Dynamics of Negative Diffusivity and Uphill Diffusion in Ternary and Single systems

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Abstract. Negative diffusivity and uphill diffusion, that contrast with Fick's laws, however which can occur in multicomponent and single systems. In this paper, authors present studied results about the dynamics of the uphill diffusion in ternary and single system. Results have shown that: i) in ternary systems, the dynamics of negative diffusivity is high concentration effect and the combination of coupled diffusion effect with high concentration effect is the dynamics of uphill diffusion; ii) in single systems, the difference of temperatures in two areas that is the dynamics of negative diffusivity and uphill diffusion.

Keywords: Dynamics of uphill diffusion; dynamics of negative diffusivity.

1 Introduction

In uphill diffusion, the diffusion flux goes up to the higher concentration area, which often occurs in multicomponent systems [1-6]. However, uphill diffusion can occur in single component system [7, 8]. In the following, authors apply the irreversible thermodynamics theory to treat dynamics of negative diffusivity and uphill diffusion in ternary system of boron and point defects (interstitial and vacancy) in silicon and single component system.

2 Dynamics of negative diffusivity and uphill diffusion in ternary system of boron and point defect in silicon

Based on irreversible thermodynamics [9], the diffusion of boron (B) and point defects (interstitial (I) and vacancy (V)) in silicon can be described. When concentration gradient of interstitial is very small and almost negligible, the diffusion flux of boron (J_B), interstitial (J_I) and vacancy (J_V) are found out [10]:

$$J_B = -D_{BB}\frac{\partial C_B}{\partial x} \tag{1}$$

$$J_I = -D_{IB}\frac{\partial C_B}{\partial x} \tag{2}$$

$$J_V = -J_B + J_I \tag{3}$$

in which, D_{BB} is intrinsic diffusivity of B and D_{IB} is mutual diffusivity of I, which are determined by:

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$$D_{BB} = \frac{1}{2}(2D_B + D_V + \frac{D_B C_B - D_I C_I}{D_V}) \tag{4}$$

$$D_{IB} = \frac{1}{2}(D_V - D_B + \frac{D_V C_V - D_I C_I}{C_B} + \frac{D_I C_I - D_B C_B}{C_V}) \tag{5}$$

Based on diffusion flux equations (1, 2, 3), we can find out the concentration profiles of B, I and V (Tab.1 and Fig.1). In which, x is distance from silicon surface.

Tab 1. Contribution of C_B, C_I, C_V, and sign diffusivity of D_{IB} in Si for 10 diffusion minutes at 1000°C.

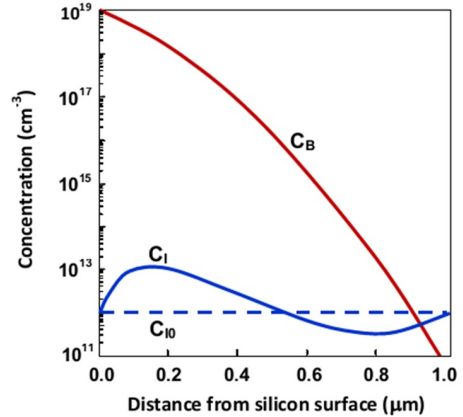| x (cm)	C_B (cm^{-3})	C_I (cm^{-3})	C_V (cm^{-3})	D_{IB} (cm^{2}s^{-1})
5.3×10^{-6}	6.5×10^{18}	2.0×10^{13}	5.8×10^{13}	-6.5×10^{-10}
1.1×10^{-5}	4.4×10^{18}	2.6×10^{13}	4.5×10^{13}	-5.2×10^{-10}
2.1×10^{-5}	1.6×10^{18}	2.4×10^{13}	4.8×10^{13}	-1.2×10^{-10}
3.2×10^{-5}	4.0×10^{17}	1.7×10^{13}	6.9×10^{13}	1.5×10^{-11}
4.2×10^{-5}	7.2×10^{16}	1.0×10^{13}	1.1×10^{14}	2.3×10^{-11}
5.3×10^{-5}	9.6×10^{15}	5.6×10^{12}	2.0×10^{14}	1.7×10^{-11}
6.4×10^{-5}	9.8×10^{14}	2.9×10^{12}	4.0×10^{14}	1.9×10^{-11}
7.4×10^{-5}	7.8×10^{13}	1.4×10^{12}	8.2×10^{14}	1.5×10^{-10}
1.0×10^{-4}	5.5×10^{10}	4.5×10^{11}	2.5×10^{15}	5.8×10^{-10}

Figure 1. Concentration profile of B and I in Si.

Eq.5 shows that: diffusion flux of interstitial does not depend on its concentration gradient, but depends on concentration gradient of boron (that is coupled diffusion effect). Calculated result of mutual diffusivity D_{IB} is presented in Tab.1, which shows that when boron concentration is high ($C_B = 10^{18} \div 10^{19}$), mutual diffusivity of interstitial becomes negative (that is high concentration effect). Thus, the combination of the coupled diffusion effect with high the concentration effect is the dynamics of uphill diffusion of interstitial in silicon.

3 Dynamics of negative diffusivity and uphill diffusion in single system

Based on Onsager's equation the general diffusion flux can be determined [11]:

$$J = -L\frac{\partial \mu}{\partial x} \tag{6}$$

in which, μ is chemical potential, L is phenomenological coefficient. The chemical potential is the function of concentration C and temperature T [12]:

$$\mu = \mu_o + KT \ln C \tag{7}$$

We propose a general diffusion model (Fig.2): the concentration C_1 in area 1 lower than concentration C_2 in area 2 but temperature T_1 in area 1 geater than that T_2 in area 2:

$$T_1 = \alpha.T_2 = \alpha.T \tag{8}$$

Where coefficient α is greater than 1, and the temperature is directly proportional to square thermal velocity, so we have:

$$u_1^2 = \alpha u_2^2 = \alpha u^2 \tag{9}$$

Finally, the diffusion flux (6) becomes:

$$J = -L\frac{mu^2}{3}\frac{lnC_2 - \alpha.lnC_1}{\Delta C}\cdot\frac{\Delta C}{\Delta x} = -D\frac{\Delta C}{\Delta x} \tag{10}$$

where D is diffusivity:

$$D = L\frac{mu^2}{3}\frac{lnC_2 - \alpha.lnC_1}{\Delta C} \tag{11}$$

Based on value and sign of diffusivity D (Tab.2), diffusion of single component can be classified in to four types, which are show in Fig.3, in which $\beta = lnC_2/lnC_1$.
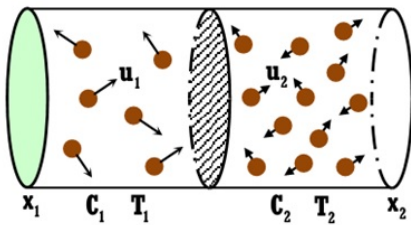

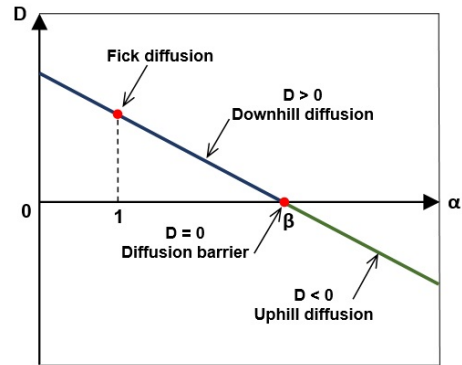**Figure 2.** General diffusion model for single component.

Figure 3. Classified diffusion in single system.

When the temperature (T_1) in low concentration area is β times more greater than the temperature (T_2) high concentration area, the diffusivity becomes negative and diffusing process is uphill diffusion. Based random walk theory [13, 14], the general diffusion model for single component were simulated in the two dimensions on two regions 1 and 2 of diffused space (Fig.4a). The program of simulation is written by the Processing language. Simulated results are presented by the motion pictures on the monitor of the computer. Fig.4a shows the positions of particles in regions 1 and 2 at the initial time t = 0, the number of particle in two parts are the same and equal to 100 ($N_1 = N_2 = 100$). Fig.4b presents the pictures of the positions of particles after the 10 minutes of diffusion, the number of particle in regions 1 and 2 are 74 and 126 ($N_1 = 74$ and $N_2 = 126$). Fig.4c shows the number of particles after the 20 minutes of diffusion are 67 and 143 ($N_1 = 67$ and $N_2 = 143$). Results show: i) At the initial time ($t = 0$): although gradient of concentration is equal to zero, the diffusion process occurs with a diffusion flux goes from region 1 (in which, the velocity of particles are high) to region 2 (in which, the velocity of particles are low). This is the osmotic diffusion; ii) After initial time: the concentration in part 2 is greater than that in part 1, but there is a diffusion flux that goes from region 1 (low concentration region) to region 2 (high concentration region). This is the uphill diffusion.

Thus, simulated result showed that the temperature T_1 in low concentration area is greater than that T_2 in high concentration area is the cause of negative diffusivity and uphill diffusion in single systems.
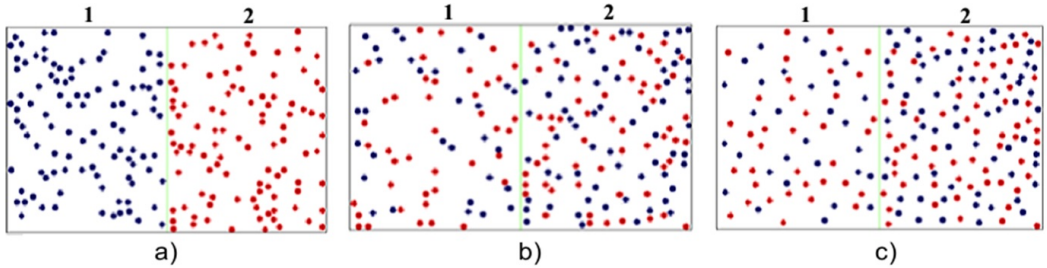
Figure 4. Result of the simulation of diffusion in single component at the simulated time of
t:
a) t = 0, b) t = 10 minutes and c) t = 20 minutes

4 Conclusions

Negative diffusivity and uphill diffusion can occur in ternary and single component systems:
i) In ternary systems, the dynamics of negative diffusivity is high concentration effect and the
combination of coupled diffusion effect with high concentration effect is the uphill diffusion;
ii) In single systems, the difference of temperatures (or thermal velocities) in two areas that
is the dynamics of negative diffusivity and uphill diffusion.

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