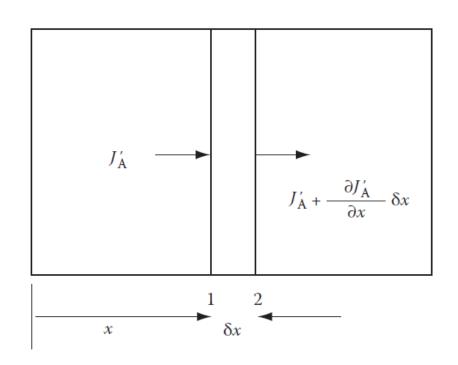
$$J_A' = -\widetilde{D} \frac{\partial C_A}{\partial x}$$

$$\frac{\partial C_A}{\partial t} = \frac{\partial}{\partial x} \left(\widetilde{D} \frac{\partial C_A}{\partial x} \right)$$



$$\widetilde{D} = X_B D_A + X_A D_B$$

Darken's equations

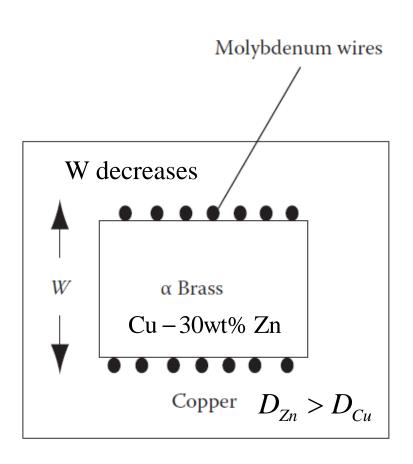


$$\widetilde{D} = \widetilde{D}_0 \exp \frac{-Q}{RT}$$

$$D_A = D_{A0} \exp \frac{-Q_A}{RT}$$

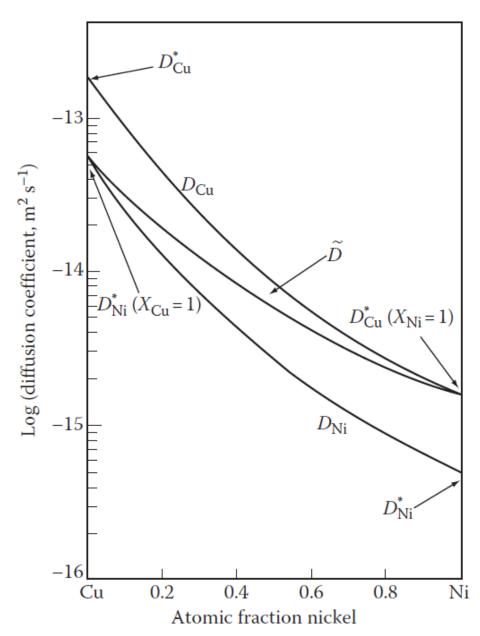
$$D_B = D_{B0} \exp \frac{-Q_B}{RT}$$

2.3.3 Diffusion in Binary Substitutional Alloys (VII)

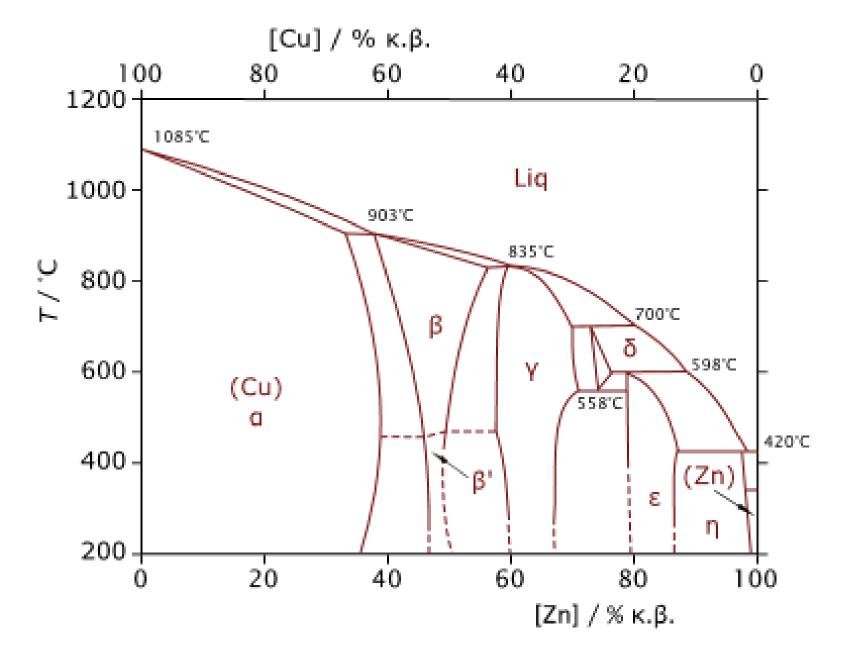


An experimental arrangement to show the Kirkendall effect. 1947. inert wires.

$$\widetilde{D} = X_B D_A + X_A D_B$$



diffusion coefficients in the Cu-Ni system at 1000°C



https://www.slideshare.net/NikolaiPriezjev

2.4 Atomic Mobility

Drift velocity is related to the diffusive flux:

$$J_B = v_B C_B$$

$$J\left(\frac{\text{atoms}}{\text{m}^2\text{s}}\right) = v\left(\frac{\text{m}}{\text{s}}\right) \cdot C\left(\frac{\text{atoms}}{\text{m}^3}\right)$$

Reasonable to guess that:

 $v_B = -M_B \frac{\partial \mu_B}{\partial x}$ Atoms Some 'Chemical' force per gradient Mobility atom

Combine both equations:

$$J_{B} = -M_{B}C_{B} \frac{\partial \mu_{B}}{\partial x}$$

$$J_{B} = -D_{B} \frac{\partial C_{B}}{\partial x}$$

2.7 High Diffusivity Paths

Found experimentally that:

$$D_l = D_{l0} \exp \frac{-Q_l}{RT}$$

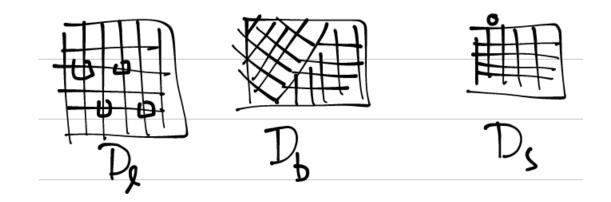
$$D_b = D_{b0} \exp \frac{-Q_b}{RT}$$

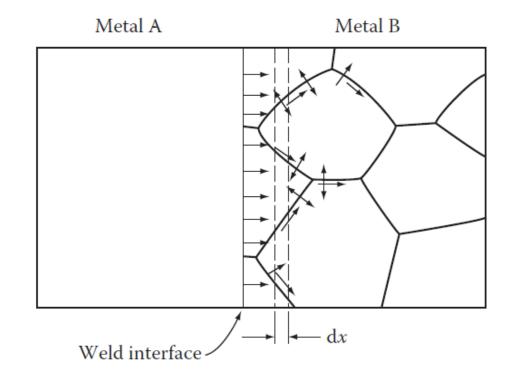
$$D_S = D_{S0} \exp \frac{-Q_S}{RT}$$

In general

$$D_{S} > D_{b} > D_{l}$$

But the size of the highway matters!





The effect of grain boundary diffusion combined with volume diffusion.

2.7 High Diffusivity Paths

$$J_{l} = -D_{l} \frac{\partial C}{\partial x}$$

$$J_{l} = -D_{l} \frac{\partial C}{\partial x} \qquad J_{b} = -D_{b} \frac{\partial C}{\partial x}$$

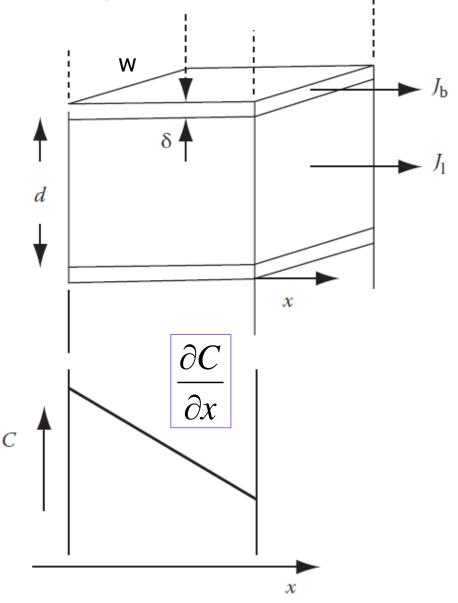
The total flux:

$$J = (J_b \delta \mathbf{w} + J_l d\mathbf{w}) / d\mathbf{w} =$$

$$= -\left(\frac{D_b \delta + D_l d}{d}\right) \cdot \frac{\partial C}{\partial x}$$

The apparent diffusion coefficient:

$$D_{app} = D_l + D_b \delta / d$$



Combined lattice and boundary fluxes during steadystate diffusion through a thin slab of material.

2.7 High Diffusivity Paths

The apparent diffusion coefficient:

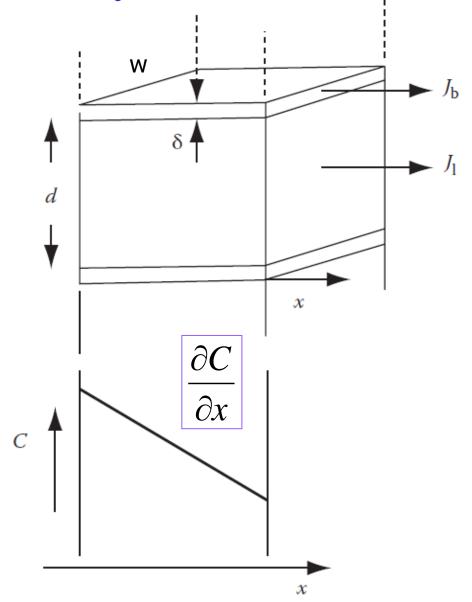
$$\frac{D_{app}}{D_l} = 1 + \frac{D_b \delta}{D_l d}$$

Grain boundary diffusion is important:

$$D_b \delta >> D_l d$$

$$D_{app} = D_l + D_b \delta / d$$

 $\delta \sim 0.5 \, \mathrm{nm}$ Grain boundary $d \sim 1 - 1000 \, \mu \mathrm{m}$ Grain size



Combined lattice and boundary fluxes during steadystate diffusion through a thin slab of material.

2.7 High Diffusivity Paths: Temperature effects

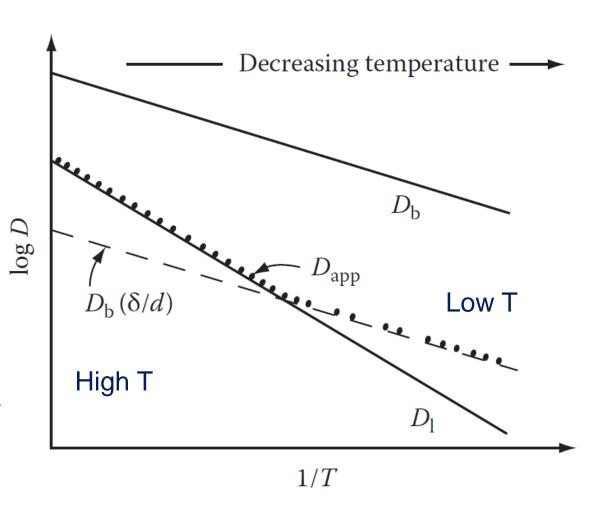
$$D_l = D_{l0} \exp \frac{-Q_l}{RT}$$

$$D_b = D_{b0} \exp \frac{-Q_b}{RT}$$

Note that:

$$D_b > D_l$$
 at all T

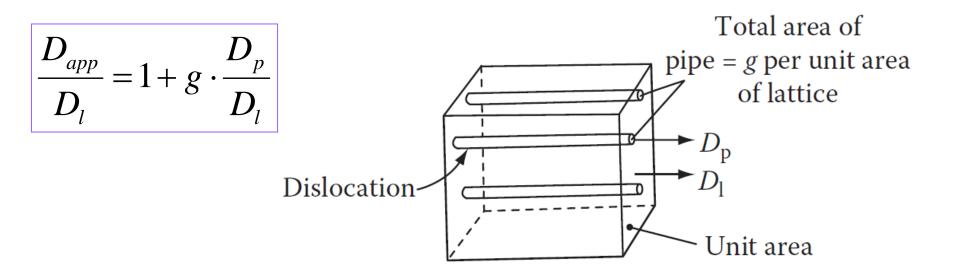
 $Q_b \approx 0.5 Q_l$ in fcc metals



 $D_{app} = D_l + D_b \delta / d$

Diffusion in a polycrystalline metal

2.7 High Diffusivity Paths: Along Dislocations



Dislocations act as a high conductivity path through the lattice.

In a well annealed material:

 $g \approx 10^{-7}$

10⁵ dislocations per mm⁻²

 10^{13} atoms mm⁻²

$$High T: g \cdot \frac{D_p}{D_l} = small$$

Low T: only dislocations

