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Response to “Comment on ‘Diffusion of n-type dopants in germanium’ ” [Appl. Phys. Rev. 2, 036101 (2015)]

Bracht, H. , Südkamp, T. , Radek, M. and Chroneos, A.

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H. Bracht, T. Südkamp, M. Radek, and A. Chroneos

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APPLIED PHYSICS REVIEWS 2, 036102 (2015)

APPLIED PHYSICS REVIEWS

Response to "Comment on 'Diffusion of n-type dopants in germanium'" [Appl. Phys. Rev. 2, 036101 (2015)]

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In this reply to the comment of Cowern *et al.*, we demonstrate on the basis of full numerical simulations of radiation enhanced dopant diffusion via the kick-out mechanism that the g/λ analysis fails to consistently describe boron (B) diffusion in germanium (Ge) under irradiation. Cowern *et al.* missed to perform a consistency check with results for the diffusivity D_I of Ge interstitials (I) determined from Ge self-diffusion under irradiation. Data deduced for D_I from the exponential B profile reported by Cowern *et al.* deviate several orders of magnitude from the self-diffusion study. This clearly disproves the validity of the kick-out mechanism to control radiation enhanced B diffusion in Ge. Exponential B profiles like those established in Ge under irradiation are also reported for silicon by Venezia *et al.* [Phys. Rev. B **69**, 125215 (2004)]. The characteristic shape is not described by the kick-out mechanism but rather explained qualitatively by the complex formation and dissolution of defect clusters. Modeling of B diffusion in Ge under irradiation performed by Schneider *et al.* [Phys. Rev. B **87**, 115202 (2013)] is fully consistent with self-diffusion under irradiation. This constraint led us to conclude that the characteristic B profiles are additionally affected by the formation of immobile B clusters. Although a direct microscopic proof of B cluster formation is still lacking, the report of Venezia *et al.* on B clustering in Si during irradiation with similar exponential B profiles also supports our interpretation of B diffusion in Ge under irradiation. *© 2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4929763]

We thank the authors for their comment on our interpretation of boron (B) diffusion in germanium (Ge) under proton irradiation mentioned in Ref. 1 and explained in Ref. 2. This gives us the opportunity to describe the limits of the g/λ approach used by Cowern *et al.*[3] for the analysis of B diffusion in Ge under non-equilibrium conditions. We will demonstrate that the g/λ approach leads to a misleading view on the properties of point defects in Ge. A careful evaluation of B diffusion in Ge under non-equilibrium conditions and, more generally, of self- and dopant diffusion in Ge under irradiation reveals that additional defect reactions beside the kick-out reaction mediate the behavior of B diffusion in Ge under irradiation. A different character of Ge self-interstitials at low and high temperatures proposed by Cowern *et al.*[3] as a result of their g/λ analysis is not supported by our analysis of their experimental B profiles, which is performed on the basis of the full differential equation system for B diffusion via the kick-out mechanism.

In this reply, we analyze the B profile measured by Bruno *et al.*[4] after concurrent proton irradiation and annealing at 550 °C for 1 h. This B profile is also considered in the Comment of Cowern *et al.*[5] to support their interpretation of I-mediated B diffusion. The B diffusion profile of Bruno *et al.* illustrated in Fig. 1 reveals an exponential profile shape. Similar profiles are obtained for B diffusion in Si under equilibrium[6] and even non-equilibrium conditions.[7] The former profiles are accurately described with the g/λ model yielding results that are consistent with analyses of B diffusion based on the full kick-out diffusion model. The latter radiation enhanced diffusion profiles are concluded to be affected by the formation and dissolution of defect clusters although the shape of the B profiles in Si obtained under irradiation resembles the exponential profile shape observed under equilibrium condition. This shape is suggested by the distribution of B in clusters and its diffusion via substitutional-interstitial exchange. It is also worth noting that the B delta spikes used by Venezia *et al.*[7] for the study of radiation enhanced B diffusion in Si possess a similar doping level as the samples considered by Cowern *et al.* in their comment. Considering that the solubility of B in Ge[8] is lower than in Si[9] and the I-supersaturation established in Ge under irradiation[2] is substantially higher than in Si,[10] B clustering in Ge during irradiation is very likely and favored by the formation of mobile BI pairs under strong I supersaturation. Accordingly, B profiles in Ge measured with secondary

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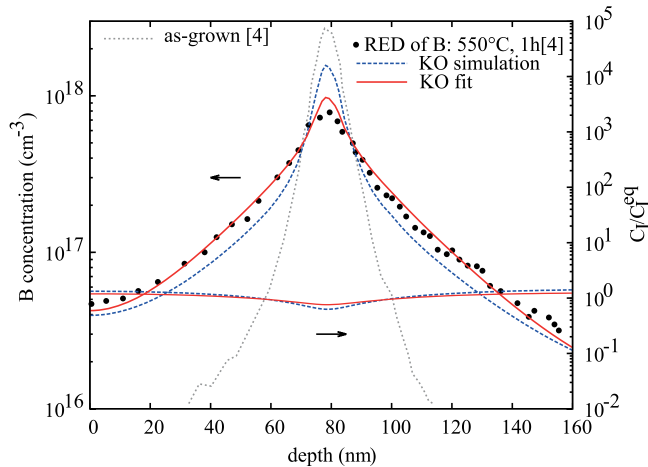

FIG. 1. Concentration profile of B before (fine dashed line) and after (•) concurrent annealing and irradiation at 550 °C for 1 h were measured by means of SIMS.[4] The upper blue dashed line shows a simulation of the radiation enhanced B diffusion on the basis of the kick-out mechanism assuming the g and λ parameters reported by Cowern *et al.*[5] and Bruno *et al.*[4] The lower blue dashed line represents the corresponding concentration C_I of Ge interstitials normalized to the equilibrium concentration C_I^{eq} and referred to the right y-axis. The red solid lines are simulations with optimized kick-out model parameters to better reproduce the experimental B profile. The corresponding g and λ parameters slightly deviate from those deduced from the g/λ analysis. These simulations on the basis of the full differential equation system describing dopant diffusion via the kick-out mechanism demonstrate that the experimental B profiles reported in the literature[3,4,11,12] can be well reproduced with the kick-out model. The simulations based on the full model reveal similar g and λ parameters as those determined from the approximated g/λ approach (see main text). For better comparison to the g/λ analysis, the formation and annihilation of self-interstitials and vacancies was not taken into account in these first simulations.

ion mass spectrometry (SIMS) after concurrent annealing and irradiation are expected to contain also a significant contribution of B atoms in clusters. Of course, this reference to B diffusion in Si under irradiation does not directly disprove the analysis and interpretation of B diffusion in Ge under irradiation reported by Cowern *et al.*[3] The reference rather indicates that dopant diffusion under irradiation can be much more complex than indicated by the shape of the measured profiles alone.

The g/λ approach was derived from the kick-out diffusion model to describe the native defect controlled diffusion of B in Si[6] analytically. The approach is applicable when this specific mode of dopant diffusion prevails under the particular experimental conditions. Cowern and coworkers[3,4,11,12] were stimulated by the exponential shape of the B profiles in Ge established under proton irradiation to assume that the radiation enhanced diffusion of B in Ge is controlled by self-interstitials. Accordingly, the transport capacity $C_{BI}D_{BI}$ is concluded to be similar or even exceeds $C_I D_I$ under radiation. However, if other defect reactions in addition to the kick-out mechanism are operative under irradiation, the analysis of B diffusion on the basis of the g/λ approach will inevitably yield misleading results on the properties of Ge self-interstitials.

In the following, we demonstrate that the g/λ model, that is, the kick-out mechanism, is not sufficient to consistently model the diffusion behavior of B in Ge under irradiation. First, we describe the B profile of Bruno *et al.*[4] illustrated in Fig. 1 on the basis of the kick-out reaction solving the underlying differential equations numerically. Assuming the values $g = 3.5 \times 10^{-4}$ s^{-1} and $\lambda = 18.8$ nm reported by Cowern *et al.*[5] and Bruno *et al.*[4] from their g/λ analysis of the same B profile and transferring these values to the model parameters of the kick-out diffusion equation system, the blue dashed lines shown in Fig. 1 are obtained. The upper blue dashed line illustrates the concentration of substitutional B_s and the lower dashed line reflects the corresponding super- and undersaturation of I that is referred to the right y-axis. A better description of the experimental profile is obtained with $g = 7 \times 10^{-4}$ s^{-1} and $\lambda = 13.3$ nm as illustrated by the red solid line. The corresponding distribution of I is indicated by the lower red solid line. Both simulations yield $g \times \lambda^2 = D_B = 1.2 \times 10^{-15}$ cm^{2}s^{-1} and are consistent with the D_B value reported by Bruno *et al.*[4] The slight deviations between the two simulations reflect the approximate nature of the g/λ approach. The simulations demonstrate that the experimental B profile shown in Fig. 1 can be reproduced by the kick-out model with transport capacities $C_{BI}D_{BI}$ and $C_I D_I$ of the mobile BI and I defects, respectively, that exceed the equilibrium quantities $C_{BI}^{eq}D_{BI}$ and $C_I^{eq}D_I$. In the g/λ approach the impact of irradiation is reflected by enhanced values of $C_{BI}D_{BI}$ and $C_I D_I$ compared to thermal equilibrium conditions. The formation of vacancies (V) and I due to irradiation and their mutual annihilation is not considered in the g/λ approach. Accordingly, the g/λ analysis of B diffusion in Ge under irradiation fails to describe the dynamic of defect production and their annihilation under irradiation. Moreover, the property of the Ge surface to inhibit the annihilation of I that is confirmed by the diffusion behavior of self- and n-type dopant atoms under irradiation[13,14] is not considered in the g/λ approach. This Ge surface property and the fact that the reaction rate for Frenkel defect annihilation decreases with decreasing temperature leads to an increasing I supersaturation with decreasing temperature that explains the increasing B diffusion with decreasing temperature observed under irradiation.[2] This scenario is not directly resolved in the g/λ analysis. It rather becomes indirectly evident in the values reported for λ at temperatures between 800 °C and 550 °C (see Fig. 2 of the Comment by Cowern *et al.*[5]).

To test the relevance of the kick-out model for B diffusion in Ge under irradiation, we added a production and annihilation term to the differential equation system that considers the formation of I and V due to irradiation and their mutual annihilation. Moreover, the property of the Ge surface to act as sink for V and to repel I is taken into account as boundary condition.[13,14] The red solid lines in Fig. 2 illustrate that the B profile of Bruno *et al.*[4] can be described on the basis of the full differential equation system for diffusion via the kick-out mechanism with additional terms that account for I- and V-formation by irradiation and their annihilation. The strong imbalance between the concentration of I and V shown by the upper and lower red dashed lines is caused by the inefficiency of the Ge surface to act as sink for I. For the simulations, we considered a Frenkel pair production rate that, according to Cowern *et al.*,[5] is three times higher than in the experiments of Schneider *et al.*[2] Other model parameters of the differential equation system are the

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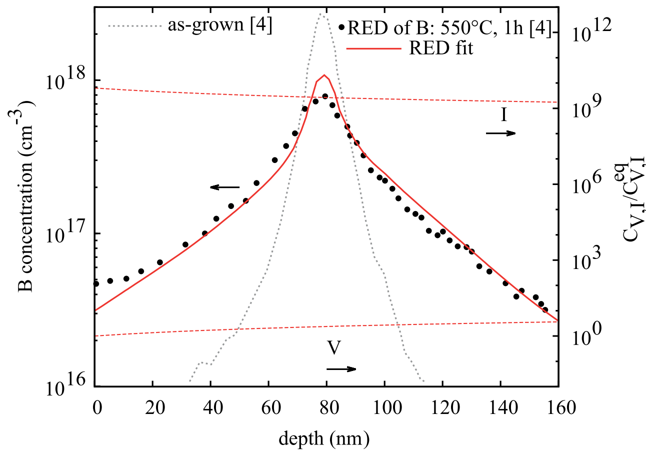

FIG. 2. SIMS concentration profile of B before (fine dashed line) and after (•) concurrent annealing and irradiation at 550 °C for 1 h.[4] The red solid line shows a fit to the experimental B profile achieved on the basis of the kick-out mechanism taking into account the formation of self-interstitials and vacancies due to irradiation and their mutual annihilation. The strong imbalance between the normalized concentration $C_{I,V}/C_{I,V}^{eq}$ of I and V is due to the Ge surface property to annihilate V and to repel I.[13,14] The simulations demonstrate that the B profiles in Ge established under irradiation are well described on the basis of the kick-out model. This, however, does not support the g/λ of Cowern *et al.* and their conclusions concerning the low and high temperature properties of I because the data deduced for the diffusion coefficient D_I is several orders of magnitude lower than those determined from self-diffusion under irradiation.[2,17] This inconsistency to Ge self-diffusion under irradiation disproves that only the kick-out mechanism controls B diffusion in Ge under irradiation. Since the g/λ analysis fails to consistently describe the B diffusion behavior in Ge under irradiation, the different properties of I at low and high temperatures are not seriously supported by Cowern *et al.*[3]

atomic fractions C_I^{eq}/C_0 and C_{BI}^{eq}/C_0 for thermal equilibrium conditions and the equilibrium transport capacities $C_{BI}^{eq}D_{BI}$ and $C_I^{eq}D_I$. The rate constants of the kick-out reaction are considered to be diffusion controlled with a capture radius $a = 5.66 \times 10^{-8}$ cm equal to the Ge lattice constant. The quantity C_I^{eq}/C_0 was calculated from its temperature dependence described by the prefactor 2.36×10^6 and I formation enthalpy 3.25 eV according to atomistic calculations.[15] $C_0 = 4.413 \times 10^{22}$ cm^{-3} is the Ge atom density. The transport capacity $C_{BI}^{eq}D_{BI}$ in thermal equilibrium or rather the quantity $C_{BI}^{eq}D_{BI}/C_{B_s}^{eq}$ was deduced from the temperature dependence of B diffusion reported by Uppal *et al.*[16] Only the model parameters C_{BI}^{eq}/C_0 and $C_I^{eq}D_I$ or rather $C_{BI}^{eq}/C_{B_s}^{eq}$ and $C_I^{eq}D_I/C_{B_s}^{eq}$ were optimized to fit the B profile shown in Fig. 2. The experimental profile is best reproduced with $C_{BI}^{eq}/C_{B_s}^{eq} = 1.05 \times 10^{-11} \pm 1.08 \times 10^{-12}$ and $C_I^{eq}D_I/C_{B_s}^{eq} = (1.90 \times 10^{-25} \pm 1.25 \times 10^{-26})$ cm^2 s^{-1}. The ratios of the equilibrium concentrations and transport capacities between BI and I are given by $C_{BI}^{eq}/C_I^{eq} = 2.23 \times 10^{-2}$ and $C_{BI}^{eq}D_{BI}/C_I^{eq}D_I = 35.1$. In particular, the latter value of $C_{BI}^{eq}D_{BI}/C_I^{eq}D_I$ describes a native defect controlled diffusion mode that seemingly is reflected by the exponential shape of the B profile. In terms of the g/λ model the values of g and λ can be calculated from the kick-out model parameters via the expressions

$$g = 4\pi a D_I C_I(x,t), \quad (1)$$

$$\lambda = \sqrt{\frac{D_{BI}}{4\pi a D_I \frac{C_{B_s}^{eq} C_I^{eq}}{C_{BI}^{eq}}}}. \quad (2)$$

Equations (1) and (2) yield

$$g\lambda^2 = \frac{C_{BI}^{eq} D_{BI}}{C_{B_s}^{eq}} \frac{C_I(x,t)}{C_I^{eq}} = D_B \frac{C_I(x,t)}{C_I^{eq}}. \quad (3)$$

The equilibrium concentration $C_{B_s}^{eq}$ of substitutional B_s is set to the maximum concentration of the B delta spike, i.e., $C_{B_s}^{eq} = 2.8 \times 10^{18}$ cm^{-3}. The I supersaturation of $C_I(x,t)/C_I^{eq} = 2.6 \times 10^9$ at B peak position is obtained from the numerical simulation (see Fig. 2). The value of $C_{BI}^{eq}D_{BI}/C_{B_s}^{eq} = 6.67 \times 10^{-24}$ cm^2 s^{-1} at 550 °C is calculated from the temperature dependence of the B diffusion coefficient reported by Uppal.[16] $C_I^{eq}/C_0 = 3 \times 10^{-14}$ is obtained from the temperature dependence reported by Vanhellemont *et al.*[15] Taking into account the Ge lattice constant $a = 5.66 \times 10^{-8}$ cm and $C_I^{eq}D_I(550\,°C) = 5.32 \times 10^{-7}$ cm^{-1} s^{-1} and $C_I(x = 80\,\text{nm})/C_I^{eq} = 2.6 \times 10^9$ from modeling the experimental B profile, Eqs. (1) and (2) yield $g = 9.8 \times 10^{-4}$ s^{-1} and $\lambda = 42$ nm, respectively. These values are of the same order as $g = 3.5 \times 10^{-4}$ s^{-1} and $\lambda = 18.8$ nm determined by means of the g/λ model.[5] This seems to demonstrate that the full kick-out and g/λ model provide similar results even under non-equilibrium conditions. However, the numerical calculations based on the full differential equation system provide additional insight. The simulations reveal that the diffusion of B under irradiation only depends on D_I and D_{BI}, i.e., higher values assumed for $C_{BI}^{eq}D_{BI}/C_{B_s}^{eq}$ or $C_I^{eq}D_I/C_{B_s}^{eq}$ can be compensated by higher values of $C_{BI}^{eq}/C_{B_s}^{eq}$ or $C_I^{eq}/C_{B_s}^{eq}$. Accordingly, the B profile illustrated in Fig. 2 is not unique with respect to g, λ and the product $g\lambda^2$. The dependence of B diffusion under irradiation on the diffusivity D_X of the mobile defect $X \in \{BI, I\}$ is also confirmed by Ge self-diffusion under irradiation.[2] In this case the situation is less complex since Ge self-diffusion under irradiation is mainly affected by D_I and, accordingly, D_I is well defined. The temperature dependence of D_I determined from radiation enhanced self-diffusion at temperatures between 554 °C and 682 °C is best described by[17]

$$D_I = (5.85 \times 10^{-4})^{+0.127}_{-5.82 \times 10^{-4}} \exp\left(-\frac{(1.32 \pm 0.42)\,\text{eV}}{k_B T}\right) \text{cm}^2\text{s}^{-1}. \quad (4)$$

This equation yields $D_I = 4.8 \times 10^{-12}$ cm^2 s^{-1} for 550 °C. Provided that the kick-out reaction solely controls B diffusion in Ge under irradiation as stated by Cowern *et al.*[3] and other previous studies,[4,11,12] the data deduced for D_I from the B profile at 550 °C must be consistent to the data deduced from self-diffusion under irradiation. However, the simulation of radiation enhanced B diffusion at 550 °C yields $D_I = 4 \times 10^{-16}$ cm^2 s^{-1}. This value deviates four orders of magnitude from the self-diffusion study. This strong inconsistency to self-diffusion demonstrates that the B profiles established under concurrent annealing and irradiation are

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not affected by the kick-out mechanism alone. Additional defect reactions must be involved that lead to the characteristic exponential B profiles under irradiation. Although experimental B profiles are well reproduced by the kick-out and g/λ model, the kick-out model fails because the results are in conflict with the D_I data obtained from radiation enhanced Ge self-diffusion. It is this inconsistency that disproves the existence of different forms of Ge self-interstitials deduced from the g/λ analysis of radiation enhanced B diffusion.

Finally, we would like to comment on the prefactor determined by Schneider *et al.*[2] from the temperature dependence of C^{eq}_{BI}. Schneider *et al.* assumed the data reported by Uppal *et al.*[16] for the B diffusion coefficient D_B. This diffusion coefficient was assigned to $D_B = C^{eq}_{BI} D_{BI}/C^{eq}_{B_s}$ although a direct proof is still lacking. This and the fact that the data determined for C^{eq}_{BI} compared to conventional diffusion anneals possess significant uncertainties in the temperature established during irradiation, a formation entropy of BI can be deduced within the limits of the experimental accuracy that led to positive migration entropies for BI. Moreover, the D_I data determined from Ge self-diffusion under irradiation are scattering also due to the difficulties to accurately determine the temperature during irradiation. Reevaluation of the data from Schneider[17] reveals a self-interstitial migration enthalpy of $H^m_I = (1.32 \pm 0.42)$ eV (see Eq. (3)). This migration enthalpy is in good agreement with the theoretical prediction of $H^m_{I^{2+}} = 1.2$ eV for the migration enthalpy of doubly positively charge I^{2+} in Ge.[18] Experiments of Haesslein *et al.*[19] support the positive charge state of I in Ge. A migration energy above 1 eV for the self-interstitial does not prove the concept of an amorphous like I-structure. This concept is also not supported by the theoretical calculations reported by Carvalho *et al.*[18]

In conclusion, simulation of B diffusion in Ge under irradiation was performed on the basis of the full differential equation system that considers B diffusion via the kick-out mechanism and the production and annihilation of vacancies and self-interstitials. Although the simulations can confirm the exponential shape of B profiles and thus seem to support the g/λ analysis, the data deduced for the diffusion coefficient D_I of self-interstitials deviate several orders of magnitude from self-diffusion experiments under irradiation. This reveals that additional defect reactions beside the kick-out reaction are involved in B diffusion under irradiation. Consequently, the conclusions drawn by Cowern *et al.*[3] on the basis of the g/λ approach are not valid and the exponential B profiles reported in the literature[3,4,11,12] do not support a different character of self-interstitials in Ge at low and high temperatures. The simulations based on the full differential equation system demonstrate that B diffusion under irradiation is mainly controlled by D_{BI} and D_I. This is also not evident in the case of the g/λ analysis, which represents an approximated solution of the kick-out diffusion equations. Our reply to the comment of Cowern *et al.* also demonstrates that simulations based on the full differential equation system are preferable to model the complex diffusion behavior under concurrent annealing and irradiation. With today's personal computers or laptops, the underlying system of coupled differential equations are solved numerically within a few seconds. These calculations are computationally inexpensive. In addition, the modeling of dopant diffusion under irradiation must be consistent to self-diffusion under irradiation. Such consistency checks are essential to derive a consistent picture on the properties of native point defects and their interaction with dopants in semiconductors. This is lacking in the analysis of Cowern *et al.*[3] and leads to the erroneous interpretation of B diffusion in Ge under irradiation. The interpretation of self- and dopant diffusion in Ge under concurrent annealing and irradiation given by Schneider *et al.*[2] is fully consistent with respect to the properties of the native point defects involved in self- and dopant diffusion. This requirement led Schneider *et al.* to propose the existence of immobile B clusters. Although a direct proof of their existence by means of, e.g., high resolution transmission electron microscopy is still lacking, the analogy to the case of B diffusion in Si under irradiation is striking and shows that the formation of immobile B defect clusters in Ge under irradiation is very likely.

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