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Evidence for diffusion-limited kinetics of ion-beam-induced epitaxial crystallization in silicon

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The theoretical predictions of point defect related models for ion-beam-induced epitaxial crystallization (IBIEC) with reaction- or diffusion-limited kinetics are compared to each other and with experimental results. It is shown that the diffusion model provides the correct nuclear energy deposition and dose rate dependence of the IBIEC rate, whereas striking differences are observed for the reaction model. A compilation of several experimental data sets indicates a uniform diffusion regime for IBIEC in a wide range of defect generation rates. In this range the IBIEC rate can be estimated by a simple formula. The expected layer thickness dependence of the IBIEC rate is verified by *in situ* time-resolved reflectivity measurements. From these results it can be concluded that point defects originating from both the amorphous and the crystalline side contribute to IBIEC. It is speculated that divacancies are the defects which diffuse and stimulate the recrystallization at the amorphous/crystalline interface.

I. INTRODUCTION

Ion irradiation through an amorphous/crystalline (a/c) interface may stimulate solid phase epitaxy or layer-by-layer amorphization in dependence of temperature and ion beam parameters.[1] From a viewpoint of application the ion-beam-induced epitaxial crystallization (IBIEC) is of interest because it occurs at temperatures much lower than necessary for the thermally induced crystallization and works also for layers deposited on substrates with contaminated surfaces.[1] Further unique features of this process are the possibilities to control precisely the lateral and vertical extent of recrystallization and to switch from IBIEC to ion-beam-induced interfacial amorphization (IBIIA). Partly, the crystal quality and dopant activation in IBIEC regrown compound semiconductors is better than obtained in thermal recrystallization. Until now IBIEC has been found in Ge,[2] Si,[3] $Ge_{1-x}Si_x$,[4] GaAs,[5] InP,[6] $NiSi_2$,[6] $CoSi_2$,[6] Al_2O_3,[7] BP,[8] and SiC.[9,10] However, it can be assumed that IBIEC is possible in many more solids.

In the case of Si a lot of experimental data were accumulated during the past years, which describe the IBIEC/IBIIA growth rate as function of deposited energy, ion dose, dose rate, temperature, frequency of pulsed beams, substrate orientation, and dopant concentration.[1,11,12] Several phenomelogical models[11,13–21] were developed which explain some aspects of the complex IBIEC/IBIIA kinetics. However, there is no model until now which provides a complete picture of the IBIEC/IBIIA process.

In most models point defects athermally produced in displacement collisions are assumed to be responsible for the low temperature recrystallization under ion beam irradiation.[12–15,17–19] With respect to the limiting process of the IBIEC kinetics these models can be divided into two groups. The first one is based on the assumption that point defect diffusion towards the a/c interface is the limiting step in the IBIEC process. Alternatively, point defect reaction at the a/c interface is supposed to govern the IBIEC kinetics. In the latter category the model of Jackson[14] is widely accepted because it was the first one which provides a quantitative estimation of the IBIEC process and fits some experimental results surprisingly well. However, a thorough analysis of this model reveals some substantial discrepancies between theoretical predictions and experimental results, which give rise to doubts on the validity of this reaction limited model of IBIEC.

In this article it is shown that the point defect diffusion model[15] provides a much better agreement with experimental data than the model of Jackson.[14] After presenting the theoretical framework of both models, the results are compared with data from the literature and our own experiments. In particular, the IBIEC rate is considered as function of energy deposited into nuclear collision, dose rate, point defect generation rate, temperature, and amorphous layer thickness. To simplify matters and due to the lack of sufficient information about IBIIA kinetics the investigation is restricted to IBIEC and the IBIEC/IBIIA transition region only.

II. IBIEC MODELS

A. Definitions and basic ideas

In general, the recrystallization rate

$$r = dV/dt \tag{1}$$

is defined as the volume (V) increase of crystalline material in an amorphous matrix with time (t). During solid phase epitaxy (SPE) the crystalline volume increases as the volume of an amorphous surface layer decreases by parallel layer shrinkage, i.e., the planar a/c interface (of constant area F) moves towards the surface. The recrystallization kinetics of SPE is usually characterized by the interface velocity

$$v = dx/dt = r/F, \tag{2}$$

where x is the regrowth distance.

In the case of IBIEC it is more convenient to consider a regrowth rate

$$r_\Phi = dx/d\Phi \tag{3}$$

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that relates the interface movement to the dose of ion irradiation Φ. This so called IBIEC rate is the volume recrystallized by one ion impact. Applying the definition of the ion dose rate

$$j = d\Phi/dt, \tag{4}$$

we obtain from Eqs. (1)–(3) the relations between IBIEC rate, interface velocity, and recrystallization rate, respectively, as

$$r_\Phi = v/j = r/(Fj). \tag{5}$$

Sometimes, the regrown thickness is normalized to the energy transferred into elastic collision to one lattice atom or, via the Kinchin-Pease-formula,[22] to the number of displacements per atom (dpa) at the a/c interface. These normalized regrowth rates are given by

$$r_{\mathrm{Sn}} = r_\Phi n_{\mathrm{Si}}/S_n \tag{6}$$

and

$$r_{\mathrm{dpa}} = r_{\mathrm{Sn}} 2E_d, \tag{7}$$

with n_{Si} being the atomic density of silicon, S_n being the nuclear deposited energy per depth, and E_d the displacement energy.

These somewhat trivial relations had to be shown here, as different quantities are used in literature to characterize the IBIEC kinetics and the terminology used there is not unambiguous. For example Jackson[14] denotes the interface velocity as rate of motion. In particular with respect to the dose rate and energy dependence of the IBIEC process, one has to carefully distinguish between IBIEC rate, interface velocity, and normalized regrowth rate.

In the point defect related models of IBIEC it is supposed that simple point defects or clusters of them produced in the collision cascades should mediate the recrystallization process at the a/c interface. It is not yet clear of what type these defects are and whether they are consumed by the recrystallization or act as "catalyst" only. One idea is that vacancy type defects at the interface lower the kinetic barrier for atomic jumps to crystalline sites. Because of the lack of detailed knowledge about the atomic transport and rearrangement mechanisms in the interface region one should try to model the recrystallization process in the most simple manner, namely, by assuming that each of these "unknown" defects transforms a microscopic volume V_c from the amorphous to the crystalline state. Therefore, the crystallization rate can be written according to formula (1) as

$$r = V_c dN_c/dt = V_c F \lambda \; dn_c/dt, \tag{8}$$

where N_c is the number and n_c the corresponding density of the elementary crystallization events, respectively, mediated by one defect at the a/c interface. The interface thickness λ is assumed to be in the order of magnitude of the atomic distance. In order to calculate the crystallization rate we must first determine how the elementary crystallization events are linked to the defect concentration in the a/c interface region. It will be shown below that there are two ways to do this in dependence on what is assumed to be the limiting process for the recrystallization—the diffusion of defects from the bulk material towards the a/c interface or the crystallization reaction of the defects at the a/c interface. Secondly, a proper balance equation must be formulated which describes the interplay between defect generation by ion irradiation and defect annihilation in the bulk.

The defect generation rate can be expressed as

$$G = Zj, \tag{9}$$

where Z is the number of atomic displacements (often interpreted as vacancies) per unit depth generated by one ion, which can be calculated, e.g., with the TRIM code,[23] and j is the ion dose rate. It should be noted that according to the Kinchin-Pease-theory[22] Z is proportional to the energy deposited into nuclear collisions S_n.

The defect annihilation term may be generally modeled by

$$A = c_A n^m, \tag{10}$$

with c_A being a rate constant, n the defect density, and m the order of defect annihilation. For example, $m=2$ means pairwise defect annihilation. According to the rate theory the rate constant c_A depends on temperature T via the Arrhenius expression

$$c_A = c_{A0} \exp(-E_A/kT), \tag{11}$$

with the activation energy E_A for the annihilation process.

Notice that the term "annihilation" may also mean that active defects are converted to other types of defects, which do not contribute to recrystallization.

B. The model of reaction limited IBIEC

Let us assume that the IBIEC process is limited by the crystallization reaction itself, i.e., the atomic rearrangement processes at the a/c interface are slow compared to the point defect diffusion from the bulk material. In this case the number of the elementary recrystallization events is directly proportional to the defect density at the interface. So, we may substitute

$$dn_c/dt = c_c n \tag{12}$$

in Eq. (8). The rate constant of the recrystallization process c_c has a temperature dependence analogous to Eq. (11) with an activation energy of E_C and is the inverse of the frequently used hopping time τ_c.[14]

The steady state concentration of defects at the interface n can be calculated by the balance equation

$$dn/dt = G - A = 0. \tag{13}$$

Inserting the expressions from Eqs. (9), (10) into Eq. (13) we obtain

$$n = (Zj/c_A)^{1/m} \tag{14}$$

and with Eqs. (12), (11), (8), (5)

$$r_\phi = c' Z^{1/m} j^{(1/m)-1} \exp(-E'/kT). \tag{15}$$

In Eq. (15) the IBIEC rate is given as function of quantities, which are usually varied in IBIEC experiments—linear density of atomic displacements Z, ion dose rate j, and temperature T, respectively. The constants are summarized in the

factor c'. The activation energy E' of the IBIEC process is a combination of both the activation energy of recrystallization and defect annealing:

$$E' = E_C - E_A/m. \quad (16)$$

In numerous experiments it was found that the IBIEC rate depends almost linearly on the energy deposited in nuclear collisions.[1,31,32] We see from Eq. (15) that this behavior may only be predicted from the model of reaction limited recrystallization if the defect annihilation would be of order $m=1$. In this case, however, the IBIEC rate would be independent of dose rate. Moreover, the IBIEC rate would not depend on temperature, if both the elementary recrystallization event and the defect annihilation are governed by atomic jumps over the same energy barrier ($E_C = E_A$), which was suggested, e.g., in the model of Jackson.[14]

The IBIEC model of Jackson is also based on the assumption of reaction limited recrystallization, i.e., the IBIEC rate is proportional to the defect density at the a/c interface. However, in contrast to the continuous defect production taken into account above, Jackson considered the discrete nature of the defect generation. According to this idea defect clouds were produced in collisional cascades caused by one energetic ion. Subsequently, the defect concentration decreases within the cascade region by pairwise defect annihilation ($m=2$) until the next ion impact into the former cascade volume.

There are two limits of the Jackson model, which should be written here in the simple and explicit notation used in this article (for comparison with quatitites originally used by Jackson[14] note the following correspondence: $R_x = v$, $\Phi = j$, $N_0 = n_i \propto Z$, $1/\tau_0 \propto j$, $1/\tau_j \propto \exp(-E_C/kT)$, $\gamma \propto Zj^{-1}\exp(-E_C/kT)$, $(a\sigma^2) = L^3$.

In dependence of the ratio of the time τ_0 between two cascades in the same area and the time τ_j for atomic jumps the IBIEC rate is given by

$$r_\phi = c_1 \ln[c_2 Z j^{-1} \exp(-E_C/kT)] \quad \text{for} \quad \tau_0/\tau_j \gg 1/(n_i L^3) \quad (17)$$

that describes the so called "single cascade regime" and

$$r_\phi = c_3 Z^{1/2} j^{-1/2} \exp(-E_C/2kT) \quad \text{for} \quad \tau_0/\tau_j \ll 1/(n_i L^3) \quad (18)$$

that describes the "cascade overlap regime." In Eqs. (17) and (18) n_i denotes the initial defect concentration in the cascade and L the critical distance for pairwise defect annihilation. The constants c summarize geometrical and time constants used in Jackson's model.

Comparing Eq. (18) with Eqs. (15), (16) we see that the "cascade overlap" limit of the Jackson model is identical to the model of reaction limited IBIEC with continuous defect generation if defects are suggested to annihilate in pairs ($m=2$) and both the atomic jumps necessary for defect annihilation and recrystallization have the same activation energy E_C. Obviously, the Jackson model provides strong deviations from the linear dependence on Z. Moreover, there is no more an Arrhenius-like temperature dependence in the case of the "single cascade" regime. Jackson explained the weak Arrhenius-like temperature dependence of IBIEC ($E_A = 0.24...0.32$ eV) (Refs. 1 and 24) by the transition from the linear temperature dependence [Eq. (17)] to the strong Arrhenius-like one for thermally stimulated SPE ($E_A = 2.7$ eV) (Ref. 25) that dominates at higher temperatures.

It should be noted here that Jackson's model includes also a steady amorphization process which competes with IBIEC. However, this amorphization process influences only the kinetics of the transition region from IBIEC to IBIIA and may be neglected at sufficiently high temperatures.

C. The diffusion-limited model

Some authors[15,26–30] assume that the IBIEC kinetics is governed by defect diffusion towards the a/c interface. According to this model each defect arriving at the interface immediately converts a microscopic volume V_C from amorphous to crystalline state and disappears due to atomic rearrangements. This means that the number of elementary crystallization events N_C in Eq. (8) is identical with the number of defects N arriving at the interface. There are some controversies whether defect fluxes coming from the amorphous, the crystalline or both sides contribute to IBIEC.[15] Therefore, it is useful to formulate first the theory for one side and to decide later what assumption is correct.

Applying the definition of the defect flux coming from the side $s=a$ (amorphous) or c (crystalline)

$$j_{D,s} = 1/F \; dN/dt, \quad (19)$$

which flows through the area F we obtain with Eqs. (5), (8) the one-side component of the IBIEC rate as

$$r_{\Phi,s} = V_C |j_{D,s}|/j. \quad (20)$$

The defect flux is given by Fick's law as

$$j_D = -D \; dn/dx, \quad (21)$$

where D is the diffusion coefficient and $n(x)$ the steady state density distribution of the defects that may be calculated by integrating the balance equation

$$D \; d^2n/dx^2 + Z(x)j - DL^{3m-5}n^m = 0 \quad (22)$$

which involves defect diffusion, generation, and annihilation. The special form of the annihilation rate constant used in Eq. (22) is the consequence of a statistical model of L^3-cell occupation with m point defects in the limit of low defect concentrations. The reoccupation time of the cells can be expressed by $\tau = L^2/D$. L is the critical distance below that defects should form immobile clusters or annihilate. It is further assumed that both defect diffusion towards the interface and defect migration necessary for the annihilation are determined by the same atomic jump processes and, therefore, may be described by one diffusion constant

$$D = D_0 \exp(-E_j/kT), \quad (23)$$

with E_j the activation energy of defect jumps, often also denoted as migration energy.

In general, Eq. (22) can be only solved numerically. However, in the case of a depth independent Z a partial integration of the nonlinear differential Eq. (22) can be carried out.[15] Assuming further that the defect concentration vanishes at both the a/c interface ($x=0$) and the surface ($x=d$)

$$n(0)=n(d)=0, \tag{24}$$

one may obtain the simple analytical expression between the defect flux at the interface and the maximum defect concentration $n_{\max}$ in the layer between the surface and the a/c interface as

$$j_D(0)=[2n_{\max}ZjD\{1-[1/(m+1)](n_{\max}/n_0)^m\}]^{1/2}, \tag{25}$$

with

$$n_0=[Zj/(DL^{3m-5})]^{1/m} \tag{26}$$

being the steady state defect concentration which would be achieved when no diffusion takes place (Eq. 22). Details of the mathematical procedure described above can be found in Ref. 15.

The usage of the maximum defect concentration is advantageous because simple limits are evident for it from Eq. (22). For layers being thicker than a critical thickness

$$d_c=(8)^{1/2}[D/(ZjL^5)]^{(m-1)/2m}L \tag{27}$$

the maximum defect concentration is mainly determined by the defect annihilation, i.e., the diffusion term may be neglected in Eq. (22). In this way we obtain

$$n_{\max}=n_0 \quad \text{for } d\gg d_c. \tag{28}$$

In the opposite case where most of the defects disappear by diffusion towards the interface the maximum defect density is

$$n_{\max}=(1/8)[Zjd^2/D] \quad \text{for } d\ll d_c, \tag{29}$$

which may be calculated after neglecting the defect annihilation term in Eq. (22). Note that $n_{\max}\ll n_0$ for $d\ll d_c$ and, therefore, the second term in Eq. (25) can be neglected.

Inserting these results into Eq. (25) we finally obtain the "thin" and "thick" layer limit of the one-side component of the IBIEC rate (20), respectively, as a function of implantation parameters Z, j and temperature T:

$$r_{\Phi,s}=\tfrac{1}{2}V_CZd \quad \text{for } d\ll d_C, \tag{30}$$

$$r_{\Phi,s}=C'_mV_CZ^{(m+1)/2m}j^{(1-m)/2m}\exp(-E_C/kT) \quad \text{for } d\gg d_C, \tag{31}$$

with

$$E_C=E_j/[2m/(m-1)] \tag{32}$$

and

$$C'_m=[2m/(m+1)]^{1/2}D_0^{(m-1)/2m}/L^{(3m-5)/2m}. \tag{33}$$

According to these theoretical results the "thin" layer limit of the IBIEC rate should be independent on temperature and dose rate. Further it is proportional to the linear density of the displacements per ion Z and depends on the layer thickness d. The layer thickness dependence of IBIEC, when found in experiments, would be an unambiguous confirmation of diffusion controlled IBIEC kinetics. However, the critical thickness d_C could be much smaller than the layer thicknesses investigated or resolved in usual IBIEC experiments. It should be emphasized, that in the case of defect fluxes coming from both the amorphous and the crystalline side, both layers must be thinner than their corresponding critical layer thicknesses in order to obtain the pure "thin" layer limit. Otherwise, the functional dependence is given by the sum of Eqs. (30) and (31). This should be the normal case in IBIEC experiments because only the amorphous layer thickness is small and decreases during ion irradiation, whereas the crystalline layer is as thick as the substrate.

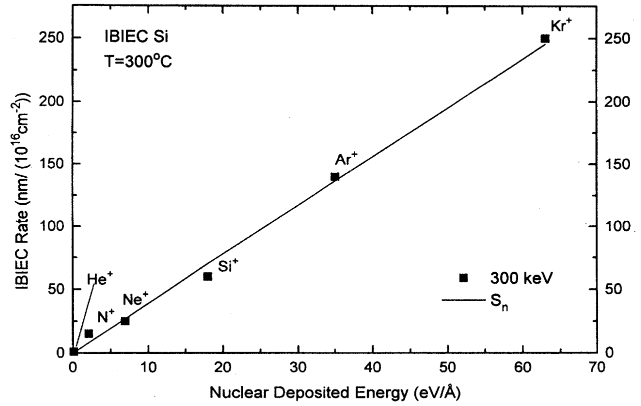

FIG. 1. IBIEC rate in dependence on the nuclear deposited energy. Experimental results after Linnros *et al.*[31] The seemingly linear energy dependence is indicated by the straight line.

Since there is no evidence for a layer thickness dependence of the regrowth rate in most experiments the "thick" layer limit should describe the "normal" IBIEC kinetics within the framework of the diffusion model. As can be seen from Eqs. (31) and (32) a linear defect annihilation process would lead to a dose rate and temperature independent IBIEC rate, which is in contradiction to experimental results. For pairwise defect annihilation we obtain

$$r_\Phi=C'_2V_CZ^{3/4}j^{-1/4}\exp(-[E_j/4]/kT) \quad \text{for } d\gg d_C. \tag{34}$$

III. COMPARISON OF EXPERIMENTAL RESULTS WITH MODEL PREDICTIONS

A. Energy dependence

Previous experiments on IBIEC have shown that at constant ion energy the IBIEC rate increases with ion mass[31] (see Fig. 1) and decreases for same ion species with increasing ion energy,[32] respectively. In both cases it was found that the IBIEC rate is, within the experimental errors, directly proportional to the part of energy deposited into nuclear collision S_n at the a/c interface and, therefore, to the displacement density Z. Numerous experiments carried out later have seemingly confirmed this obvious linear dependence on S_n. However, according to the point defect models presented above such a linear behavior is expected only for a linear defect annihilation process, where no dose rate or temperature dependence exists [Eqs. (15), (31)].

For pairwise defect annihilation the reaction model predicts a square root dependence or even a much weaker logarithmic behavior in the case of Jackson's single cascade model [Eqs. (17), (18)]. This is in striking contradiction to

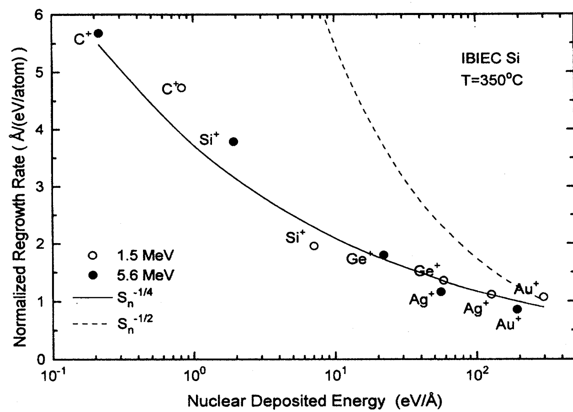

FIG. 2. Normalized regrowth rate in dependence on nuclear deposited energy S_n. Experimental results after Kinomura *et al.*[33] The theoretical predictions of the diffusion and the Jackson model are given by the solid and the broken line, respectively.

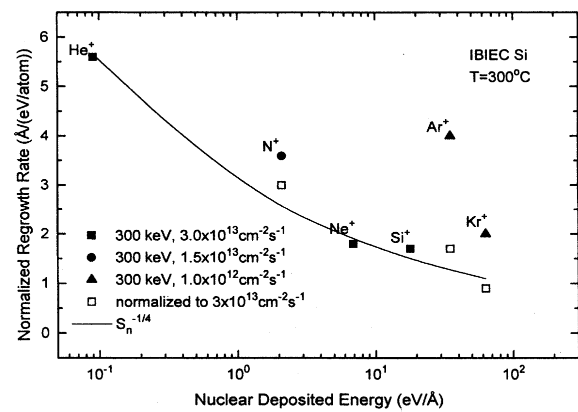

FIG. 3. Normalized regrowth rate in dependence on nuclear deposited energy S_n. Experimental results after Linnros *et al.*[31] Note that these are the same as presented in Fig. 1. Empty squares are experimental results normalized to the dose rate of 3×10^{13} $cm^{-2}\,s^{-1}$. The theoretical prediction of the diffusion model is given by the solid line.

the experimental findings. Although the diffusion model provides a sublinear dependence [$\propto Z^{3/4} \propto S_n^{3/4}$; see Eq. (34)] on the nuclear energy deposition, too, the deviation from the linearity is much smaller than in the reaction model. It is very difficult to prove such a small difference in experiments because of implantation and analysis inaccuracies. However, a simple mathematical trick recently applied by Kinomura *et al.*[33] can help to render the deviation from linearity more evident. The IBIEC rate divided by the deposited nuclear energy, i.e., the normalized regrowth rate [Eq. (6)], should not depend on the deposited nuclear energy, if the IBIEC rate is exactly proportional to the displacements. Otherwise, a clear functional dependence should be found.

Kinomura *et al.*[33] have investigated the IBIEC process for several ions at energies of 1.5 and 5.6 MeV, respectively. In Fig. 2 the experimental results of the normalized regrowth rate are represented as function of the nuclear energy deposition. It can be clearly seen that the normalized regrowth rate is not constant, but follows very well a $S_n^{-1/4}$ dependence as predicted by the diffusion model with pairwise defect annihilation. For comparison the curve expected from the cascade overlap regime of the Jackson model ($S_n^{-1/2}$) is drawn, too.

Interestingly, the previous results of Linnros *et al.*[31] (Fig. 1) on the energy dependence of the IBIEC rate show also strong deviations from the linear behavior, if they are represented in a normalized plot (Fig. 3). As can be seen in Fig. 3 the results for He, Ne, and Si irradiation, which was carried out at the same dose rate, can be fitted again by an $S_n^{-1/4}$ curve. However, the regrowth rates obtained from ion irradiation at lower dose rates are higher as expected. Considering the dose rate dependence predicted by the diffusion model with pairwise defect annihilation [Eq. (34)], the regrowth rates measured at lower dose rates j can be normalized to $j_0=3\times10^{13}$ $cm^{-2}\,s^{-1}$ by multiplying them with $(j/j_0)^{1/4}$. In this way an excellent agreement between theoretical prediction and experimental results is obtained.

Finally, it should be noted that the deviation from the linear energy dependence of the IBIEC rate becomes evident in experimental studies only if the nuclear deposited energy is varied over orders of magnitudes and all other implantation parameters are kept constant.

B. Dose rate dependence

The dose rate dependence of IBIEC was investigated by several authors. Williams *et al.*[32] studied the IBIEC process with 80 keV He^+ ion irradiation at 440 °C (Fig. 4). They found that the IBIEC rate decreases with increasing dose rate. Similar results were obtained by Linnros and Holmen[34] for 300 keV inert gas ion implantation in silicon and silicon-on-sapphire material at different temperatures. In Fig. 5 the results obtained at 400 °C are shown. However, only three different dose rates were used in these previous studies. This is clearly insufficient for a reliable comparison with theory. Therefore, a more comprehensive and precise study of the dose rate dependence of IBIEC stimulated with 300 keV Si^+ at 400 °C was performed by Heera *et al.*[26] (Fig. 6). In all these experiments the measured data are in good agreement

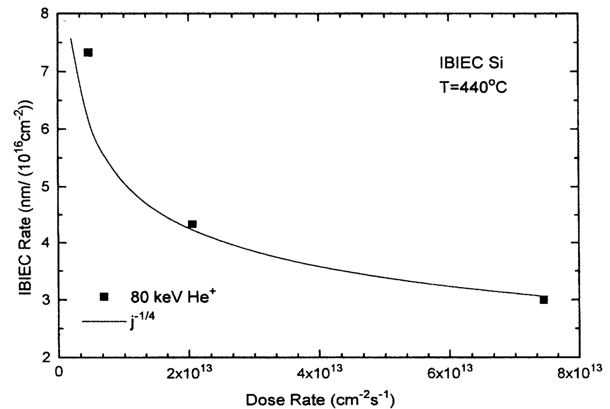

FIG. 4. IBIEC rate in dependence on the dose rate. Experimental results after Williams *et al.*[32] The theoretical predictions of the diffusion model are given by the solid line.

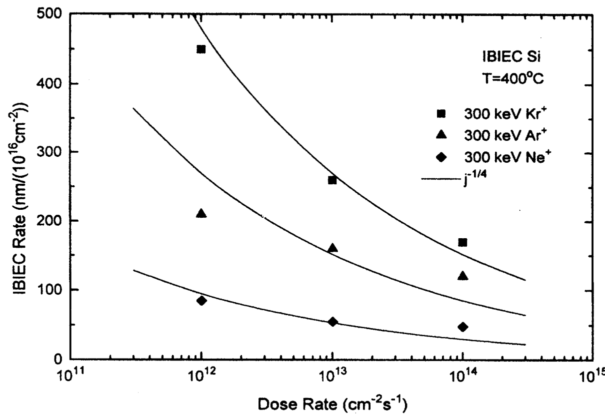

FIG. 5. IBIEC rate in dependence on the dose rate at 400 °C. Experimental results after Linnros *et al.*[34] The theoretical predictions of the diffusion model are given by the solid lines.

with the $j^{-1/4}$ behavior (solid lines in Figs. 4–6), which is expected from the diffusion model with pairwise defect annihilation [Eq. (34)]. Note, that it is not possible to fit these experimental results with the Jackson model, which predicts a logarithmic dependence for the single cascade regime [Eq. (17)] and a square root dependence for the cascade overlap regime [Eq. (18)], respectively.

Deviations from the $j^{-1/4}$ dependence were found near the transition temperature from IBIEC to IBIIA.[34,35] It was demonstrated that the critical temperature for this growth reversion increases with increasing nuclear energy deposition and/or dose rate.[35] According to the data given in Ref. 35 the following empirical relation exists between the critical temperature, displacement rate, and dose rate

$$\exp(-1.2 \text{ eV}/kT_R)=3.5\times10^{-42} \text{ cm}^4\text{ s } Z^2 j. \quad (35)$$

C. Temperature dependence

Several experimental studies[1,24] have demonstrated that the activation energy of IBIEC is between 0.24 and 0.32 eV for $T_R<T\leqslant 400$ °C and then increases to 2.7 eV for thermally stimulated solid phase epitaxy ($T>550$ °C). These findings were explained with point defects necessary for recrystallization. It was assumed that in the case of thermally induced SPE the activation energy is the sum of the energy of defect formation and migration, whereas for IBIEC only the defect migration energy is necessary because the defects are formed in a ballistic way. The activation energy of IBIEC has been often attributed to the vacancy migration energy, because the migration energy of neutral vacancies in silicon is 0.33 eV.[41] It was shown by the author in a previous paper[15] that this idea cannot be correct. According to the diffusion and also to the reaction model (overlap regime) the IBIEC activation energy is determined by both the activation energy of the elementary atomic jump processes and the order of defect annihilation. Only a fraction of the activation energy of the elementary process is predicted to be the IBIEC activation energy for higher orders of defect annihilation [see Eqs. (15), (16) and (31), (32)].

Under certain experimental conditions the a/c interface does not move during ion irradiation, because of the balance between IBIEC and IBIIA

$$r_{\Phi,\text{IBIEC}}-r_{\Phi,\text{IBIIA}}=0. \quad (36)$$

Following Jackson's idea to model the amorphization by a temperature and dose rate independent rate,[14] we obtain with expression (34) the relationship between the critical temperature T_R and the irradiation parameters j,Z as

$$\exp(-E_j/kT_R)=(r_{\Phi,\text{IBIIA}}/V_C C_2')^4 Z^{-3} j. \quad (37)$$

According to the diffusion model with pairwise defect annihilation the activation energy for defect jumps E_j should be four times the IBIEC activation energy [Eq. (34)], i.e.,

$$E_j=0.96\ldots1.28 \text{ eV}. \quad (38)$$

This is in excellent agreement with the experimental results of Linnros *et al.*[35] [Eq. (35)], who found $E_j=1.2$ eV.

In order to obtain the same dependence on the displacement density as in Eq. (35) the IBIIA rate must be proportional to $Z^{5/4}$. This is a reasonable assumption. Recent investigations on IBIIA indicate a linear nuclear energy dependence, which is very close to $Z^{5/4}$.[12] It should be noted, however, that there is also a temperature and dose rate dependence of IBIIA and, therefore, the considerations above could be too rough.

D. Defect generation rate dependence

According to definitions [(5),(9)] and the model predictions [(15),(31)] the interface velocity can be represented as function of the defect generation rate and temperature

$$v=v_0\exp(-E/kT)=cG^\alpha\exp(-E/kT), \quad (39)$$

with

$$\alpha=1/m$$

for the reaction model (cascade overlap) and

$$\alpha=(m+1)/2m$$

for the diffusion model (thick layer), respectively.

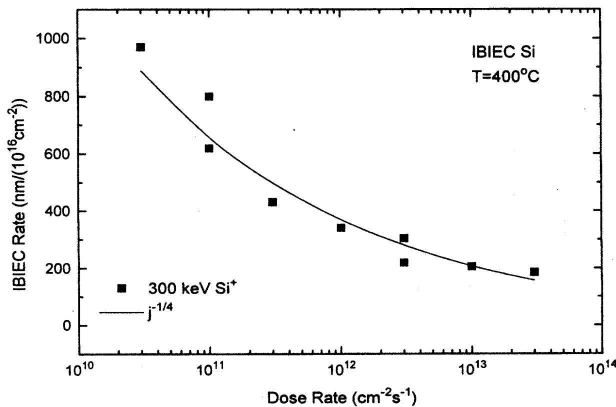

FIG. 6. IBIEC rate in dependence on the dose rate. Experimental results after Heera *et al.*[26] The theoretical predictions of the diffusion model are given by the solid line.

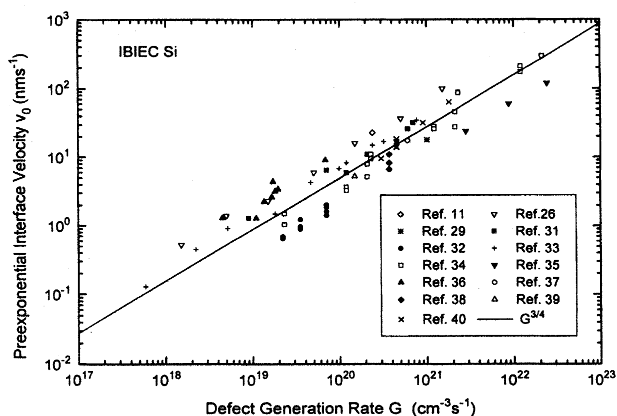

FIG. 7. Preexponential factor of the a/c-interface velocity during IBIEC as function of the defect generation rate. The solid line is the best fit of the experimental results [Eq. (40)]. Note that the figure compiles numerous experimental results obtained at quite different experimental conditions.

A double logarithmic plot of v_0 over G gives a straight line with slope α. We have analyzed a big number of IBIEC data taken from literature[11,26,31–40] in order to look for such a dependency and to determine α. In order to avoid complications by the IBIIA process only IBIEC results are considered, which were obtained at least 40 K above the critical temperature [Eq. (35)]. As shown in Fig. 7 there is a good correlation of the data points with a straight line of slope 3/4. This is exactly the result expected from the point diffusion model with pairwise defect annihilation in the bulk. It should be emphasized that there is a uniform IBIEC regime in the wide range of defect generation rates from 6×10^{17} to 2×10^{22} $cm^{-3}\,s^{-1}$. The scattering of the data points could be caused by both experimental inaccuracies (e.g., target temperature) and different material quality (e.g., defect and doping level).

An approximate value for the constant in Eq. (39) can be extracted from the experimental data plotted in Fig. 7. Together with the known slope and the activation energy a simple formula for a rough quantitative estimation of the IBIEC rate can be derived, which works for a very wide range of experimental conditions and is very useful when planning IBIEC experiments:

$$r_\Phi = 5\times10^{-22}\ \mathrm{cm}^{13/4}\,\mathrm{s}^{-1/4}\,Z^{3/4} j^{-1/4}\exp(-0.3\ \mathrm{eV}/kT)$$

$$\text{for } T_R < T \leq 400\ ^\circ\mathrm{C} \text{ and } d > d_C. \quad (40)$$

E. Layer thickness effects

The most striking feature of the diffusion model compared to the reaction model is the existence of a layer thickness dependence of the IBIEC rate [Eq. (30)]. In order to find out such thickness effects the IBIEC rate must continuously be recorded up the disappearance of the amorphous layer with a high degree of precision. This is only possible by *in situ* measurement methods as, e.g., the time resolved reflectivity (TRR) analysis method.[1,25] According to Eq. (27) the critical thickness below which the thickness effect should appear is itself a function of temperature and ion beam parameters. Proper experimental conditions should be chosen

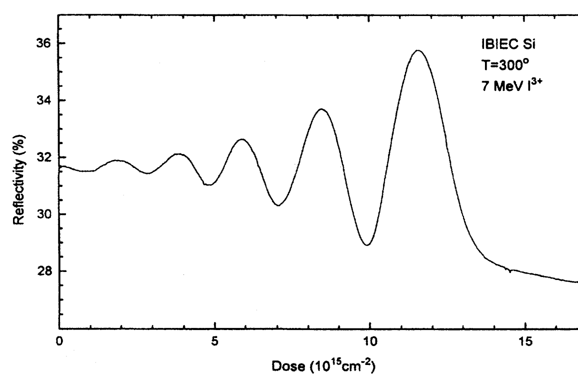

FIG. 8. Time resolved reflectivity trace measured during IBIEC as function of fluence.[41] The peak-to-valley distance corresponds to a regrown thickness of 36.2 nm.

to maximize the critical thickness. This means that the temperature should be as high as possible whereas the dose rate and the displacement density should be kept as low as possible. Priolo *et al.*[1] investigated the depth dependence of the IBIEC process for 600 keV Kr^+ ions at 350 °C by TRR measurements. Indeed, the IBIEC rate was found to decrease with shrinking amorphous layer thickness. However, this effect was attributed to the decline of the nuclear energy deposition profile in this depth range. Results on IBIEC rate decrease with increasing ion fluence during 300 keV ion irradiation[31] can be interpreted in a similar manner. Therefore the depth dependence of IBIEC were studied by TRR measurements with ions of several MeV kinetic energy.[42] In this case the nuclear deposited energy is almost constant in the near surface region. In Fig. 8 the measured reflectivity during IBIEC with 7 MeV I^{3+} ion irradiation at 300 °C is shown in dependence on the ion dose. In this curve every peak-to-valley distance corresponds to a regrown layer thickness of 36.2 nm. So it is possible to represent the IBIEC rate as function of the amorphous layer thickness (Fig. 9). It is obvious that after an initial stage with an almost constant regrowth rate of about 36 nm/10^{15} cm^{-2} the IBIEC rate decreases with shrinking amorphous layer thickness. The depth

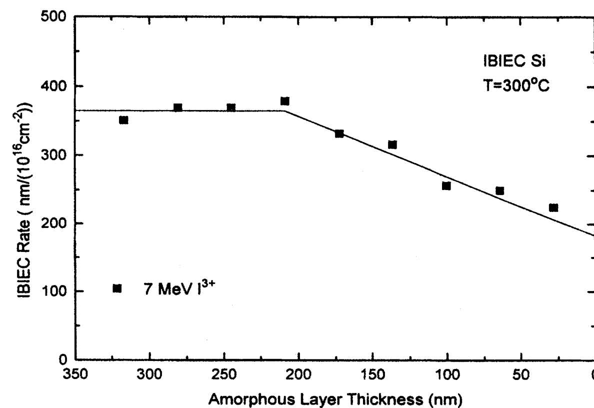

FIG. 9. IBIEC rate as function of the remaining amorphous layer thickness as calculated from the TRR trace in Fig. 8. The solid line is only to guide the eye.

dependence of the IBIEC rate for amorphous layer thicknesses below 200 nm cannot be explained by a profile of nuclear energy deposition. Assuming a linear depth dependence as theoretically predicted from Eq. (30) the IBIEC rate for zero thickness can be extrapolated to be about 18 nm/10^{15} cm^{-2}, which is just the half of the initial one. Because the IBIEC rate does not vanish it is clear that defects coming from the crystalline side participate in the recrystallization process. Moreover, the bisection of the IBIEC rate implies that defects coming from the amorphous layer should contribute to IBIEC with the same efficiency. Previous results on the effect of channeled ions on the IBIEC rate[28] can be interpreted in an analogous way. The IBIEC rate is reduced to one-half of its initial value when the ions enter the target in channel direction and, therefore, do not produce atomic displacements in the crystalline layer.

Further evidence for defect diffusion during IBIEC is given by recent experiments from Stepina *et al.*[29] who used low energy ions which do not arrive at the *a/c* interface, but nevertheless stimulated the regrowth process.

IV. CONCLUSIONS

It was demonstrated in this article that the defect diffusion model with pairwise defect annihilation in the bulk describes the IBIEC rate as function of nuclear deposited energy and dose rate in good agreement with experimental results, whereas a thorough analysis of the reaction limited model of IBIEC reveals some striking differences between theoretical predictions and experimental findings. Moreover, the diffusion model is valid in a wide range of defect generation rates as indicated by a compilation of many experimental IBIEC data. From these data the unknown constant of the model is determined and a formula for a quantitative prediction of the IBIEC rate in silicon is derived. As a consequence of the model the migration energy of the defects which induce the crystallization should be four times the IBIEC activation energy, i.e., between 0.96 and 1.2 eV. From this reason single vacancies with migration energies between 0.18 and 0.33 eV cannot be the IBIEC stimulating defects. However, a possible candidate for the IBIEC stimulating defect could be the divacancy with a migration energy of about 1.3 eV.[41] This assumption is quite reasonable because several investigations have shown that divacancies are the most available vacancy-type defect in silicon after ion implantation at room temperature.[43] Moreover, a recent positron annihilation study has demonstrated that tri- and tetravacancies are formed during IBIEC.[44] Therefore, it can be speculated that the pairing process considered in the diffusion model is the agglomeration of divacancies to tetravacancies.

Further evidence for the diffusion model of IBIEC was provided by the experimentally proved decrease of the IBIEC rate with shrinking amorphous layer thickness. Because the IBIEC rate extrapolated to zero thickness is about one-half of the initial rate it can be concluded that defects coming from both the amorphous and the crystalline side contribute to IBIEC to the same magnitude. The existence of vacancy-type defects in ion irradiated amorphous silicon has recently been shown by van den Hoven *et al.*[45]

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