Self-diffusion in compressively strained Ge

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Under a compressive biaxial strain of $\sim 0.71\%$, Ge self-diffusion has been measured using an isotopically controlled Ge single-crystal layer grown on a relaxed $Si_{0.2}Ge_{0.8}$ virtual substrate. The self-diffusivity is enhanced by the compressive strain and its behavior is fully consistent with a theoretical prediction of a generalized activation volume model of a simple vacancy mediated diffusion, reported by Aziz *et al*. [Phys. Rev. B **73**, 054101 (2006)]. The activation volume of (-0.65 ± 0.21) times the Ge atomic volume quantitatively describes the observed enhancement due to the compressive biaxial strain very well. © *2011 American Institute of Physics*. [doi:10.1115/1.4004462]

I. INTRODUCTION

Compressively strained Ge (s-Ge) grown on a relaxed $Si_{1-x}Ge_x$ virtual substrate (VS) is attracting much attention as a promising candidate for a next generation higher mobility p-type MOSFET.[1–3] Knowledge of self-diffusion in compressively strained Ge, which is required to understand the kinetics of the dopant diffusion and activation, is essential for the development of the s-Ge based MOS technology. In addition, the investigation of self-diffusion is important, not only to design appropriate device processing schemes, but also from the physics point of view. Self-diffusion is the most fundamental process of atomic transport in a solid. A study of self-diffusion can provide valuable information on the properties of native point defects, which are responsible for the diffusion processes. In particular, Ge self-diffusion in Ge takes place by a simple vacancy-mediated mechanism. The s-Ge matrix is, therefore, an ideal system to study and to understand the effect of strain on self-diffusion in a solid.

Recently, Si and Ge self-diffusion in relaxed $Si_{1-x}Ge_x$ has been widely investigated by probing the concentration profiles of stable or radio isotopes as tracers in relaxed $Si_{1-x}Ge_x$ epitaxial layers ($0 \leq x \leq 1$).[4–8] In this case, a decrease in the activation enthalpy with increasing Ge content, x, has been observed. The influence of compressive strain on the Si/Ge interdiffusion has been described in the literature by Cowern *et al.*[9] In their case, enhanced Ge diffusion has been observed. On the contrary, the number of self-diffusion studies in strained materials is extremely limited. Zangenberg *et al*. have reported retardation (enhancement) of Ge self-diffusion in $Si_{0.9}Ge_{0.1}$ under the tensile (compressive) strain of 0.21%.[4] However, a theoretical description of the observed phenomena was not achieved because of the complication induced by the existence of two constituents, Si and Ge, and due to multi-diffusion mechanisms involving interstitials and vacancies. Aziz *et al.*[10–12] have theoretically predicted the thermodynamic effects of hydrostatic pressure and biaxial strain on dopant- and self-diffusion mediated by the simple vacancy and interstitial mechanism in a crystalline solid based on a generalized activation volume model. They demonstrated the consistency of the prediction for antimony (Sb) diffusion in hydrostatically compressed Si and in biaxially strained Si (tensile) and SiGe (compressive). For self-diffusion, however, the theoretical finding has never been experimentally tested.

Here we report on an experimental study of self-diffusion in Ge under biaxial compressive strain. The experimental observation is quantitatively described in terms of the behavior of the vacancies in Ge by means of the activation volume with relevant previous experimental diffusion results.

II. EXPERIMENTAL

Figure 1 shows the schematic illustration of a heterostructure employed in this work. A 50-nm-thick $^{nat}Si_{0.2}{}^{nat}Ge_{0.8}$ buffer layer was grown by solid-source molecular beam epitaxy (MBE) on a $^{nat}Si_{0.2}{}^{nat}Ge_{0.8}$ VS grown by chemical vapor deposition.[13] The MBE growth of the Ge isotope superlattice (SL) consisting of alternating layers of isotopically enriched ^{70}Ge (^{70}Ge: 96.3%, ^{72}Ge: 2.1%, ^{73}Ge: 0.1%, ^{74}Ge: 1.2%, and ^{76}Ge: 0.3%) and ^{nat}Ge (^{70}Ge: 20.5%, ^{72}Ge: 27.4%, ^{73}Ge: 7.8%, ^{74}Ge: 36.5%, and ^{76}Ge: 7.8%) was performed on the $^{nat}Si_{0.2}{}^{nat}Ge_{0.8}$ buffer layer using the procedure described in Ref. 14. The thickness of each isotope layer was 8 nm. Finally, a 50-nm-thick relaxed $^{nat}Si_{0.2}{}^{nat}Ge_{0.8}$ cap layer was MBE-grown on the top as a stressor. Two ^{nat}Ge (10 nm) barrier layers sandwiching the Ge isotope SL were inserted to avoid interdiffused Si reaching the Ge isotope SL during diffusion annealing. The total thickness of the strained Ge layer was 60 nm including the Ge isotope SL and barrier layers. The sample was cut into a number of smaller pieces and they were annealed at various diffusion temperatures between 475 and 600 °C for 0.5–61 h in a

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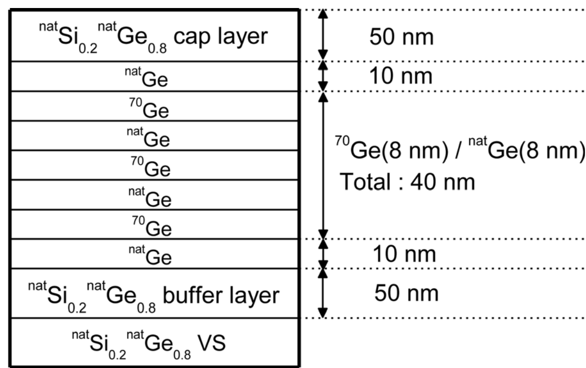

FIG. 1. Schematic illustration of the relaxed $Si_{0.2}Ge_{0.8}$ (50 nm)/compressively s-Ge isotope SL (60 nm)/relaxed $Si_{0.2}Ge_{0.8}$ (50 nm) heterostructures fabricated in this work.

resistively heated furnace under a flow of 99.999% Ar at 1.3 l/min. The samples were placed face to face with bulk Ge samples in order to avoid Ge loss from the surface during annealing.

The strains in the heterostructures were evaluated by asymmetrical (224) reciprocal space mapping (RSM) using the x-ray diffraction (XRD) system, X'pert MRD. Figure 2 shows the intensity peaks of the Si substrate, relaxed $Si_{0.2}Ge_{0.8}$ VS, and the s-Ge layer before and after annealing at 550 °C for 6 h, where Q_x and Q_y correspond to horizontal and vertical lattice constants, respectively. The Q_x of s-Ge is in excellent agreement with that of the relaxed $Si_{0.2}Ge_{0.8}$. The compressive strain in s-Ge before annealing is 0.71%. This strain corresponds to an in-plane stress of 0.95 GPa. The compressive strains in s-Ge after the diffusion annealing at 550 °C are 0.71% for 1 h, 0.68% for 3 h, and 0.66% for 6 h, which are well within the 8% relaxation compared to the initial strain. These results confirm that the heterostructures remained pseudomorphic to the underlying VS throughout the diffusion annealing. The cross-sectional transmission electron microscopy with a JEM-2100 F operating at 200 kV before and after the diffusion annealing showed that the low threading dislocation densities of $\sim 4 \times 10^6$ cm^{-2} remained constant during the annealings. The depth profiles of the concentration of ^{74}Ge in the s-Ge isotope SL were measured by secondary ion mass spectroscopy (SIMS) using PHI ADEPT1010 with a Cs^+ primary ion beam accelerated at 1.0 kV. The Ge self-diffusivities in compressively s-Ge were determined by fitting the SIMS depth profiles using a partial differential equation solver, ZOMBIE.[15]

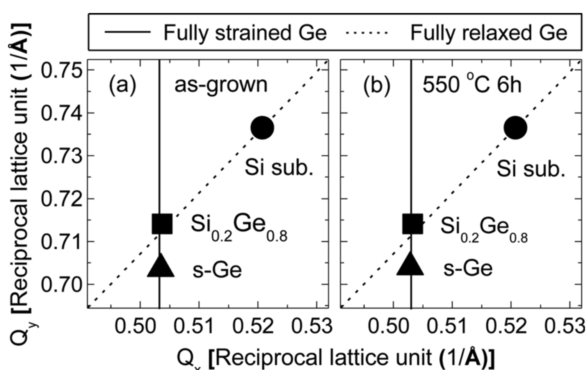

FIG. 2. Intensity peaks of the Si substrate, relaxed SiGe VS, and compressively s-Ge layer examined with asymmetrical (224) RSM using the XRD system (a) before, and (b) after annealing at 550 °C for 6 h.

III. RESULTS AND DISCUSSION

Figure 3 shows the depth profiles of ^{74}Ge in the heterostructures measured by SIMS before and after annealing at 550 °C for 1–6 h along with simulation results by ZOMBIE. We obtained a Ge self-diffusivity value of 6.40×10^{-18} cm^{2}s^{-1} for this case. Figure 4 compares the temperature dependencies of the Ge self-diffusivities in the strained and unstrained Ge obtained in this work. The unstrained Ge is a simple Ge isotope SL grown on a (001)-oriented Ge substrate employed in our previous studies.[16–18] The Ge self-diffusivities in the unstrained Ge agree with those obtained for bulk Ge reported in Ref. 19. The experimentally obtained Ge self-diffusivities are described by an Arrhenius expression, $D^{SD} = D_0^{SD}\exp(-H^{SD}/k_BT)$, with the pre-exponential factor, D_0^{SD}, the Boltzmann constant, k_B, the absolute temperature, T, and the activation enthalpy, H^{SD}; where $D_0^{SD} = 35.8\,\text{cm}^2\,s^{-1}$ and $H^{SD} = 3.07$ eV for compressively s-Ge, and $D_0^{SD} = 17.1\,\text{cm}^2\,s^{-1}$ and $H^{SD} = 3.11$ eV for unstrained Ge. The latter set of values agrees with $D_0^{SD} = 25.4$ cm$^2 s^{-1}$ and $H^{SD} = 3.13$ eV reported for bulk Ge in the temperature range of 429 to 904 °C.[19] Therefore, the enhancement of the Ge self-diffusivity by a factor of ~ 3.5 due to the compressive biaxial strain of $\sim 0.71\%$ is experimentally established in the temperature range of 475 to 600 °C. Annealing at higher temperatures introduces the relaxation of s-Ge and at lower temperatures does not induce a large enough change in the ^{74}Ge concentration profile for SIMS evaluation.

We now turn our attention to the origin of the enhancement by a factor of ~ 3.5. It is well-established that Ge self-diffusion in unstrained Ge is mediated by the simple vacancy mechanism.[18–21] The similarity between the activation enthalpy of s-Ge and that of unstrained Ge, as shown in Fig. 4, allows us to assume that the simple vacancy mechanism is also responsible for Ge self-diffusion in Ge with a

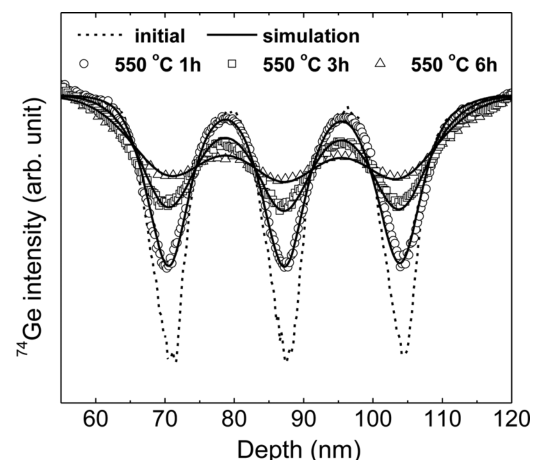

FIG. 3. SIMS and simulated depth profiles of ^{74}Ge in the relaxed $Si_{0.2}Ge_{0.8}$/s-Ge isotope SL/relaxed $Si_{0.2}Ge_{0.8}$ heterostructures. The dashed line, open circles, open squares, and open triangles represent the SIMS depth profiles before and after annealing at 550 °C for 1, 3, and 6 h, respectively. The solid curves are the simulation results.

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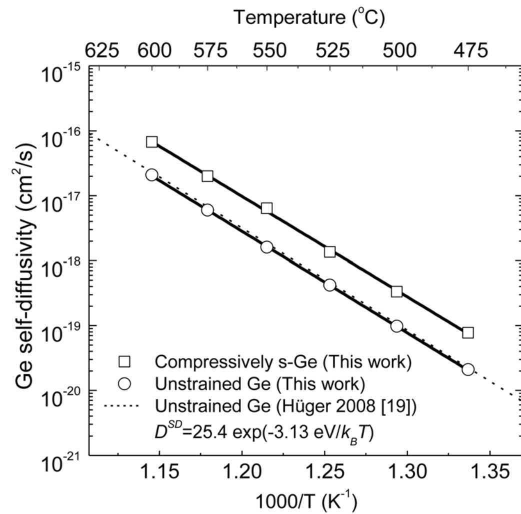

FIG. 4. Temperature dependences of Ge self-diffusivities in compressively strained and unstrained Ge. The open squares and open circles, respectively, represent the temperature dependences of Ge self-diffusivities in compressively strained and unstrained Ge obtained in this work. The solid lines show the best fits based on an Arrhenius expression. The dashed line represents the temperature dependence of the Ge self-diffusivities in unstrained Ge reported in Ref. 19.

compressive biaxial strain of $\sim 0.71\%$. The effect of biaxial strain on diffusivity has been characterized by the change in activation enthalpy, Q' with biaxial strain, $\varepsilon_{\text{biax}}$: [9–12,22,23]

$$Q' = -k_B T \frac{\partial \ln D^{SD}}{\partial \varepsilon_{\text{biax}}}\bigg|_T. \tag{1}$$

By employing Eq. (1) and our self-diffusivities, we deduced the change in the activation enthalpy of $Q' = -13 \pm 4\,\text{eV}$ per unit compressive strain. For Sb diffusion, Kringhøj *et al.*[22] reported $Q' = -13 \pm 3\,\text{eV}$ per unit strain for compressively strained $Si_{0.91}Ge_{0.09}$ and $Q' = 17 \pm 5\,\text{eV}$ for tensile Si. The similarity of the activation enthalpy of Sb to that of Ge is understandable since they both diffuse via the simple vacancy mechanism.[4] It has been proposed that the effect of stress (pressure) on diffusion in solids can be thermodynamically treated by the concept of an activation volume.[10–12,20,24,25] The change in the activation enthalpy with compressive strain can be explained by the activation volume based on the behavior of native point defects. From transition state theory, self-diffusivity can be written as, $D^{SD} = \gamma a^2 \nu f \exp(-G^{SD}/k_B T)$,[11,12,24,25] where γ is the geometrical constant, a is the lattice parameter, ν is the attempt frequency, and f is the correlation factor. Here, G^{SD} is the Gibbs free energy of point defects responsible for the self-diffusion processes. The activation volume is defined by the pressure derivative of the Gibbs free energy,

$$V^{SD} = \frac{\partial G^{SD}}{\partial p}\bigg|_T. \tag{2}$$

In addition, the pressure derivative of self-diffusivity is described as

$$k_B T \frac{\partial \ln D^{SD}}{\partial p}\bigg|_T = k_B T \frac{\partial \ln(\gamma a^2 \nu f)}{\partial p}\bigg|_T - \frac{\partial G^{SD}}{\partial p}\bigg|_T. \tag{3}$$

Since the first term on the right hand side of Eq. (3) represents a small correction well within a few percent of V^{SD}, the observed increase in D^{SD} with p is attributable to the activation volume term.[20,24,25] Therefore, V^{SD} can be described by

$$V^{SD} = -k_B T \frac{\partial \ln D^{SD}}{\partial p}\bigg|_T, \tag{4}$$

and becomes positive (negative) when D^{SD} decreases (increases) with p. For the case of the vacancy mechanism, V^{SD} is the sum of two components, $V^{SD} = V^F + V^M$, where the formation volume, V^F is the volume change of the system upon formation of a vacancy in its standard state, and the migration volume, V^M is the additional volume change when the vacancy reaches the saddle point in its migration path.[10–12,20] For a (001) thin-film geometry, the predominant source of vacancies is the free surface.[10–12] When a vacancy is created in crystalline Ge, one Ge atom leaves a lattice site and migrates to the surface. The resulting increase of the crystal volume corresponding to one Ge atomic volume (Ω_{Ge}) is 2.26×10^{-29} m³/atom.[26] However, V^F depends on the degree to which the atoms surrounding the vacancy relax toward the vacancy center. Thus, V^{SD} is decreased by the relaxation volume (V^R) which takes a negative value. In addition, V^M is also negative due to the fact that the diffusing atom via a vacancy is expanding a constriction at the saddle point in its migration path.[20] Then, the activation volume of the Ge self-diffusion under hydrostatic pressure is described as

$$V^{SD} = \Omega_{\text{Ge}} + V^R + V^M, \tag{5}$$

and is smaller than $+1\Omega_{\text{Ge}}$. Aziz *et al.*[10–12] further generalized the activation volume for a non-hydrostatic stress state by defining an activation strain tensor. The formation strain tensor ($\boldsymbol{V}^F$) is described as

$$\boldsymbol{V}^F = \Omega_{\text{Ge}} \begin{bmatrix} 0 & & \\ & 0 & \\ & & 1 \end{bmatrix} + \frac{V^R}{3} \begin{bmatrix} 1 & & \\ & 1 & \\ & & 1 \end{bmatrix}. \tag{6}$$

The first term on the right-hand side of Eq. (6) shows the volume change corresponding to Ω_{Ge} at the free surface when a vacancy is created inside the crystal. The second term indicates that the relaxation volume propagates out elastically to surfaces and provides equal contributions in all directions.[11] In addition, the migration strain tensor ($\boldsymbol{V}^M$) is defined as

$$\boldsymbol{V}^M = \begin{bmatrix} V_\perp^M & & \\ & V_\perp^M & \\ & & V_\parallel^M \end{bmatrix}, \tag{7}$$

where $V_\perp^M$ and $V_\parallel^M$ are the volume changes perpendicular and parallel to the direction of the net transport at the saddle point in its migration path. Under hydrostatic stress, the activation volume is the scalar activation volume conventionally defined in Eq. (5), which is the sum of the trace of Eq. (6), $\Omega_{\text{Ge}} + V^R$, and that of Eq. (7), V^M. In the presence of biaxial stress (σ_{biax}), the work needed to create a lattice site at the surface is zero because there is no normal force at the surface. Furthermore, the part of V^R that propagates on the free surface and interacts with the zero normal stress does not contribute to the work performed against the stress field. Therefore, the contribution of the activation volume to self-

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diffusion under biaxial stress via the vacancy mechanism, $V_{\rm biax}^{SD}$, can be described as

$$V_{\rm biax}^{SD} = \frac{2}{3}V^R + V^M - V_{||}^M, \tag{8}$$

which is dominated by the relaxation and migration volume terms. Therefore, $V_{\rm biax}^{SD}$ is negative. According to the strain tensor model and activation volume described in Eq. (4), the contribution of the activation volume to self-diffusion under biaxial stress can be described as

$$V_{\rm biax}^{SD} = -k_B T \frac{\partial \ln D_{\rm biax}^{SD}}{\partial \sigma_{\rm biax}}\bigg|_T. \tag{9}$$

Based on Eq. (9) and the Ge self-diffusivities obtained experimentally ($D_{\rm biax}^{SD}$) in this work, we find, $V_{\rm biax}^{SD} = (-0.65 \pm 0.21)\Omega_{\rm Ge}$. $V_{\rm biax}^{SD} \approx -\Omega_{\rm Ge}$ implies that upon the formation and migration of a vacancy, there exists a strong inward constriction of the lattice. The increase of Ge self-diffusivities in s-Ge results from the decrease of the activation enthalpy by the negative work performed against the stress field, $\sigma_{\rm biax}V_{\rm biax}^{SD}$. The compressive biaxial strain of 0.71% is expected to produce a small change of -0.09 ± 0.03 eV in the activation enthalpy. From the fittings by the Arrhenius expression, the difference between the activation enthalpy of self-diffusion in compressively strained Ge, 3.07 eV (this work), and in unstrained Ge, 3.13 eV (Ref. 19), can be found to be -0.06 eV ($= 3.07 - 3.13$ eV). This value is within the accuracy of the value of -0.09 ± 0.03 eV deduced from the generalized activation volume model. Furthermore, from the comparison between the biaxial and hydrostatic stress states [Eqs. (5) and (8)], the following relationship can be given:

$$V^{SD} - \frac{3}{2}V_{\rm biax}^{SD} = \Omega_{\rm Ge} + \left(V_{||}^M - V_{\perp}^M\right). \tag{10}$$

If the anisotropy in $\boldsymbol{V}^M$ is negligibly small, the right-hand side of Eq. (10) should become $+1\Omega_{\rm Ge}$. Aziz *et al.*[10–12] have described the contribution of the activation volume under biaxial stress, $V_{\rm biax}$, to the diffusion as $-(Q'/Y)$, where the biaxial modulus, Y, is the ratio of Young's modulus to one minus the Poisson ratio. For the Sb diffusion,[11,12] they have demonstrated the consistency of the predicted relationship by the values of Q' reported by Kringhøj *et al.*[22] and an activation volume $V = +0.066\Omega$ determined for hydrostatically compressed Si in their work. The results are $V + (3/2)(Q'/Y) = (+1.20 \pm 0.33)\Omega$ for tensile Si and $V + (3/2)(Q'/Y) = (+0.93 \pm 0.20)\Omega$ for the compressively strained $Si_{0.91}Ge_{0.09}$ alloy, which agrees with the prediction of $+1\Omega$ ($Y = 180.5$ GPa for these cases). Werner *et al.*[20] reported positive values of the activation volume (V^{SD}) for Ge self-diffusion in Ge under hydrostatic pressure in the temperature range of 603 to 813 °C. Inserting the $V_{\rm biax}^{SD}$ deduced in this study and $V^{SD} = +0.24\Omega_{\rm Ge}$ at 603 °C reported in Ref. 20 into Eq. (10), we find $V^{SD} - (3/2)V_{\rm biax}^{SD} = (+1.22 \pm 0.21)\Omega_{\rm Ge}$, which is fully consistent with the $+1\Omega_{\rm Ge}$ prediction.

IV. CONCLUSIONS

In conclusion, we have designed and conducted an experiment to reliably obtain the Ge self-diffusivity in biaxially compressed Ge. The compressive strain enhances Ge self-diffusion. The degree of the enhancement is described quantitatively by the theoretical prediction of a generalized activation volume model of a simple vacancy mediated diffusion.[10–12]

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