Diffusion Mechanisms and Intrinsic Point-Defect Properties in Silicon

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Introduction

High-purity silicon used for the growth of single crystals is a material with a high resistivity. Small traces of foreign atoms, which are mainly substitutionally dissolved on lattice sites, make the material highly conductive and therefore suitable for electronic applications. The controlled incorporation of extrinsic point defects in silicon is the main task for the production of electronic devices. Homogeneous doping is generally achieved by adding a controlled amount of the dopant element to the silicon melt. However, the fabrication of electronic devices like diodes, transistors, and complex integrated circuits requires spatially inhomogeneous dopant distributions. Control of the inhomogeneous doping profiles demanded by the considerations outlined in the article by Packan in this issue requires a detailed knowledge of the atomic mechanisms of dopant diffusion in silicon, the properties of intrinsic point defects like vacancies (V) and self-interstitials (I), and the interactions among different point defects.

This article provides a brief overview of the atomic mechanisms of self- and foreign-atom diffusion in silicon and the properties of the intrinsic defects involved. The experimentally observed diffusion coefficient D_A of an element A, which includes the silicon self-atom and different foreign atoms, will be interpreted on the basis of the diffusion mechanisms generally considered for that particular element. For more detailed information on diffusion phenomena in silicon, the interested reader is referred to the excellent reviews by Fahey et al.[1] and Hu.[2]

Mechanisms of Diffusion in Solids

Figure 1 illustrates various mechanisms for the diffusion of an element A in a solid such as silicon. The diffusion of mainly interstitially dissolved foreign atoms (A_i), like hydrogen or the $3d$ transition elements in silicon, proceeds via interstitial lattice sites. No intrinsic point defects are involved in this direct interstitial mechanism.

Direct diffusion of atoms on substitutional sites (A_s) can occur by means of a direct exchange with an adjacent silicon atom or a ring mechanism. No experimental evidence has been found for these direct mechanisms, since the diffusion of A_s by indirect mechanisms is usually more favorable.

Various indirect diffusion mechanisms, which involve intrinsic point defects, are illustrated in Figure 1b. These mechanisms can be expressed by the point-defect reactions

$$A_s + V \Longleftrightarrow AV, \tag{1}$$

$$A_s + I \Longleftrightarrow AI, \tag{2}$$

$$A_s + I \Longleftrightarrow A_i, \tag{3}$$

and

$$A_s \Longleftrightarrow A_i + V. \tag{4}$$

Reactions 1 and 2 represent the vacancy and interstitialcy mechanisms, respectively. Isolated intrinsic defects approach substitutional impurities and form next-nearest AV and AI defect pairs due to Coulomb attraction and/or minimization of local
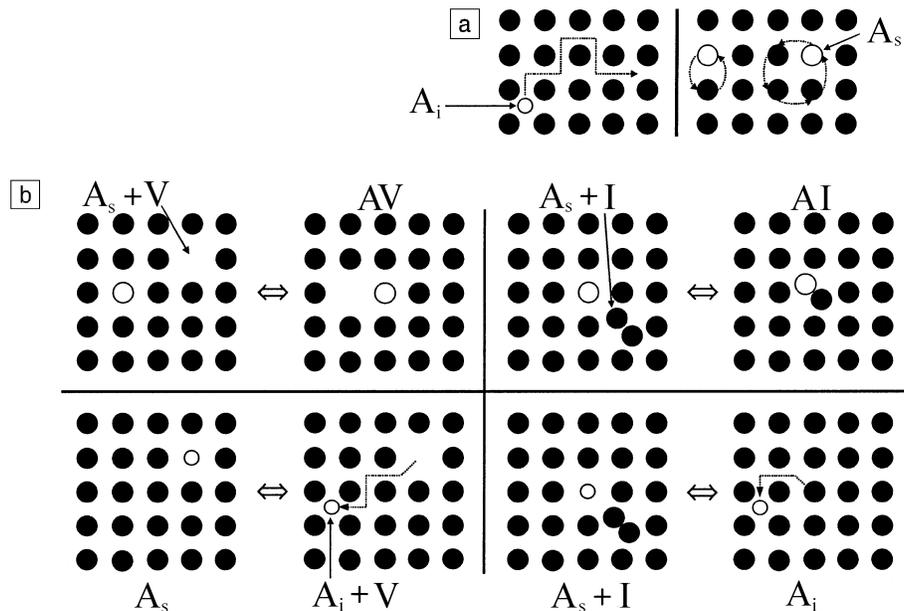

Figure 1. Schematic two-dimensional representation of (a) direct and (b) indirect diffusion mechanisms of an element A in a solid. A_i, A_s, V, and I denote interstitially and substitutionally dissolved foreign atoms, vacancies, and silicon self-interstitials, respectively. AV and AI are defect pairs of the corresponding defects.

strain. For long-range migration of A_s, the AV pair must partially dissociate, and the vacancy has to diffuse to at least a third nearest-neighbor site in the diamond lattice and return along a different path to complete the diffusion step. In contrast, dopant diffusion via the interstitialcy mechanism only occurs if the AI pair does not dissociate. Reactions 3 and 4 are the kick-out and the dissociative (or Frank–Turnbull) mechanisms, respectively. They describe the diffusion behavior of hybrid elements that are mainly dissolved on substitutional sites, but that move as interstitial defects (A_i).

Mass transport in solids can be treated on the basis of Fick's law of diffusion. In one dimension, the diffusion equation takes the form

$$\frac{\partial C_A}{\partial t} - \frac{\partial}{\partial x}\left(D_A \frac{\partial C_A}{\partial x}\right) = G_A, \qquad (5)$$

where C_A and D_A, respectively, are the concentration and diffusion coefficients of a point defect A as a function of time t and position x. Possible reactions between A and other defects are taken into account by G_A. If no reactions take place (i.e., $G_A = 0$), and a constant concentration C_A^{eq} is maintained at the surface, the solution of Equation 5 is given by

$$C_A = C_A^{eq}\left[1 - \text{erf}\left(\frac{x}{2\sqrt{D_A t}}\right)\right], \qquad (6)$$

with a concentration-independent diffusion coefficient D_A. This solution holds for the diffusion of mainly interstitially dissolved foreign atoms like hydrogen, lithium, and the $3d$ transition metals in silicon, provided that their diffusion is not affected by complex formation between the foreign atom and other defects. Fitting of the concentration profiles yields the direct interstitial diffusion coefficient D_{A_i}. Contrary to interstitial diffusion, the diffusion of mainly substitutionally dissolved dopants often results in diffusion profiles that deviate from Equation 6. In this case, the experimentally obtained diffusion coefficient D_A is a complex quantity that comprises not only the individual diffusion coefficient of the point defect governing the process, but also the equilibrium concentrations of the other defects involved in the defect reaction (see the following sections).

Intrinsic Point Defects and Silicon Self-Diffusion

The diffusion of isolated vacancies and self-interstitials in silicon can be described on the basis of Equation 5, with $G_{V,I} = 0$, assuming local equilibrium. The direct diffusion of vacancies and self-interstitials is determined by their individual diffusion coefficients D_V and D_I. Experimentally, it is difficult to measure $D_{V,I}$ because concentration profiles of vacancies and self-interstitials are not easily accessible. However, the indirect diffusion of silicon can be investigated with the aid of tagged silicon atoms. Recently, the self-diffusion coefficient of silicon D_{Si} has been accurately measured over a wide temperature range, using silicon isotope heterostructures.[3] Taking into account all possible contributions to self-diffusion (namely, contributions due to the vacancy mechanism D_{Si}^V, the interstitialcy mechanism D_{Si}^I, and the direct exchange of silicon atoms D_{ex}), the self-diffusion coefficient is given by

$$D_{Si} = D_{Si}^V + D_{Si}^I + D_{ex}$$
$$= \xi_V C_V^{eq} D_V + \xi_I C_I^{eq} D_I + D_{ex}. \qquad (7)$$

Since a successful jump of a tagged silicon atom to a next-nearest lattice site depends on the probability that an intrinsic defect is located next to the tagged atom, D_{Si} not only depends on the diffusion coefficients $D_{V,I}$, but also on the concentrations of vacancies and self-interstitials, which equal the thermal-equilibrium concentrations $C_{V,I}^{eq}$, under appropriate experimental conditions. After the site exchange, the intrinsic defect is still next to the tagged self-atom. Accordingly, the reverse jump is highly probable. These jumps, which do not contribute to the long-range diffusion, are taken into account by the correlation factors $\xi_{V,I}$. In contrast to the indirect diffusion mechanisms, diffusion via direct exchange is not correlated; that is, the correlation factor equals 1, characteristic for a random walk.

Measurements of D_{Si} alone are not definitive in establishing the mechanism of self-diffusion. In order to identify this mechanism, additional related information is required and is obtained from the analysis of self- and foreign-atom diffusion experiments involving nonequilibrium of intrinsic defects. Processes that can be used experimentally to produce nonequilibrium concentrations of self-interstitials and vacancies are discussed in the article by Cowern and Rafferty in this issue. In the next section, we briefly summarize the main conclusions about self- and foreign-atom diffusion that have been drawn from such experiments.

Foreign-Atom Diffusion

The temperature dependence of the diffusion coefficient of some foreign atoms in silicon, in comparison with self-diffusion, is shown in Figure 2.[4]

Interstitial Elements

Elements that mainly diffuse via the direct interstitial mechanism are indicated by the short-dashed lines in Figure 2. It is typical of these elements that a deviation of the vacancy and self-interstitial concentrations from thermal equilibrium does not affect their diffusion. The slower diffusion of interstitial oxygen, compared with the other interstitial impurities, is explained by the bond-centered position of interstitial oxygen, whose motion requires the breaking of two silicon bonds.[5,6]

Hybrid Elements

Gold, sulfur, platinum, and zinc are hybrid elements that are mainly substitutionally dissolved ($C_{A_s}^{eq} \gg C_{A_i}^{eq}$), but move as interstitial defects. The diffusion behavior of these foreign atoms is accurately described on the basis of the interstitial-substitutional exchange mechanisms, that is, the kick-out and dissociative mechanisms (see Reactions 3 and 4). For appropriate experimental conditions, the equations for diffusion via interstitial-substitutional exchange can be reduced to a single partial-differential equation like that given by Equation 5, with $G_A = 0$ and

$$D_A = D_A^I + D_A^V$$
$$= \frac{C_I^{eq} D_I}{C_{A_s}^{eq}}\left(\frac{C_{A_s}^{eq}}{C_{A_s}(x)}\right)^2 + \frac{C_V^{eq} D_V}{C_{A_s}^{eq}}, \qquad (8)$$

where $C_{A_s}^{eq}$ represents the equilibrium concentration of A_s,[7,8] D_A is given by the sum of a concentration-dependent self-interstitial contribution D_A^I and a concentration-independent vacancy contribution D_A^V. Each contribution is associated with the transport coefficient $C_{I,V}^{eq} D_{I,V}$ of the particular intrinsic defect. This self-interstitial- and vacancy-controlled mode of the diffusion of hybrid elements holds in crystals with low densities of dislocations and other extended defects. The diffusion in crystals with a high density of dislocations, and in dislocation-free silicon under iso-concentration conditions, is characterized by an A_i-controlled diffusion coefficient:[9]

$$D_A = D_A^{A_i} = \frac{C_{A_i}^{eq} D_{A_i}}{C_{A_s}^{eq}}. \qquad (9)$$

$D_A^{A_i}$ is the equilibrium diffusion coefficient of the hybrid elements. The thermal equilibrium of self-interstitials and vacancies is maintained during the diffusion process by, for example, dislocations that act as sinks or sources for these defects. Diffusion data of the hybrid elements shown in Figure 2 represent this A_i-controlled diffusivity.
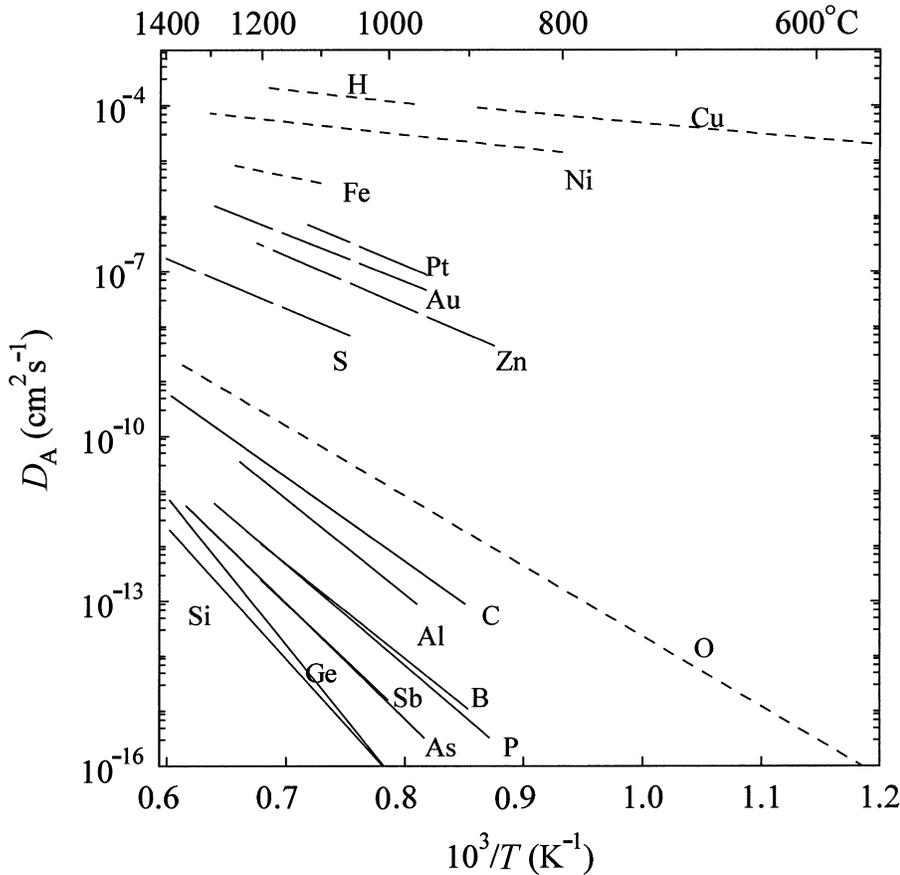
Figure 2. Temperature dependence of the diffusion coefficient of foreign atoms (A) in silicon, compared with self-diffusion. Solid lines represent diffusion data of elements that are mainly dissolved substitutionally and diffuse via the vacancy or interstitialcy mechanism. Long-dashed lines (— —) illustrate diffusion data for hybrid elements, which are mainly dissolved on the substitutional lattice site, but their diffusion proceeds via a minor fraction in an interstitial configuration. The short-dashed lines (– – – –) indicate the elements that diffuse via the direct interstitial mechanism.

Dopant Elements

The diffusion of the common dopants boron, phosphorus, arsenic, and antimony is always faster than silicon self-diffusion (see Figure 2), irrespective of whether the atom has a smaller (e.g., boron, phosphorus) or larger (arsenic, antimony) atomic radius than silicon. This can be understood as an indication that dopant diffusion is also mediated by vacancies and self-interstitials. Considering lattice distortion, small dopants attract self-interstitials and repel vacancies, whereas bigger dopants are more attractive for vacancies than for self-interstitials. The diffusion of dopants is described on the basis of the vacancy and interstitialcy mechanisms represented by Reactions 1 and 2, respectively. Both reactions are mathematically equivalent to the kick-out mechanism (Reaction 3). V and AV in the vacancy mechanism play similar roles as I and A_i in the kick-out re-

action, whereas in the interstitialcy mechanism, AI substitutes for A_i in the kick-out reaction. Correspondingly, the AV- and AI-controlled dopant diffusion is characterized by a diffusivity given by[1,4]

$$D_A = D_A^{AV} + D_A^{AI}$$

$$= \frac{C_{AV}^{eq} D_{AV}}{C_{A_i}^{eq}} + \frac{C_{AI}^{eq} D_{AI}}{C_{A_i}^{eq}}. \quad (10)$$

C_{AV}^{eq} and C_{AI}^{eq} denote the equilibrium concentration of AV and AI. The pair diffusivity D_{AV} and D_{AI} are complex quantities that comprise correlation and dissociation effects.[10] Equation 10 holds for dopant diffusion if the concentration of intrinsic point defects is in thermal equilibrium, and the dopant concentration does not exceed the intrinsic carrier concentration at the diffusion temperature (electronically intrinsic conditions). No direct ex-

perimental evidence has been found for the self-interstitial-controlled mode of dopant diffusion, which would yield a concentration-dependent diffusion coefficient like the first term on the right-hand side of Equation 8. The vacancy-controlled mode[10] is considered to be important for phosphorus diffusion under high doping levels.[11,12]

Equations 7, 8, and 10 show that both vacancies and self-interstitials may be involved in self- and foreign-atom diffusion. Accurate data of the individual contributions are of great significance for modeling dopant diffusion under nonequilibrium conditions.

Vacancy and Self-Interstitial Contributions to Self-Diffusion

Introducing the so-called fractional interstitial component of dopant diffusion,

$$f_A^{AI} = \frac{D_A^{AI}}{D_A^{AI} + D_A^{AV}} = \frac{D_A^{AI}}{D_A}, \quad (11)$$

the ratio between the dopant-diffusion coefficient under nonequilibrium and equilibrium conditions is given by[1]

$$\frac{D_A'}{D_A} = (1 - f_A^{AI})\frac{C_V}{C_V^{eq}} + f_A^{AI}\frac{C_I}{C_I^{eq}}. \quad (12)$$

This equation describes the impact of a super- or undersaturation of intrinsic point defects on dopant diffusion. Figure 3 illus-
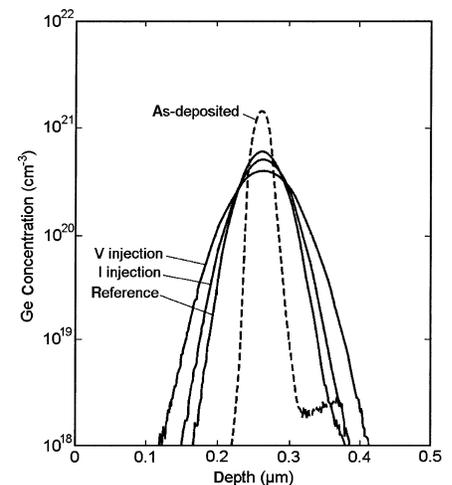

Figure 3. Concentration profiles of a buried germanium epitaxial layer in silicon, measured by secondary-ion mass spectrometry before (dashed curve) and after (solid curves) diffusion at 1050°C for 80 min under boundary conditions that cause either vacancy (V) or self-interstitial (I) injection, or no injection of intrinsic point defects.[13]

trates the diffusion of the Group IV element germanium under equilibrium and nonequilibrium conditions.[13] The broadening of the germanium-doped buried epitaxial silicon layer increases both by vacancy and self-interstitial injection, compared with annealing in inert ambient. Under the same experimental conditions, an antimony-doped buried layer reveals enhanced and retarded diffusion under vacancy and self-interstitial injection, respectively[13] (not shown in Figure 3). The analysis of the germanium and antimony diffusion profiles yields $f_{Ge}^{GeI} = 0.3$–0.4 and $f_{Sb}^{SbI} \leq 0.02$ for the fractional interstitial component at 1050°C.

Similar diffusion experiments with boron- and antimony-doped buried layers yield $f_B^{BI} \geq 0.98$ for 860°C and $f_{Sb}^{SbI} \leq 0.01$ for 790°C.[14] Generally, f_B^{BI} and f_{Sb}^{SbI} are considered to be almost constant for all temperatures between 800°C and 1200°C. Accordingly, boron and antimony diffuse almost entirely via the interstitialcy and vacancy mechanisms, respectively. Like boron, the diffusion of phosphorus is mainly mediated by the interstitialcy mechanism, whereas the fractional interstitial component of arsenic appears to be lower than that of phosphorus, indicating a higher vacancy component for arsenic than for phosphorus (see, for example, References 1 and 4). The fractional interstitial components of diffusion for some common dopants are listed in Table I.

In analogy to dopant diffusion, the fractional interstitial component f_{Si}^I of silicon self-diffusion can be deduced from self-diffusion experiments under equilibrium and nonequilibrium conditions. Such self-diffusion experiments were recently performed with isotopically enriched silicon structures, yielding $f_{Si}^I \approx 0.67$ for temperatures between 1000°C and 1100°C.[17] More detailed information about the vacancy and self-interstitial contributions to self-diffusion can be deduced from the diffusion behavior of hybrid elements. The diffusion coefficient of these elements in dislocation-free silicon is directly related to $C_I^{eq}D_I$ and $C_V^{eq}D_V$ (see Equation 8). These self-interstitial- and vacancy-related transport coefficients can be determined

separately from in- and out-diffusion experiments, which have been recently performed with zinc at temperatures between 850°C and 1200°C.[9,18,19] The temperature dependence of $C_I^{eq}D_I$ and $C_V^{eq}D_V$ deduced from these experiments is shown in Figure 4, compared with the recently measured self-diffusion data. The agreement between the direct silicon self-diffusion and the sum of the vacancy and self-interstitial contributions to self-diffusion implies that self-diffusion is mediated both by vacancies and self-interstitials.[20,21]

Diffusion at High Doping Levels

In semiconductors, point defects can occur in different charge states and may introduce energy levels within the bandgap. Occupation of the energy states depends on the Fermi-level position. Under extrinsic conditions, that is, when the hole concentration or electron concentration introduced by doping exceeds the intrinsic carrier concentration, the Fermi level deviates from its intrinsic condition E_f^{in}. As a consequence, the concentration of charged defects in thermal equilibrium changes.[22] Assuming a defect X [X $\in$ (V, I, A$_s$, A$_i$, AV, AI)] in heavily p-type silicon, which introduces a donor level at $E_X^{0/+}$ (as illustrated in Figure 5), the formation of the positively charged defect X$^+$ is energetically more favorable in p-type silicon than under intrinsic conditions. Because the total concentration of X is given by the sum of the equilibrium concentrations of the various charge states, C_X^{eq} is altered if the Fermi level changes. Correspondingly, D_A is also affected, which is correlated with the equilibrium concentrations of the defects mediating the diffusion (see Equations 7–10).

The doping dependence of silicon self-diffusion,[23] as well as the diffusion of the hybrid element gold in heavily p-type silicon,[24] points to positively charged self-interstitials I$^+$ with a donor level located at 0.3–0.4 eV above the valence-band edge E_V. Positively charged defects also govern the diffusion of p-type dopants like boron, aluminum, and gallium in p-type material, whereas negatively charged defects mediate the diffusion of n-type dopants like phosphorus, arsenic, and antimony at

high electron concentrations.[4] For example, the diffusion coefficients of arsenic as a function of the phosphorus background concentration are shown in Figure 6.[25,26] For doping concentrations lower
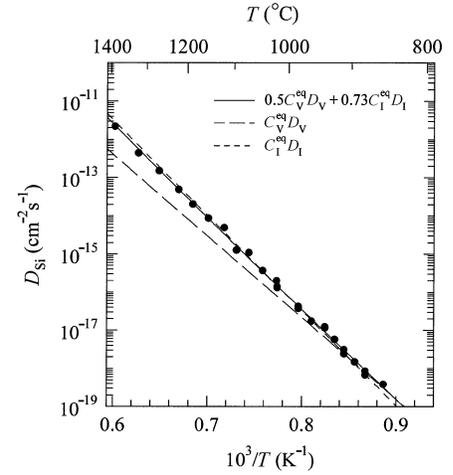
Figure 4. Temperature dependence of the measured direct silicon self-diffusion coefficient D$_{Si}$ (symbols) compared with the products C$_I^{eq}$D$_I$ (short-dashed line)[9] and C$_V^{eq}$D$_V$ (long-dashed line)[18,19] between the thermal equilibrium concentration and the diffusion coefficient of the particular intrinsic point defect. The results given for C$_I^{eq}$D$_I$ and C$_V^{eq}$D$_V$ were obtained from zinc diffusion experiments. The remarkable agreement between D$_{Si}$ and ξ_VC$_V^{eq}$D$_V$ + ξ_IC$_I^{eq}$D$_I$ (solid line), taking into account the calculated correlation factors $\xi_I = 0.73$ and $\xi_V = 0.5$, implies that both vacancies and self-interstitials mediate silicon self-diffusion.
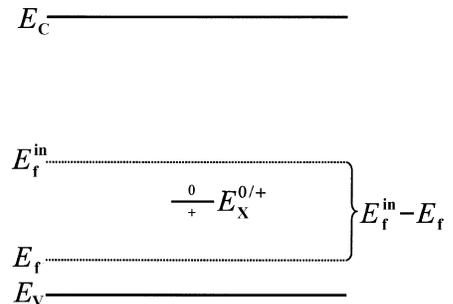
Figure 5. Schematic illustration of the effect of the Fermi-level position on the formation of charged point defects. For p-type doping conditions, the energy of formation E$_f$ of a positively charged defect X$^+$, which introduces a defect state within the bandgap, is lowered by $\Delta E = E_f^{in} - E_f$, compared with intrinsic conditions.

Table I: Fractional Interstitial Component f_A^{AI} of Dopant Diffusion in Silicon.

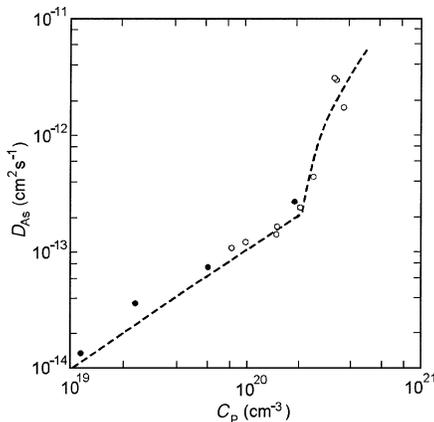| Element (A)	f_A^{AI}	Temperature (°C)	Reference
B	≥0.94	810, 860	14
Ga	≥0.95	1050	13
P	≥0.93	1100	15
As	≈0.4	1100	16
Sb	≤0.02	790, 1050	13, 14

Figure 6. Diffusion coefficient D_{As} of arsenic in silicon at 1050°C as a function of phosphorus background concentration C_P (open symbols are from Reference 25, closed symbols are from Reference 26). The dashed line was calculated with the vacancy-percolation model.[25]

Table II: Thermodynamic Properties of Self-Interstitials (I) and Vacancies (V) in Silicon, Obtained from Experimental and Theoretical Studies.[a]

A	$H_A^f + H_A^m$	$S_A^f + S_A^m$	H_A^f	S_A^f	H_A^m	S_A^m
I	4.95 eV	13.2 k	3.2–3.5 eV	4–6 k	1.4–1.8 eV	7–9 k
V	4.24 eV	6.3 k	2.0–4.0 eV	1–6 k	0.2–2.2 eV	0–6 k

[a] See References 9 and 20 and references therein.
H^f and H^m are the enthalpy of formation and migration, respectively.
S^f and S^m are the corresponding entropy of formation and migration, respectively.
k is the Boltzmann constant.

than 2×10^{20} cm^{-3}, arsenic diffusion depends linearly on background concentration, which points to a singly negatively charged defect mediating the diffusion process. The strong increase of arsenic diffusion at doping concentrations exceeding 2×10^{20} cm^{-3}, which also has been observed for germanium, tin, and antimony diffusion, is proposed to be caused by a vacancy-percolation phenomenon.[25] Other models, based on more complex vacancy-impurity pairs than that involved in Reaction 1, were also proposed to explain the observed doping dependence.[27]

An unambiguous identification of the processes operative under high doping levels is difficult because, with increasing doping level, not only the position of the Fermi level but also impurity-defect interactions and clustering of point defects can significantly affect impurity diffusion. New experimental approaches and a careful reanalysis of previous results may provide more information about the diffusion processes and charge states of point defects in silicon under extrinsic doping conditions.

Intrinsic Point-Defect Properties

The properties of intrinsic point defects are of fundamental significance for modeling diffusion processes under equilibrium and nonequilibrium conditions. The enthalpy of formation $H_{V,I}^f$ and migration $H_{V,I}^m$ and the corresponding entropies $S_{V,I}^f$ and $S_{V,I}^m$ of vacancies and self-interstitials, which are considered to be fairly reliable, are summarized in Table II.[20,21] Accurate data for $H_{V,I}^f + H_{V,I}^m$ and $S_{V,I}^f + S_{V,I}^m$ were obtained from metal out- and in-diffusion experiments.[9,18,19] These results are sufficient for modeling self- and foreign-atom diffusion under equilibrium conditions. For modeling of diffusion processes under nonequilibrium conditions, the individual values for $H_{V,I}^f$, $H_{V,I}^m$, $S_{V,I}^f$, and $S_{V,I}^m$ have to be known. Recent experimentally and theoretically determined values for the self-interstitial-related properties are in good agreement.[9,20] However, data reported for the vacancy-related properties are not as consistent. Values of H_V^m between 0.18 and 0.45 eV, depending on the vacancy charge state, were found at cryogenic temperatures,[28] whereas H_V^m seems to be around 2 eV at typical silicon processing temperatures; thus a temperature dependence of H_V^m seems to be indicated. A temperature dependence is expected if the vacancy is spread out over several atomic volumes. In order to check this concept of extended intrinsic point defects,[29] more reliable information about H_V^m, especially at high temperatures, is required.

Concluding Remarks

Predictive modeling of all processing steps used in silicon-device technologies requires a detailed knowledge about (1) the properties of intrinsic point defects, (2) the mechanisms of dopant diffusion under intrinsic and extrinsic doping conditions, (3) the distribution of intrinsic defects and of their agglomerates in silicon after crystal growth, (4) surface and bulk reaction processes causing vacancy and self-interstitial injection, and (5) gettering mechanisms. Our present understanding of items (1) and (2) is summarized in this article. Items (3)–(5) are discussed in accompanying articles in this issue. In the last 10 years, much progress has been achieved in determining the vacancy and self-interstitial contributions of self- and foreign-atom diffusion. Additionally, the physical mechanism of transient-enhanced diffusion (TED) of dopants in ion-implanted silicon has been characterized.[30] Despite this progress, the properties of intrinsic defects that are important for modeling are still not accurately known. Furthermore, detailed information about diffusion mechanisms, defect interactions, and defect states under extrinsic doping conditions is lacking. Diffusion experiments with buried dopant marker layers in silicon superlattices have considerably contributed to our present understanding of TED and the individual vacancy and self-interstitial contributions of dopant diffusion. In a similar way, silicon-isotope heterostructures, which have already been successfully used for self-diffusion experiments under equilibrium[3] and nonequilibrium conditions,[17] may serve as test structures to study directly the impact of dopant diffusion on self-diffusion. These experiments are expected to provide information about dopant-defect interactions and intrinsic point-defect properties at high doping levels.

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30. P.A. Stolk, H.-J. Gossmann, D.J. Eaglesham, D.C. Jacobson, C.S. Rafferty, G.H. Gilmer, M. Jaraíz, J.M. Poate, H.S. Luftman, and T.E. Haynes, *J. App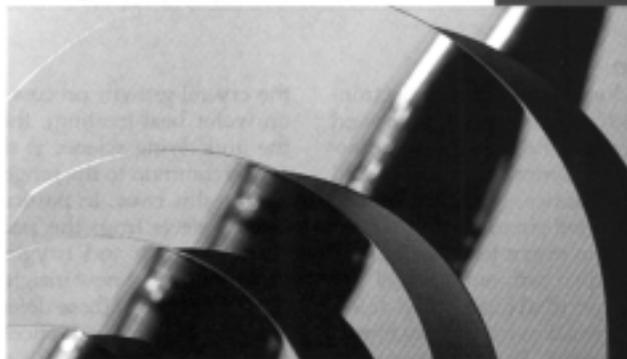l. Phys.* **81** (1997) p. 6031. □