A growth kinetics model of rate decomposition for $Si_{1-x}Ge_x$ alloy based on dimer theory*

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According to the dimer theory on semiconductor surface and chemical vapor deposition(CVD) growth characteristics of $Si_{1-x}Ge_x$, two mechanisms of rate decomposition and discrete flow density are proposed. Based on these two mechanisms, the Grove theory and Fick's first law, a CVD growth kinetics model of $Si_{1-x}Ge_x$ alloy is established. In order to make the model more accurate, two growth control mechanisms of vapor transport and surface reaction are taken into account. The paper also considers the influence of the dimer structure on the growth rate. The results show that the model calculated value is consistent with the experimental values at different temperatures.

Keywords: dimer theory, rate decomposition, discrete flow density mechanisms, growth kinetics

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1. Introduction

The international technology roadmap for semiconductors (ITRS) suggests that Si-based strained technology including the strained $Si_{1-x}Ge_x$, Si, and Ge will be used to enhance the mobility of devices and circuits; it will also be the key to achieving high-field transmission for long periods of time. With the advantages of its higher mobility, cuttable energy-band, compatibility with the Si process and low cost, $Si_{1-x}Ge_x$ alloy material is widely used in microwave devices, mobile communications and high-frequency circuits.[1–3] The main preparation of $Si_{1-x}Ge_x$ alloy is various CVD technology, the mainstream processes of which are reduced pressure chemical vapor deposition (RPCVD) and ultrahigh vacuum chemical vapor deposition (UHVCVD).[4,5]

Up to now, CVD growth kinetics model of $Si_{1-x}Ge_x$ alloy is surface reactions control based on the collision mechanism. This type of model does not take the control mechanism of gas transmission in CVD growth into account, so it can only be applied to characterize the growth kinetics of low temperature strained $Si_{1-x}Ge_x$, but not suitable for high-temperature relaxation $Si_{1-x}Ge_x$. Therefore, the accuracy and adaptability can hardly characterize the CVD growth kinetics model of $Si_{1-x}Ge_x$ material.[6]

According to the surface dimer theory of the Si-based semiconductor and CVD growth characteristics of $Si_{1-x}Ge_x$ material, this paper proposes rate decomposition and discrete flow density mechanisms. From the Grove theory and Fick's first law, CVD growth kinetics model is established which can be applied in strained or relaxation $Si_{1-x}Ge_x$. In order to make the model more precise, this paper not only takes two growth control mechanisms of vapor transport and surface reaction into account, but also considers the influence of the dimer structure on the growth rate, which is different from the model only considering the mechanism of surface reactions control.

2. Dimer theory and rate decomposition mechanism

2.1. Growth characteristics of $Si_{1-x}Ge_x$ alloy

The initial theory about the CVD growth kinetics of $Si_{1-x}Ge_x$ thought that due to the characteristics of $Si_{1-x}Ge_x$ alloy, the growth rate can be simply regarded as the addition of the growth rates of Si and Ge. However, recent experimental studies[5–8] showed that the CVD growth rates of both $Si_{1-x}Ge_x$ and Si increase slowly with the increase of GeH_4 flow, which indicates that the simple addition cannot be applied in the accurate growth kinetics model.

2.2. Dimer theory

In the surface of a Si-based semiconductor, each atom has two unstable dangling bonds with the presence of only one electron. In order to reduce the number of dangling bonds and maintain stability in the dimer theory,[9,10] two adjacent atoms in the surface are paired to form a strong σ bond and a weak π bond; strong bonds directly lead to the 2×1 rebuilding of surface atoms, which form dimer rows.[9,10] There are four kinds of dimer totally, they are Si–Si, Ge–Si, Si–Ge, Ge–Ge.

Gas such as SiH_4, GeH_4, and H_2 are adsorbed by the dimer on the surface of $Si_{1-x}Ge_x$. Consequently, the chemical bonds of Si–H, Ge–H, Si–SiH_3, Si–GeH_3, Ge–H, Ge–GeH_3, etc. have the possibility to form.

Table 1 demonstrates that the bond energy of Ge–H is about 5.4 kcal/mol smaller than that of Si–H. Hence, compared with Si, the activation energy of H atoms desorption

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on the Ge surface is lower. In addition, Si–Si bond energy is larger than that of Si–Ge and Ge–Ge, while Ge–Ge bond energy is the smallest. Thus, to the same kind of Si-source (SiH_4 or SiH_2Cl_2) or Ge-source (GeH_4), the adsorption activation energy varies with different dimers and the corresponding activation energies of the surface adsorption are also different, as shown in Table 2.[11]

Table 1. Bond energy of Si-compounds and Ge-compounds.

Chemical bond	Si–H	Ge–H	Si–Si	Ge–Ge	Si–Ge
Bond energy/kcal·mol^{-1}	92.5	87.1	76.6	71.7	74.2

Table 2. SiH_4/GeH_4 activation energy of dimer adsorption on the surface of $Si_{1-x}Ge_x$.

Dimer	Surface reaction	Si–H$_4$(X = Si)	GeH$_4$(X = Ge)
Si–Si	H–Si+Si–XH_3	4.3	4.7
Si–Ge	H–Si+Ge–XH_3	2.2	2.4
Ge–Si	H–Ge+Si–XH_3	8.6	8.8
Ge–Ge	H–Ge+Ge–XH_3	6.8	6.1

Experiment shows that the desorption activation energy of the H-atom is influenced by the Ge-atom, but only a little change appears. Therefore, in the condition that the top-level atoms are Si, the desorption activation energy is substantially the same when H-atoms desorb from a Si–Si homogeneous dimer and a Si–Ge heterogeneous dimer. While, for the case that the top-level atoms are Ge, the corresponding activation energy decreases by approximately 10 kcal/mol, as shown in Table 3.

Table 3. Desorption activation energy of H-atoms from different dimers on the surface of $Si_{1-x}Ge_x$.[11]

Dimer	Si–Si	Ge–Ge	Si–Ge	Ge–Si
Activation energy/kcal·mol^{-1}	64.07	52.89	63.29	53.57

As the content of Ge is low (typically about 0.2), the Ge–Si dimer is relatively much less than the Si–Si dimer, but the difference of their adsorption activation energy is only 4.3 kcal/mol. Thus, the adsorption activation energy of Si-source on the Si–Si dimer and Ge–Si dimer can be approximately regarded as the difference between the adsorption activation energy (exothermic reaction) and the desorption activation energy (endothermic reaction) on the Si–Si dimer. Similarly, the adsorption activation energy of a Si-source on the Si–Ge dimer and Ge–Ge dimer can be approximately estimated as the difference between the adsorption activation energy and the desorption activation energy on the Si–Ge dimer.

2.3. Rate decomposition mechanism

Due to the low desorption activation energy on the surface, the Ge atoms on the growth surface are equivalent to the desorption center, which adsorb H atoms on its surface. The existence of Ge atoms reduces the adsorption activation energy of a Si source and the desorption activation energy of H atoms. Thus, increasing the availability of the growth surface and raising the surface growth rates of Si and Ge, ultimately the growth rate of the $Si_{1-x}Ge_x$ is also improved. In conclusion, the CVD growth rate of $Si_{1-x}Ge_x$ alloy significantly increases with the Ge content (GeH_4 flow) growing.

Since the growth surface is $Si_{1-x}Ge_x$ alloy, known in dimer theory, the adsorption activation energy of a Si source adsorbed on Si atoms and Ge atoms are not the same, and the reaction rate constants are also different. These two cases should be considered separately. Because there are four different dimers in the CVD growth process of $Si_{1-x}Ge_x$ alloy and adsorption activation energy on the Si–Si dimer and Ge–Si dimer of Si-source are different. This paper proposes that the activation energy of a Si source in the surface decomposition reaction and the final growth rates are different.

Based on the rate decomposition mechanism proposed in this paper, surface reaction rate R_{SiGe} of $Si_{1-x}Ge_x$ CVD growth can be divided into three parts: the surface reaction growth rate $R_{\mathrm{Si-Si}}$ of a Si source adsorbed on the dimer surface whose top atoms are Si; the surface reaction growth rate $R_{\mathrm{Si-Ge}}$ of a Si source adsorbed on the dimer surface whose top atoms are Ge; and the Ge surface reaction growth rate R_{Ge} that Ge atoms are adsorbed on the dimmer surface whose top atoms are Si or Ge, as shown as

$$R_{\mathrm{SiGe}} = R_{\mathrm{Si}} + R_{\mathrm{Ge}} = R_{\mathrm{Si-Si}} + R_{\mathrm{Si-Ge}} + R_{\mathrm{Ge}}, \tag{1}$$

where $R_{\mathrm{Si}} = R_{\mathrm{Si-Si}} + R_{\mathrm{Si-Ge}}$.

According to the Arrhenius,[12] the growth rate is determined by the growth rate constant k_{s}, the relationship between activation energy E and k_{s} is

$$k_{\mathrm{s}} = k_{\mathrm{s0}}\, \mathrm{e}^{-E/kT}. \tag{2}$$

Known from the adsorption and desorption mechanisms of the Si source, decomposition activation energy $E_{\mathrm{Si-Si}}$ of Si source on the Si–Si dimer or Si–Ge dimer of which the top level atom is Si, can be considered as the difference between the adsorption activation energy of a Si source and the desorption activation energy of H atoms, that is

$$E_{\mathrm{Si-Si}} = (E_{\mathrm{d_{Si-Si}}} - E_{\mathrm{a_{Si-Si}}})(1-x) + (E_{\mathrm{d_{Si-Ge}}} - E_{\mathrm{a_{Si-Ge}}})x. \tag{3}$$

In Eq. (3), x is Ge content in $Si_{1-x}Ge_x$ alloy, $E_{\mathrm{d_{Si-Si}}}$ and $E_{\mathrm{d_{Si-Ge}}}$ are the activation energies of H atoms desorbed from Si–Si and Si–Ge dimer, respectively, of which the top level atoms are Si, $E_{\mathrm{a_{Si-Si}}}$ and $E_{\mathrm{a_{Si-Ge}}}$ are the activation energies of SiH_4 absorbed on Si–Si and Si–Ge dimers, respectively, of which the top level atoms are also Si.

Similarly, when the top level atoms are Ge, the decomposition activation energy $E_{\mathrm{Si-Ge}}$ of a Si source on the Ge–Ge dimer or Ge–Si dimer can be calculated by

$$E_{\mathrm{Si-Ge}} = (E_{\mathrm{d_{Ge-Si}}} - E_{\mathrm{a_{Ge-Si}}})(1-x) + (E_{\mathrm{d_{Ge-Ge}}} - E_{\mathrm{a_{Ge-Ge}}})x. \quad (4)$$

The same as a Si source, the decomposition activation energy $E_{\mathrm{Ge-Ge}}$ of a Ge source is shown as

$$E_{\mathrm{Ge-Ge}} = (E_{\mathrm{d_{Ge-Ge}}} - E_{\mathrm{a_{Ge-Ge}}})x^2 + [(E_{\mathrm{d_{Ge-Si}}} - E_{\mathrm{a_{Ge-Si}}}) + (E_{\mathrm{d_{Si-Ge}}} - E_{\mathrm{a_{Si-Ge}}})](1-x)x + (E_{\mathrm{d_{Si-Si}}} - E_{\mathrm{a_{Si-Si}}})(1-x)^2. \quad (5)$$

The meaning of the components in Eqs. (4) and (5) correspond to those in expression (3).

3. CVD growth characteristics and the discrete flow density mechanism

Research shows that the growth process of $Si_{1-x}Ge_x$ alloy can be seen as not only the independent growth of Si and Ge, but also the relationship of their competition.[13] Therefore, this paper proposes discrete flow density mechanisms of the CVD growth of $Si_{1-x}Ge_x$ alloy based on the Grove theory.[14] In this mechanism, vapor transport flux density F_1 is characterized by $F_{1\mathrm{Si}}$ and $F_{1\mathrm{Ge}}$, which are vapor transport flux densities of Si-source (SiH_4, Si_2HCl_2) and Ge-source (GH_4) respectively. Surface reaction flux density F_2 is similar. In other words, the growth rates of both Si and Ge in $Si_{1-x}Ge_x$ alloy by CVD epitaxial growth can be characterized by its gas diffusion flow density and flux density on surface reaction.

Figure 1 shows the schematic of the model of discrete flow density, $F_{1\mathrm{Si}}$ and $F_{1\mathrm{Ge}}$ are the diffusion flow densities from the main flow of the Si-source and Ge-source to the surface of the substrate, respectively, and $F_{2\mathrm{Si}}$ and $F_{2\mathrm{Ge}}$ are the flux densities on surface reaction of Si-source and Ge-source, respectively.

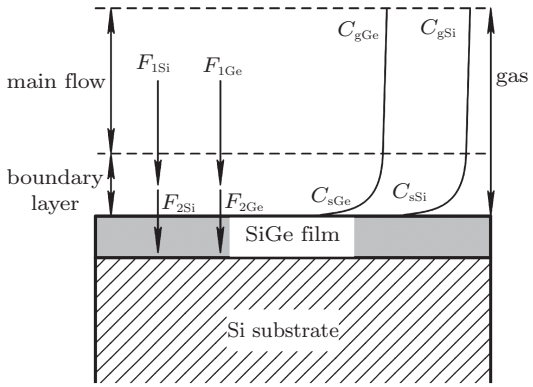


Fig. 1. Discrete flow density model of $Si_{1-x}Ge_x$ alloy by CVD.

Based on Fick's first law, the following equations can be concluded

$$F_{1\mathrm{Si}} = h_{\mathrm{g,Si}}(C_{\mathrm{g,Si}} - C_{\mathrm{s,Si}}), \quad (6)$$

$$F_{1\mathrm{Ge}} = h_{\mathrm{g,Ge}}(C_{\mathrm{g,Ge}} - C_{\mathrm{s,Ge}}), \quad (7)$$

$$F_{2\mathrm{Si}} = k_{\mathrm{s,Si}}C_{\mathrm{s,Si}}, \quad (8)$$

$$F_{2\mathrm{Ge}} = k_{\mathrm{s,Ge}}C_{\mathrm{s,Ge}}, \quad (9)$$

where $k_{\mathrm{s,Si}}$ and $k_{\mathrm{s,Ge}}$ are the reaction rate constants, $h_{\mathrm{g,Si}}$ and $h_{\mathrm{g,Ge}}$ are the gas mass transport coefficients, $C_{\mathrm{g,Si}}$ and $C_{\mathrm{g,Ge}}$ are the concentrations of the main flow, $C_{\mathrm{s,Si}}$ and $C_{\mathrm{s,Ge}}$ are the concentrations on the surface. The above symbols with subscripts Si and Ge represent the Si-source and the Ge-source, respectively.

4. Growth rate of $Si_{1-x}Ge_x$ alloy by CVD

According to the Grove theory and Fick's first law, assuming the number of atoms required to generate a unit volume of film as N_0, the growth rate of film in the stable state R can be expressed as[6,14]

$$R = \frac{Yk_{\mathrm{s}}h_{\mathrm{g}}C_{\mathrm{T}}}{(k_{\mathrm{s}}+h_{\mathrm{g}})N_0}, \quad (10)$$

where Y is the mole percent of reactants, C_{T} is the total number of molecules per cubic centimeter (including the reactants and the dilution gas) and the relationship between concentration of reactants C_{g}, Y and C_{T} is $C_{\mathrm{g}} = YC_{\mathrm{T}}$.

Due to the mechanism of rate decomposition, from expressions (1) and (8), the discrete flow density on the surface of Si can be represented as

$$F_{2,\mathrm{Si}} = k_{\mathrm{s,Si-Si}}C_{\mathrm{s,Si-Si}} + k_{\mathrm{s,Si-Ge}}C_{\mathrm{s,Si-Ge}}, \quad (11)$$

where $k_{\mathrm{s,Si-Si}}$ and $k_{\mathrm{s,Si-Ge}}$ are the reaction rate constants of Si, $C_{\mathrm{s,Si-Si}}$ and $C_{\mathrm{s,Si-Ge}}$ are the source concentrations of Si on the surface of dimer Si and dimer Ge, respectively. The variation of $C_{\mathrm{s,Si-Si}}$ and $C_{\mathrm{s,Si-Ge}}$ with the total concentrations of source Si absorbed on the growth surface $C_{\mathrm{s,Si}}$ is as follows:

$$C_{\mathrm{s,Si-Si}} = C_{\mathrm{s,Si}}(1-x), \quad (12)$$

$$C_{\mathrm{s,Si-Ge}} = C_{\mathrm{s,Si}}x, \quad (13)$$

where x is the component of Ge in the $Si_{1-x}Ge_x$ alloy.

In the equilibrium, $F_1 = F_2$, according to Eqs. (6) and (11), we can obtain

$$k_{\mathrm{s,Si-Si}}C_{\mathrm{s,Si-Si}} + k_{\mathrm{s,Si-Ge}}C_{\mathrm{s,Si-Ge}} = h_{\mathrm{g,Si}}(C_{\mathrm{g,Si}} - C_{\mathrm{s,Si}}). \quad (14)$$

Then, from Eqs. (12)–(14), $C_{\mathrm{s,Si}}$ can be expressed as

$$C_{\mathrm{s,Si}} = \frac{h_{\mathrm{g,Si}}C_{\mathrm{g,Si}}}{k_{\mathrm{s,Si-Si}}(1-x) + k_{\mathrm{s,Si-Ge}}x + h_{\mathrm{g,Si}}}. \quad (15)$$

Based on the the Grove theory and Eqs. (12), (13), and (15), each rate model can be concluded as

$$R_{\mathrm{Si-Si}} = \frac{k_{\mathrm{s,Si-Si}}C_{\mathrm{s,Si-Si}}}{N_0}$$

$$= \frac{k_{s,Si-Si}C_{s,Si}(1-x)}{N_0}$$
$$= \frac{k_{s,Si-Si}(1-x)h_{g,Si}C_{g,Si}}{N_0[k_{s,Si-Si}(1-x)+k_{s,Si-Ge}x+h_{g,Si}]}, \quad (16)$$

$$R_{Si-Ge} = \frac{k_{s,Si-Ge}C_{s,Si-Ge}}{N_0}$$
$$= \frac{k_{s,Si-Ge}C_{s,Si}x}{N_0}$$
$$= \frac{k_{s,Si-Ge}xh_{g,Si}C_{g,Si}}{N_0[k_{s,Si-Si}(1-x)+k_{s,Si-Ge}x+h_{g,Si}]}, \quad (17)$$

$$R_{Ge} = \frac{k_{s,Ge}h_{g,Ge}C_{g,Ge}}{[h_{g,Ge}+C_{g,Ge}]}. \quad (18)$$

From the mechanism of discrete flow density proposed before and Eqs. (16), (17), and (18), the growth rate of $Si_{1-x}Ge_x$ alloy by CVD is

$$R_{SiGe} = \frac{h_{g,Si}[k_{s,Si-Si}(1-x)+k_{s,Si-Ge}x]C_TY_{Si}}{[k_{s,Si-Si}(1-x)+k_{s,Si-Ge}x+h_{g,Si}]N_{0,Si}} + \frac{k_{s,Ge}h_{g,Ge}C_TY_{Ge}}{[k_{s,Ge}+h_{g,Ge}]N_{0,Ge}}. \quad (19)$$

5. Model verification

5.1. Growth experiment of $Si_{1-x}Ge_x$ alloy by RPCVD

In this paper, the growth experiment of strained $Si_{1-x}Ge_x$ alloy was conducted by an ASM-EpSilon E2000 RPCVD system. 6-inch p-type (100) Si wafer being single-side polished was adopted as the substrate and the growth source was SiH_4–GeH_4–H_2, of which the flow of H_2 and SiH_4 were 20000 slm and 20 slm, respectively.

5.2. Determination of model parameters

5.2.1. Activation energy E_a and reaction rate constant k_s

The research shows that the activation energy drops about 6 kcal/mol with the increase of the desorption temperature by 90 K.[2] Hence, the variation of activation energy with temperature can be estimated as the following expression:

$$E_{T_2} = E_{T_1} - \frac{6(T_2-T_1)}{90}, \quad (20)$$

where E_{T_2} and E_{T_1} represent the activation energy at temperatures T_2 and T_1, respectively.

According to the rate decomposition mechanism and the expressions (3)–(5), and (20), the activation energy and the corresponding rate constants of SiH_4 and GeH_4 in adsorption and decomposition reactions at different temperatures can be calculated, as shown in Table 4.

Table 4. Reaction rate constants and activation energy.

T/K	Activation energy $E_{a_{Si-Si}}$/kJ·mol^{-1}	Activation energy $E_{a_{Si-Ge}}$/kJ·mol^{-1}	Activation energy E_{aGe}/kJ·mol^{-1}	Rate constant $k_{s,Si-Si}$/cm·s^{-1}	Rate constant $k_{s,Si-Ge}$/cm·s^{-1}	Rate constant $k_{s,Ge}$/cm·s^{-1}
823	253.8	188.874	222.411	2.3516×10^9	4.1344×10^9	3.4622×10^{10}
873	239.8	174.874	208.411	4.5104×10^9	6.9084×10^9	4.7661×10^{10}
923	225.8	160.874	194.411	8.3653×10^9	1.8153×10^{10}	4.7956×10^{10}

5.2.2. Total number of gas molecules C_T

According to the ideal gas equation $pV = nRT$, the total number of gas molecules per unit volume C_T can be expressed as the following equation:

$$C_T = \frac{nN_0}{V} = \frac{pN_0}{RT}, \quad (21)$$

where V is the volume of the reaction chamber, T is the reaction temperature, R is the gas constant, n is the number of moles and N_0 is Avogadro's constant.

Table 5 shows C_T at different temperatures calculated by Eq. (21), the reaction pressure is 40 Torr.

Table 5. The total number C_T at different temperatures.

T/K	Avogadro's constant/mol^{-1}	Gas constant /J·mol^{-1}·K^{-1}	Reaction pressure/Torr	C_T/cm^{-3}
823	6.02×10^{23}	8.314472	40	4.6803×10^{17}
873	6.02×10^{23}	8.314472	40	4.4122×10^{17}
923	6.02×10^{23}	8.314472	40	4.1732×10^{17}

5.3. Verification results

The growth rates of strained $Si_{1-x}Ge_x$ alloy by RPCVD at different GeH_4/SiH_4 flow ratios and different temperatures are obtained according to Eq. (19). The comparison of results between the calculated value and the experimental value is shown in Figs. 2 and 3. These figures demonstrate the variation of strained $Si_{1-x}Ge_x$ alloy growth rates with GeH_4 flow and temperature. As seen from these figures, the strained $Si_{1-x}Ge_x$ alloy growth rate increases not only with the GeH_4 flow growing, but also with temperature increasing, which is consistent with the results in Refs. [4], [5], and [7].

The average errors of the RPCVD growth rate model in this paper are 3.5525%, 1.1425%, and 1.274%, at 550 °C, 600 °C, and 650 °C respectively, as shown in Table 6. The error is much smaller than that in Ref. [6], in which the mechanism of rate decomposition was not considered.

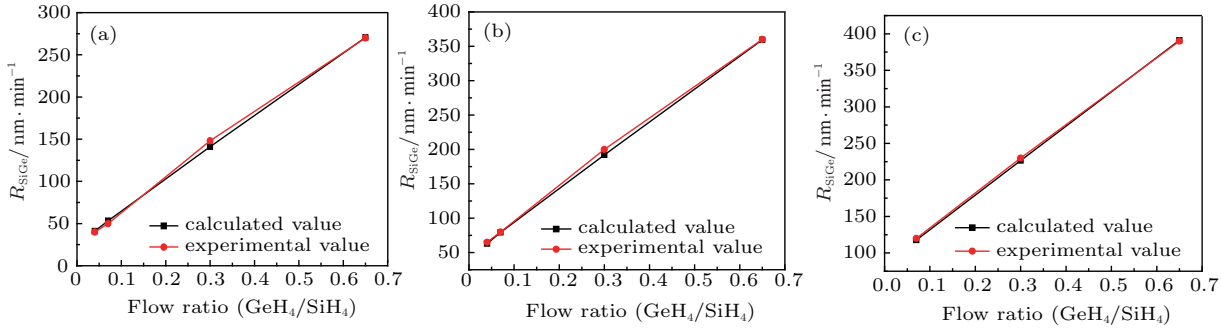


Fig. 2. (color online) Relationship between $Si_{1-x}Ge_x$ growth rates and GeH_4/SiH_4 flow ratios at (a) 823 K, (b) 873 K, (c) 923 K.

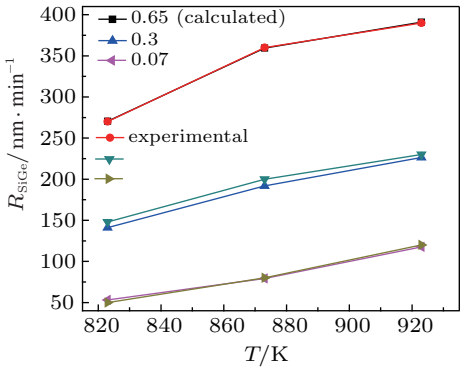


Fig. 3. (color online) Relationship between growth rates and temperatures when flow ratios are 0.65, 0.3, and 0.07, respectively.

Table 6. Model error at different GeH_4/SiH_4 flow and temperatures.

Sample	Flow	Error in this paper/%			Error in Ref. [6]
	F_{GeH_4}/sccm	550 °C	600 °C	650 °C	625 °C
1	0.8	2.30	3.21	/	/
2	1.4	6.40	0.78	2.00	/
3	6.0	5.34	0.40	1.55	/
4	13.0	0.17	0.18	0.27	/
Average error/%		3.55	1.14	1.27	7.12

6. Conclusions

According to the dimer theory, the Grove theory and CVD growth characteristics of $Si_{1-x}Ge_x$, this paper proposes rate decomposition and discrete flow density mechanisms and establishes a model of the CVD growth rate. Through the growth experiment of $Si_{1-x}Ge_x$ alloy by RPCVD, this paper compares the model calculated value with the experimental value. The results show that at 550 °C, 600 °C, and 650 °C, the average errors are 3.5525%, 1.1425%, and 1.274%, respectively, which have much smaller errors and are more accurate than those in Ref. [6]. Therefore, the rate decomposition mechanism based on the surface dimer theory turns out to be correct.

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