

**STUDY ON SELF-DIFFUSION IN ALLOY AuCuSi
WITH FCC STRUCTURE: DEPENDENCE ON TEMPERATURE, PRESSURE,
CONCENTRATION OF SUBSTITUTIONAL ATOMS, CONCENTRATION
OF INTERSTITIAL ATOMS AND STRAIN**

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Abstract. This article presents a self-diffusion theory, including analytical expressions for activation energy, pre-exponential factor, and self-diffusion coefficient, dependent on temperature, pressure, the concentration of substitutional atoms, the concentration of interstitial atoms, and strain for substitutional and interstitial ternary alloys with the FCC structure, based on the statistical moment method (SMM). The theoretical results are applied to numerical calculations for the AuCuSi alloy. The SMM numerical results for AuCuSi are compared with those for AuSi, AuCu, and Au. The variation of self-diffusion quantities with temperature and strain in AuCuSi follows the same patterns as in AuCu, AuSi, and Au. The variation with the interstitial atom concentration in AuCuSi follows the same pattern as in AuSi, while the variation with the substitutional atom concentration follows the same pattern as in AuCu. The SMM numerical results for Au's self-diffusion quantities agree well with experimental data and other calculation results. Other SMM numerical results for self-diffusion quantities are novel and predict future experimental outcomes.

Keywords: AuCuSi, self-diffusion, strain, binary alloy, ternary alloy, interstitial atom, substitutional atom, activation energy, pre-exponential factor, self-diffusion coefficient, SMM.

1. Introduction

Diffusion and the influence of stress on the diffusion of metals and semiconductors have attracted the attention of many researchers [1]-[10]. The theory of self-diffusion in a crystal via the vacancy mechanism has been investigated using a detailed dynamical model. It has been shown that the parameters determining the diffusion coefficient can be defined in terms of the crystal's normal coordinates [1]. A calculation that simplifies

the pair correlation functions has also been presented [2]. Activation volume and energy have been calculated from diffusion measurements at high temperatures and pressures [3]. A thermodynamic formalism has been developed to illuminate the predominant point defect mechanism of self- and impurity diffusion in silicon, providing a rigorous basis for point defect-based interpretation of diffusion experiments in biaxially strained epitaxial layers in the Si–Ge system [4]. The thermodynamics of diffusion under hydrostatic and non-hydrostatic stress has been presented for single crystals free of extended defects [5]. Enhanced Sb diffusion in biaxially compressed Si_{1-x}Ge_x layers (x = 0.1 and 0.2) has been investigated [6]. The measurement of the two-dimensional island density after submonolayer deposition has been used to determine the effect of material surface reconstruction and strain on surface diffusion [7]. Molecular dynamics simulations of adatom diffusion on strained (111) Lennard-Jones surfaces have been presented [8]. The discovery of bulk metallic glasses (BMGs) has sparked considerable interest in understanding their mechanical behavior [9]. The diffusion coefficients of hydrogen in body-centered cubic (bcc) iron have been evaluated via density functional theory and small-polaron theory calculations [10]. The statistical moment method (SMM) has been applied to study diffusion characteristics in metals and alloys [11]-[17]. Studies on self-diffusion in metals and binary interstitial alloys with BCC and FCC structures using the statistical moment method (SMM) have been reviewed in previous works [16], [17].

In this work, for the first time, we present analytical expressions for self-diffusion quantities such as activation energy, pre-exponential factor, and self-diffusion coefficient, depending on temperature, pressure, the concentration of substitutional atoms, the concentration of interstitial atoms, and strain for the substitutional and interstitial ternary alloy with the FCC structure, based on the SMM. The theoretical results are applied to numerical calculations for the AuCuSi alloy.

2. Content

Using the Mie-Lennard-Jones potential and the following approximations to describe the interactions Au-Au, Cu-Cu, Si-Si, Au-Cu, Au-Si, and Cu-Si

$$\varphi(r) = \frac{C}{n-m} \left[m \left(\frac{r_0}{r} \right)^n - n \left(\frac{r_0}{r} \right)^m \right], \quad (1)$$

$$\varphi_{\text{Au-Cu}} \approx \frac{1}{2} (\varphi_{\text{Au-Au}} + \varphi_{\text{Cu-Cu}}), \varphi_{\text{Au-Si}} \approx \frac{1}{2} (\varphi_{\text{Au-Au}} + \varphi_{\text{Si-Si}}), \varphi_{\text{Cu-Si}} \approx 0. \quad (2)$$

The potential parameters D , r_0 , m , and n are given in Table 1.

Table 1. The parameters of potential Mie-Lennard-Jones for interactions Au-Au, Cu-Cu and Si-Si [18]

Interaction	m	n	C (10^{-16} erg)	r_0 (10^{-10} m)
Au-Au	5.5	10.5	4683	2.875
Cu-Cu	5.5	11.0	6470	2.549
Si-Si	6.0	12.0	45128	2.295

We can calculate the activation energy Q , the pre-exponential factor D_0 , and the self-diffusion coefficient D of defective metal Au (the main metal in ternary interstitial and substitutional alloy AuCuSi) at zero pressure and temperature T according to the following formulas:

$$Q(0,T) = -u_0(0,T) + \Delta\psi^R(0,T) - \Delta\psi^{R(1)}(0,T), \quad (3)$$

$$\Delta\psi^R = (B^R - 1)\psi^R, \Delta\psi^{R(1)} = (B^{R(1)} - 1)\psi^{R(1)}, \quad B^R \approx 1 + \frac{u_0^R}{\psi^R}, \quad B^{R(1)} \approx 1 + \frac{u_0^R - \Delta\psi^R}{2\psi^{R(1)}}, \quad (4)$$

$$\psi^R = \left[1 - n_v n_1 + n_v (B^R - 1)\right] \psi + n_v n_a \psi^{(1)}, \quad (5)$$

$$\begin{aligned} \psi = Nu_0 + 3N\theta[x + \ln(1 - e^{-2x})] + \frac{3N\theta^2}{k^2} \left[\gamma_2 Y^2 - \frac{2\gamma_1}{3} \left(1 + \frac{Y}{2}\right) \right] + \\ + \frac{6N\theta^3}{k^4} \left[\frac{4}{3} \gamma_2^2 \left(1 + \frac{Y}{2}\right) Y - 2(\gamma_1^2 + 2\gamma_1\gamma_2) \left(1 + \frac{Y}{2}\right) (1+Y) \right], \end{aligned} \quad (6)$$

$$n_v = \exp\left(\frac{u_0^R}{4k_{Bo}T}\right), \quad (7)$$

$$D_0(0,T) = \frac{n_1 f \omega r_1^2}{2\pi} \exp\left(\frac{s_v^f}{k_{Bo}}\right), \quad (8)$$

$$s_v^f = \frac{\frac{\partial u_0^R}{\partial T} - (B-1) \frac{\partial \psi^R}{\partial T} - \frac{g_v^f + \frac{\partial u_0^R}{\partial T} - (B-1) \frac{\partial \psi^R}{\partial T}}{k_{Bo} T^2} g_v^f + k_{Bo} \ln(N+1)}{n_1 + n_2}, \quad (9)$$

$$g_v^f = -\frac{u_0}{2} + (B-1)\psi, \quad (10)$$

$$D(0,T) \approx D_0(0,T) \exp\left[-\frac{Q(0,T)}{k_{Bo}T}\right], \quad (11)$$

where u_0 is the cohesive energy of an atom of perfect metal, u_0^R is the cohesive energy of an atom of defective metal, ψ is the Helmholtz free energy of perfect metal, ψ^R is the Helmholtz free energy of defective metal, $\psi^{(1)}$ is the Helmholtz free energy of the metal with a missing atom on the first coordination sphere, N is the total number of atoms of the metal, $\theta = k_{Bo}T$, k_{Bo} is the Boltzmann constant, k , γ_1 and γ_2 are the crystal parameters,

$x = \frac{\hbar\omega}{2\theta}$, $\omega = \sqrt{\frac{k}{m}}$ is the vibration frequency of the atom, m is the mass of the atom,

$\hbar = \frac{h}{2\pi}$, h is the Planck constant, $Y \equiv x \coth x$, n_1 is the number of atoms on the first

coordination sphere, n_v is the equilibrium vacancy concentration, f is the correlation factor, ω in eq.(8) is the vibration frequency of the diffused atom, r_1 in eq. (8) is the length of an effective jump for the diffused atom, s_v^f is the entropy of vacancy formation determined in an approximation of the first two coordination spheres with the center as

the vacancy and the atomic numbers as n_1 and g_v^f is the change in the Gibbs thermodynamic potential of the formation of a vacancy. The numerical calculations for Au at $P = 0$, $T = 1000$ K, and $T = 1250$ K are shown in Table 2.

Table 2. Activation energy Q , pre-exponential factor D_0 , and self-diffusion coefficient D for Au at $P = 0$, $T = 1000$ K, and $T = 1250$ K calculated by SMM and according to EXPT of Gatos and Kurtz [19], Markin et al. (1957)[20], Okkerse (1956) [21]

Method	D_0 (cm^2/s)	Q (kcal/mol)	D (cm^2/s) (1000 K)	D (cm^2/s) (1250 K)
SMM	0.13	41.76	9.76×10^{-11}	7.68×10^{-9}
EXPT of Gatos and Kurtz [19]	0.265	45.3	3.32×10^{-11}	3.18×10^{-9}
EXPT of Okkerse [21]	0.031	39.4	7.58×10^{-11}	4.00×10^{-9}
EXPT of Markin et al. [20]	0.091	41.7	6.99×10^{-11}	4.65×10^{-9}

Table 2 shows the values of activation energy Q , pre-exponential factor D_0 , and self-diffusion coefficient D of Au at $P = 0$, $T = 1000$ K, and $T = 1250$ K calculated by SMM and according to experimental data from Gatos and Kurtz [19], Markin et al. [20] and Okkerse [21]. The SMM and experimental results for the Q of Au agree well with each other, especially since the error between SMM results and experiment [20] is only 1%.

We can calculate the activation energy Q of interstitial alloy AC ($A = \text{Au}$, $C = \text{Si}$) at $P = 0$ according to the following formulas

$$Q(0, T) = -u_0(0, T) + \Delta\psi^R(0, T) - \Delta\psi^{R(1)}(0, T), \quad (12)$$

$$\Delta\psi^R = (B^R - 1)\psi^R, \Delta\psi^{R(1)} = (B^{R(1)} - 1)\psi^{R(1)}, B^R \approx 1 + \frac{u_0^R}{\psi^R}, B^{R(1)} \approx 1 + \frac{u_0^R - \Delta\psi^R}{2\psi^{R(1)}}, \quad (13)$$

$$\psi^R = \sum_X \left[1 - n_v n_1 + n_v (B_X^R - 1) \right] c_X \psi_X + n_v n_a \psi_X^{(1)} - T(S_c^* + S_c), \quad (14)$$

$$\psi = \sum_X c_X \psi_X - TS_c, \quad (15)$$

$$\begin{aligned} \psi_X = N_X u_{0X} + 3N\theta \left[x_X + \ln(1 - e^{-2x_X}) \right] + \frac{3N_X \theta^2}{k_X^2} \left[\gamma_{2X} Y_X^2 - \frac{2\gamma_{1X}}{3} \left(1 + \frac{Y_X}{2} \right) \right] + \\ + \frac{6N_X \theta^3}{k_X^4} \left[\frac{4}{3} \gamma_{2X}^2 Y_X \left(1 + \frac{Y_X}{2} \right) - 2(\gamma_{1X}^2 + 2\gamma_{1X}\gamma_{2X}) \left(1 + \frac{Y_X}{2} \right) (1 + Y_X) \right], \end{aligned} \quad (16)$$

$$n_v = \exp\left(-\frac{g_v^f(AC)}{k_{Bo}T}\right) = n_v^A \exp\left(-\frac{c_C g_v^f(C)}{k_{Bo}T}\right) \approx n_v^A, n_v^A = \exp\left(-\frac{\sum_{X=A, A_1, A_2} c_X g_v^f(X)}{k_{Bo}T}\right), \quad (17)$$

$$g_v^f(X) = \frac{1}{N} \left[n_1 (\psi_X^{(1)} - \psi_X) + (B_X - 1) \psi_X \right], B_X \approx 1 + \frac{u_{0X}}{\psi_X}, \quad (18)$$

where $X = A, A_1, A_2$ and C ($A = \text{Au}, C = \text{Si}$), A are the atoms at the lattice nodes of the FCC lattice of metal A , A_1 are the atoms A at the face center of the FCC unit cell of the alloy, A_2 are the atoms A at the vertices of the alloy's FCC unit cell, C are the atoms at the body center of the FCC unit cell of the alloy, ψ is the Helmholtz free energy of perfect alloy, ψ^R is the Helmholtz free energy of defective alloy, $\psi^{(1)}$ is the Helmholtz free energy of the alloy with a missing atom on the first coordination sphere, ψ_X is the Helmholtz free energy of material X , $c_X = \frac{N_X}{N}$ is the concentration of atoms X , $c_A = 1 - 15c_C, c_{A_1} = 6c_C, c_{A_2} = 8c_C$, N_X is the number of atoms X , S_c is the configurational entropy of the alloy AB , S_c^* is the configurational entropy of the alloy AB and vacancies, $u_{0X}, k_X, \gamma_{1X}, \gamma_{2X}$ are the cohesive energy and the crystal parameters of the material X , $Y_X \equiv x_X \coth x_X, x_X = \frac{\hbar\omega_X}{2\theta}, \omega_X = \sqrt{\frac{k_X}{m_X}}$. The numerical calculations for AuSi at $P = 0$ are shown in Table 3.

Table 3. Temperature and Si concentration depend on activation energy Q (kcal/mol) and pre-exponential factor D_0 (cm²/s) for Au and AuSi at $P = 0$ calculated by SMM

T (K)	$c_{\text{Si}} = 0$		$c_{\text{Si}} = 1\%$		$c_{\text{Si}} = 3\%$	
	Q (kcal/mol)	D_0 (cm ² /s)	Q (kcal/mol)	D_0 (cm ² /s)	Q (kcal/mol)	D_0 (cm ² /s)
977	41.47	0.10	43.89	0.05	48.67	0.01
1000	41.76	0.13	44.21	0.06	49.09	0.01
1050	42.31	0.17	44.86	0.09	49.96	0.02
1100	42.84	0.22	45.48	0.12	50.77	0.04
1150	43.35	0.27	46.07	0.16	51.54	0.05
1200	43.82	0.33	46.62	0.21	52.28	0.08
1250	44.27	0.39	47.15	0.26	52.98	0.10
1300	44.70	0.46	47.65	0.31	53.65	0.14
1321	45.11	0.52	48.14	0.37	53.93	0.15

The temperature and Si concentration dependence of the activation energy Q and the pre-exponential factor D_0 for Au and AuSi at $P = 0$ calculated by SMM are summarized in Table 3. According to this table, the interstitial atom Si strongly influences diffusion in AuSi. For AuSi at 1321 K, when c_{Si} increases from 0 to 3%, Q increases from 45.11 kcal/mol to 53.93 kcal/mol (19.6% increase), D_0 decreases from 0.52 cm²/s to 0.15 cm²/s (down 71.2%). For both Au and AuSi, Q and D_0 increase with temperature. When the temperature increases from 977 K to 1321 K, the ratio $\Delta Q/\Delta T$ is 1.0×10^{-2} kcal/(mol.K) for Au, 1.2×10^{-2} kcal/(mol.K) for AuSi with $c_{\text{Si}} = 1\%$ and 1.5×10^{-2} kcal/(mol.K) for AuSi with $c_{\text{Si}} = 3\%$.

We can calculate the activation energy Q of ternary interstitial and substitutional alloy ABC ($A = \text{Au}, B = \text{Cu}, C = \text{Si}$) at $P = 0$ according to the following formulas:

$$Q(0, T) = -u_0(0, T) + \Delta\psi^R(0, T) - \Delta\psi^{R(1)}(0, T), \quad (19)$$

$$\Delta\psi^R = (B^R - 1)\psi^R, \Delta\psi^{R(1)} = (B^{R(1)} - 1)\psi^{R(1)}, B^R \approx 1 + \frac{u_0^R}{\psi^R}, B^{R(1)} \approx 1 + \frac{u_0^R - \Delta\psi^R}{2\psi^{R(1)}}, \quad (20)$$

$$\psi^R = \sum_X \left[1 - n_v n_1 + n_v (B_X^R - 1) \right] c_X \psi_X + n_v n_d \psi_X^{(1)} - T(S_c^* + S_c), \quad (21)$$

$$\psi = \sum_X c_X \psi_X + TS_c^{AC} - TS_c, \quad (22)$$

$$\psi_X = N_X u_{0X} + 3N\theta \left[x_X + \ln(1 - e^{-2x_X}) \right] + \frac{3N_X \theta^2}{k_X^2} \left[\gamma_{2X} Y_X^2 - \frac{2\gamma_{1X}}{3} \left(1 + \frac{Y_X}{2} \right) \right] + \frac{6N_X \theta^3}{k_X^4} \left[\frac{4}{3} \gamma_{2X}^2 Y_X \left(1 + \frac{Y_X}{2} \right) - 2(\gamma_{1X}^2 + 2\gamma_{1X} \gamma_{2X}) \left(1 + \frac{Y_X}{2} \right) (1 + Y_X) \right], \quad (23)$$

$$n_v = \exp\left(-\frac{g_v^f(ABC)}{k_{Bo}T}\right) = n_v^A \exp\left(-\frac{c_B g_v^f(B)}{k_{Bo}T}\right) \exp\left(-\frac{c_C g_v^f(C)}{k_{Bo}T}\right) \approx n_v^A \exp\left(-\frac{c_B g_v^f(B)}{k_{Bo}T}\right), n_v^A = \exp\left(-\frac{\sum_{X=A, A_1, A_2} c_X g_v^f(X)}{k_{Bo}T}\right), \quad (24)$$

$$g_v^f(X) = \frac{1}{N} \left[n_1 (\psi_X^{(1)} - \psi_X) + (B_X - 1) \psi_X \right], B_X \approx 1 + \frac{u_{0X}}{\psi_X}, \quad (25)$$

where $X = A, A_1, A_2, B$ and C ($A = \text{Au}$, $B = \text{Cu}$, $C = \text{Si}$), A are the atoms at the lattice nodes of the FCC lattice of metal A, A_1 are the atoms at the face center of the FCC unit cell of the alloy, A_2 are the atoms A at the vertices of the alloy's FCC unit cell, B are the substitutional atoms A_1 at the face center of the FCC unit cell, C are the atoms at the body center of the FCC unit cell of the alloy, ψ_X is the Helmholtz free energy of material X, $c_A = 1 - c_B - 15c_C$, $c_{A_1} = 6c_C$, $c_{A_2} = 8c_C$, S_c^{AC} , S_c , and are configurational entropies of the alloy AC, the alloy ABC, the alloy ABC, and vacancies. The numerical calculations for AuCuSi at $P = 0$ are shown in Table 4 and Table 5.

Table 4. Temperature, Cu concentration, and Si concentration dependences of pre-exponential factor D_0 (cm²/s) for Au, AuCuSi at $P = 0$ calculated by SMM

$T(\text{K})$	$c_{\text{Cu}} = 0\%$, $c_{\text{Si}} = 0\%$	$c_{\text{Cu}} = 3\%$, $c_{\text{Si}} = 1\%$	$c_{\text{Cu}} = 5\%$, $c_{\text{Si}} = 3\%$
977	0.10	0.06	0.01
1000	0.12	0.07	0.01
1050	0.17	0.11	0.02
1100	0.22	0.15	0.04
1150	0.27	0.20	0.06
1200	0.33	0.26	0.08
1250	0.39	0.32	0.11
1300	0.46	0.40	0.15
1321	0.52	0.43	0.16

The temperature, Cu concentration, and Si concentration dependence of the pre-exponential factor D_0 for Au, and AuCuSi at $P = 0$ calculated by SMM is summarized in Table 4. According to this table, as the temperature increases, D_0 increases. The substitutional atom Cu affects diffusion less than the interstitial atom Si. For example, for AuCuSi at the same concentration $c_{\text{Si}} = 1\%$ and temperature $T = 977$ K, when c_{Cu} increases from 0% to 3%, D_0 increases from 0.05 cm²/s to 0.06 cm²/s. This is because the size of atom the of Cu is close to the size of atom the of Au (Cu is located near Au in the periodic table).

Table 5. Temperature, Cu concentration, and Si concentration dependences of activation energy Q (kcal/mol) for Au and AuCuSi at $P = 0$ calculated by SMM

$T(\text{K})$	$c_{\text{Cu}} = 0\%,$ $c_{\text{Si}} = 0\%$	$c_{\text{Cu}} = 3\%,$ $c_{\text{Si}} = 1\%$	$c_{\text{Cu}} = 5\%,$ $c_{\text{Si}} = 3\%$
977	41.47	44.53	49.53
1000	41.75	44.85	49.96
1050	42.31	45.53	50.86
1100	42.84	46.17	51.70
1150	43.35	46.77	52.50
1200	43.82	47.33	53.26
1250	44.27	47.87	53.98
1300	44.70	48.39	54.68

The temperature, Cu concentration, and Si concentration dependence of the activation energy Q for Au and AuCuSi at $P = 0$ calculated by SMM are summarized in Table 5. According to these results, as the temperature increases, the Q of Au and AuCuSi both increase. At the same temperature, if the Si concentration increases, Q increases sharply. When Cu concentration increases, Q also increases but not significantly. For example, for AuCuSi at temperature $T = 1100$ K, $c_{\text{Si}} = 3\%$ when c_{Cu} increases from 0 to 5%, Q increases from 50.77 kcal/mol to 51.70 kcal/mol (an increase of only 1.8%).

The numerical calculations for AuSi and AuCuSi at $P = 0$ are shown in Table 6.

The temperature, Cu concentration, and Si concentration dependence of the self-diffusion coefficient D for Au, AuSi, and AuCuSi at $P = 0$ calculated by SMM and according to experimental data from Markin et al. (1957) [20] is summarized in Table 6. According to these results for both Au, AuSi, and AuCuSi, D increases significantly with temperature. When the temperature rises from 977 to 1321 K, the increase rate of D with a temperature of Au $\Delta D/\Delta T$ calculated by SMM is 7.82×10^{-12} cm²/(sK) and 3.33×10^{-11} cm²/(sK) according to experimental data of Markin et al. [20]. For AuCuSi, as c_{Si} increases, D decreases sharply. Specifically, for AuCuSi at temperature $T = 1321$ K, when c_{Si} increases from 0 to 3%, D decreases by 98.9%. When c_{Cu} increases, D decreases but not significantly. The results calculated by SMM for Au agree well with the experimental data.

Table 6. Temperature, Cu concentration, and Si concentration dependences of self-diffusion coefficient D (cm^2/s) for Au, AuSi, and AuCuSi at $P = 0$ calculated by SMM and according to EXPT of Markin et al. (1957) [20]

T (K)	$c_{\text{Cu}} = 0\%$, $c_{\text{Si}} = 0\%$		$c_{\text{Cu}} = 0\%$, $c_{\text{Si}} = 1\%$	$c_{\text{Cu}} = 3\%$, $c_{\text{Si}} = 1\%$	$c_{\text{Cu}} = 5\%$, $c_{\text{Si}} = 3\%$
	Markin[20]	SMM			
977.0	4.22×10^{-11}	5.34×10^{-11}	7.60×10^{-12}	6.24×10^{-12}	7.59×10^{-14}
1000.0	-	9.76×10^{-11}	1.33×10^{-11}	1.10×10^{-11}	1.47×10^{-13}
1015.5	9.76×10^{-11}	1.25×10^{-10}	1.91×10^{-11}	1.58×10^{-11}	2.27×10^{-13}
1045.0	1.08×10^{-10}	2.30×10^{-10}	3.70×10^{-11}	3.10×10^{-11}	4.95×10^{-13}
1075.0	3.01×10^{-10}	4.13×10^{-10}	6.96×10^{-11}	5.89×10^{-11}	1.05×10^{-12}
1100.0	-	6.55×10^{-10}	1.14×10^{-10}	9.79×10^{-11}	1.89×10^{-12}
1109.5	5.56×10^{-10}	7.76×10^{-10}	1.34×10^{-10}	1.18×10^{-10}	2.35×10^{-12}
1181.5	1.66×10^{-9}	2.57×10^{-9}	4.88×10^{-10}	4.41×10^{-10}	1.08×10^{-11}
1186.5	1.81×10^{-9}	2.78×10^{-9}	5.31×10^{-10}	4.81×10^{-10}	1.20×10^{-11}
1200.0	-	3.41×10^{-9}	6.64×10^{-10}	6.04×10^{-10}	1.56×10^{-11}
1221.5	3.29×10^{-9}	4.70×10^{-9}	9.37×10^{-10}	8.60×10^{-10}	2.33×10^{-11}
1284.0	7.26×10^{-9}	11.14×10^{-9}	2.39×10^{-9}	2.25×10^{-9}	7.02×10^{-11}
1300.0	-	1.37×10^{-8}	2.99×10^{-9}	2.84×10^{-9}	9.13×10^{-11}
1321.0	1.15×10^{-8}	1.77×10^{-8}	3.98×10^{-9}	3.83×10^{-9}	1.28×10^{-10}

The strain and pressure dependence of the pre-exponential factor D_0 and the self-diffusion coefficient D for metal Au, interstitial alloy AC, and interstitial and substitutional alloy ABC are determined by

$$D_0(\varepsilon) = D_0(0, T) \exp \left[\frac{\sigma_0 \varepsilon^\alpha (V^r + V^m)}{k_{\text{Bo}} T} \right], \quad (26)$$

$$V^r = N \frac{\sqrt{2}}{2} (a^{R3} - a^3), \quad (27)$$

$$D(\varepsilon) = D_0(\varepsilon) \exp \left[-\frac{Q(0, T)}{k_{\text{Bo}} T} \right], \quad (28)$$

$$D_{0x}(P) = D_{0x}(0, T) \exp \left(-\frac{PV^a(X)}{k_{\text{B}} T} \right), \quad (29)$$

$$V^a(X) = V^f(X) + V^m(X) = v(X) + V^r(X) + V^m(X), \quad (30)$$

$$v(X) = V^f(X) - V^r(X), \quad (31)$$

$$V^a(X) = -k_{\text{B}} T \frac{\partial \ln D_x(T, P)}{\partial P}, \quad (32)$$

$$D_x(P) = D_{0x}(P) \exp \left[-\frac{Q_x(0, T)}{k_{\text{B}} T} \right], \quad (33)$$

where V^a is the vacancy activation volume, V^f is the vacancy formation volume, $V^r(X)$ is the lattice recovery volume, $V^m(X)$ is the activation volume of vacancy migration, and a and a^R respectively are the mean nearest neighbor distances between

the two atoms A in perfect and defective metal A, interstitial alloy AC, interstitial and substitutional alloy ABC, $\sigma = \sigma_0 \varepsilon^\alpha$ is the stress, ε is the strain, σ_0 , and α are material constants. The numerical calculations for Au, AuSi, and AuCuSi are shown in tables from Table 7 to Table 10.

Table 7. Temperature, Si concentration, and strain dependence of self-diffusion coefficient $D(\text{cm}^2/\text{s})$ for AuSi calculated by SMM

c_{Si} (%)	T (K)	$\varepsilon = 1\%$	$\varepsilon = 3\%$	$\varepsilon = 5\%$
0	700	6.336×10^{-15}	5.216×10^{-15}	3.899×10^{-15}
	900	7.053×10^{-12}	5.671×10^{-12}	3.705×10^{-12}
	1100	5.895×10^{-10}	4.584×10^{-10}	1.898×10^{-10}
1	700	0.416×10^{-15}	0.343×10^{-15}	0.258×10^{-15}
	900	0.830×10^{-12}	0.670×10^{-12}	0.450×10^{-12}
	1100	1.003×10^{-10}	0.786×10^{-10}	0.367×10^{-10}
3	700	0.002×10^{-15}	0.001×10^{-15}	0.001×10^{-15}
	900	0.010×10^{-12}	0.009×10^{-12}	0.006×10^{-12}
	1100	0.026×10^{-10}	0.021×10^{-10}	0.012×10^{-10}

The temperature, Si concentration, and strain dependence of the self-diffusion coefficient D for AuSi calculated by SMM are summarized in Table 7. According to these results, D decreases as the strain increases (or the stress increases). This rule for AuSi is completely consistent with the rule for Au according to studies by Aziz (1997) [4] and Hung et al. (2007) [16].

The temperature, Cu concentration, Si concentration, and strain dependence of self-diffusion coefficient D for Au, AuCuSi calculated by SMM are summarized in Table 8. According to this result, for Au, and AuCuSi at the same temperature, when the strain increases, D decreases. For AuCuSi at the same temperature, Si concentration, and strain, when c_{Cu} increases, D decreases. For example, for AuCuSi at the same temperature $T = 700\text{K}$ and strain $\varepsilon = 1\%$ when c_{Cu} increases from 0 to 3%, D decreases from 0.416×10^{-15} to $3.290 \times 10^{-16} \text{cm}^2/\text{s}$. When c_{Cu} increases to 5%, D continues to decrease to $8.686 \times 10^{-19} \text{cm}^2/\text{s}$.

Table 8. Temperature, Cu concentration, Si concentration, and strain dependence of self-diffusion coefficient $D(\text{cm}^2/\text{s})$ for AuCuSi calculated by SMM

$c_{\text{Si}}, c_{\text{Cu}}$ (%)	T (K)	$\varepsilon = 1\%$	$\varepsilon = 3\%$	$\varepsilon = 5\%$
$c_{\text{Cu}} = 0\%$ $c_{\text{Si}} = 0\%$	700	6.336×10^{-15}	5.216×10^{-15}	3.899×10^{-15}
	900	7.053×10^{-12}	5.671×10^{-12}	3.705×10^{-12}
	1100	5.895×10^{-10}	4.584×10^{-10}	1.898×10^{-10}
$c_{\text{Cu}} = 3\%$ $c_{\text{Si}} = 1\%$	700	3.290×10^{-16}	2.424×10^{-16}	1.837×10^{-16}
	900	7.893×10^{-13}	5.161×10^{-13}	3.319×10^{-13}
	1100	6.694×10^{-11}	6.370×10^{-11}	2.296×10^{-11}
$c_{\text{Cu}} = 5\%$ $c_{\text{Si}} = 3\%$	700	8.686×10^{-19}	7.611×10^{-19}	5.830×10^{-19}
	900	6.465×10^{-15}	5.425×10^{-15}	3.732×10^{-15}
	1100	1.795×10^{-12}	1.450×10^{-12}	0.737×10^{-12}

Table 9. Pressure and Si concentration dependence of self-diffusion coefficient $D(\text{cm}^2/\text{s})$ for AuSi at $T = 1183 \text{ K}$ calculated by SMM and according to EXPT of Dickerson (1954)[22]

P (10^8 Pa)	$c_{\text{Si}} = 0\%$		$c_{\text{Si}} = 2\%$	$c_{\text{Si}} = 3\%$
	<i>TN</i> [43]	<i>SMM</i>	<i>SMM</i>	<i>SMM</i>
0.00	1.80×10^{-9}	2.721×10^{-9}	9.53×10^{-11}	1.696×10^{-11}
2.00	1.50×10^{-9}	2.193×10^{-9}	7.62×10^{-11}	1.351×10^{-11}
4.05	1.32×10^{-9}	1.760×10^{-9}	6.08×10^{-11}	1.075×10^{-11}
6.06	1.18×10^{-9}	1.421×10^{-9}	4.88×10^{-11}	8.601×10^{-12}
7.57	0.91×10^{-9}	1.211×10^{-9}	4.14×10^{-11}	7.289×10^{-12}
9.08	0.92×10^{-9}	1.032×10^{-9}	3.52×10^{-11}	6.185×10^{-12}

(EXPT [22] measured in the range of temperature from 1181 K to 1194 K and calculated by SMM at 1183 K)

Table 10. Pressure, Cu concentration, Si concentration, and strain dependence of self-diffusion coefficient $D(\text{cm}^2/\text{s})$ for Au, AuCuSi at $T = 1183 \text{ K}$ calculated by SMM

P (10^8 Pa)	$c_{\text{Cu}} = 0\%$ $c_{\text{Si}} = 0\%$	$c_{\text{Cu}} = 4\%$ $c_{\text{Si}} = 2\%$	$c_{\text{Cu}} = 5\%$ $c_{\text{Si}} = 3\%$
	<i>SMM</i>	<i>SMM</i>	<i>SMM</i>
0.00	2.721×10^{-9}	6.200×10^{-11}	1.116×10^{-11}
2.00	2.193×10^{-9}	4.939×10^{-11}	8.862×10^{-12}
4.05	1.760×10^{-9}	3.918×10^{-11}	7.018×10^{-12}
6.06	1.421×10^{-9}	3.126×10^{-11}	5.597×10^{-12}
7.57	1.211×10^{-9}	2.641×10^{-11}	4.729×10^{-12}
9.08	1.032×10^{-9}	2.232×10^{-11}	4.001×10^{-12}

The pressure and Si concentration dependence of the self-diffusion coefficient D for AuSi at $T = 1183 \text{ K}$, calculated by SMM, are summarized in Table 9. The pressure dependence of the self-diffusion coefficient D for Au at $T = 1183 \text{ K}$, calculated by SMM agrees well with the experimental data of Dickerson (1954) [22]. The pressure, Cu concentration, Si concentration, and strain dependence of the self-diffusion coefficient D for Au, AuCuSi at $T = 1183 \text{ K}$, calculated by SMM are summarized in Table 10. According to these results, for both Au, AuSi, and AuCuSi at the same temperature and component concentration, when the pressure increases, the activation energy and the self-diffusion coefficient both decrease.

When $c_C = 0$, the pressure and strain dependence of diffusion quantities for the alloy ABC is the pressure and strain dependence of diffusion quantities for the substitutional alloy AB. When $c_B = 0$, the pressure and strain dependence of the diffusion quantities for the alloy ABC is the pressure and strain dependence of diffusion quantities for the interstitial alloy AC. When $c_B = 0$ and $c_C = 0$, the pressure and strain dependence of the diffusion quantities for the alloy ABC is the pressure and strain dependence of diffusion quantities for the main metal A.

The effects of pressure and strain on diffusion quantities such as activation energy, pre-exponential factor, and self-diffusion coefficient of metals have been studied by Aziz [4], [5] and by SMM [11], [16]. We have extended the results of the study of the self-diffusion of metals to binary intercalation alloys [15] and to ternary substitutional and interstitial alloys in this work.

3. Conclusions

This article presents a self-diffusion theory, including analytical expressions for activation energy, pre-exponential factor, and self-diffusion coefficient, depending on temperature, pressure, the concentration of substitutional atoms, the concentration of interstitial atoms, and strain for substitutional and interstitial ternary alloys with the FCC structure, based on the statistical moment method (SMM). The theoretical results are applied to numerical calculations for the AuCuSi alloy. The SMM numerical results for AuCuSi are compared with those for AuSi, AuCu, and Au. The variation of self-diffusion quantities with temperature and strain in AuCuSi follows the same patterns as in AuCu, AuSi, and Au. The variation with the interstitial atom concentration in AuCuSi follows the same pattern as in AuSi, while the variation with the substitutional atom concentration follows the same pattern as in AuCu. The SMM numerical results for Au's self-diffusion quantities agree well with experimental data and other calculation results. Other SMM numerical results for self-diffusion quantities are novel and predict future experimental outcomes.

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