

Damage buildup in Si under bombardment with MeV heavy atomic and molecular ions

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Damage buildup in Si under bombardment with MeV heavy atomic and molecular ions

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Accumulation of structural disorder in Si bombarded at $-196\text{ }^{\circ}\text{C}$ with $0.5\text{ MeV }^{209}\text{Bi}_1$ and $1\text{ MeV }^{209}\text{Bi}_2$ ions (the so-called molecular effect) is studied by Rutherford backscattering/channeling spectrometry. Results show that the damage buildup is sigmoidal even for such heavy-ion bombardment at liquid nitrogen temperature. This strongly suggests that, for the implant conditions of this study, the buildup of lattice damage cannot be considered as an accumulation of completely disordered regions. Instead, damage-dose curves are well described by a cascade-overlap model modified to take into account a catastrophic collapse of incompletely disordered regions into an amorphous phase after damage reaches some critical level. Results also show that Bi_2 ions produce more lattice damage than Bi_1 ions implanted to the same dose. The ratio of lattice disorder produced by Bi_2 and Bi_1 ions is 1.7 near the surface, decreases with depth, and finally becomes close to unity in the bulk defect peak region. Parameters of collision cascades obtained using ballistic calculations are in good agreement with experimental data. The molecular effect is attributed to a spatial overlap of (relatively dense) collision subcascades, which gives rise to (i) nonlinear energy spike processes and/or (ii) an increase in the defect clustering efficiency with an effective increase in the density of ion-beam-generated defects. © 2001 American Institute of Physics. [DOI: 10.1063/1.1404426]

I. INTRODUCTION

Over the past two decades, studies of semiconductors exposed to bombardment with MeV ions have received increasing interest. Such interest has been stimulated by the fact that implantation with MeV ions offers considerable control and flexibility over ion ranges and energy deposition over large depths. Because ion implantation always produces lattice disorder, which affects all the properties of the material, considerable research effort has been made to understand and control material properties that are modified by implantation-produced lattice damage.

One of the physically interesting ion-beam-damage effects is the so-called molecular effect (ME). It is considered that the ME takes place when the level of implantation-produced lattice disorder differs during bombardment with atomic and molecular ions of equal velocity. The ME in semiconductors has received extensive studies over the past two decades (see, for example, Refs. 1–4 and references therein). Research interest in the ME has basically been driven by the fact that ME studies are capable of providing important information on fundamental ion-beam-damage processes in semiconductors such as the amorphization behavior and the effect of the density of collision cascades on

implantation-produced lattice disorder. An understanding of these processes is often necessary if potential applications of ion implantation for device fabrication are to be fully exploited.

Often, the ratio of the levels of ion-beam-produced stable lattice damage under molecular and atomic ion bombardment regimes (γ) is used as a quantitative measure for the ME. When $\gamma \neq 1$, it is considered that the ME takes place. Most of the ME studies, using Rutherford backscattering/channeling (RBS/C) spectrometry to measure lattice disorder, have revealed that $\gamma > 1$ in the case of heavy-ion bombardment (see, for example, Refs. 1–5), with the exception of some reports on electron microscopy studies,^{6,7} which have suggested that $\gamma < 1$ in some cases.

The nature of the ME is generally explained within the framework of the nonlinear energy spike concept, such as displacement and/or thermal spikes.^{1,2} In the case of displacement spikes, it is assumed that, if the density of atomic displacements within a collision cascade exceeds some threshold value (usually ~ 10 at. %), the damaged lattice suffers a catastrophic collapse to an amorphous phase. For a thermal spike to occur, it is widely believed that the average energy density deposited into the volume of a collision cascade should be close to (or above) the average energy required for material melting (usually up to a few eV/atom). In this case, highly disordered regions are produced as a result

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of the formation and subsequent rapid quenching of pseudo-liquid (“molten”) regions within collision cascades. These displacement and thermal spike processes are nonlinear with the density of nuclear energy deposition. Hence, a spatial overlap of collision cascades (or subcascades) produced by atoms which form a molecular ion may result in an increase in the level of implantation-produced lattice disorder due to such nonlinear energy spike processes.^{1,2}

It is important to note that the ME in Si during heavy-ion bombardment has previously been studied mostly for lattice disorder integrated over the whole ion penetration depth.^{1,2} Typical energies of heavy ions in such ME studies were tens of keV. However, it is clear that, due to ion scattering, the efficiency of the spatial overlap of collision cascades produced by the atoms which form a molecular ion changes with ion penetration depth. Hence, for a better understanding of the physical processes responsible for the ME, it is important to study the ME at different depths.

We are aware of only one report for relatively heavy-ion bombardment where the ME was studied as a function of depth for Si bombarded at room temperature with Ge cluster ions with an energy of ~ 1 MeV/atom.⁵ However, the authors of Ref. 5 did not keep the value of beam flux [in atoms/(cm²s)] constant during bombardment with atomic and molecular Ge ions. As has previously been discussed in detail in Ref. 4, such a condition on beam flux is rather important in ME studies in semiconductors, particularly in the case of Si bombarded with light ions or with MeV heavy ions at room temperature when dynamic annealing processes (responsible for the flux effect) are rather pronounced. Hence, the interpretation of the ME data reported in Ref. 5 is rather complicated since contributions from both the flux effect and the ME should be taken into account.

In this article, we report on a study of disorder depth profiles in Si bombarded with MeV heavy ions (²⁰⁹Bi). In order to minimize dynamic annealing and to eliminate contribution from the flux effect, we study Si bombarded at liquid nitrogen temperature with a constant beam flux value [in Bi atoms/(cm²s)]. Results show that parameters of collision cascades obtained using ballistic calculations are in good agreement with experimental data.

II. EXPERIMENT

Phosphorus doped (100) Si wafers (5–10 Ω cm) were bombarded with 0.5 MeV ²⁰⁹Bi₁⁺ and 1 MeV ²⁰⁹Bi₂⁺ ions at -196°C over the dose range from 2×10^{12} to 1.5×10^{13} cm⁻² with a beam flux of $\sim 6 \times 10^9$ Bi atoms/(cm²s) (for both atomic and molecular ions) using an ANU 1.7 MV tandem accelerator (NEC, 5SDH-4). During implantation, samples were tilted by $\sim 7^\circ$ relative to the incident ion beam to minimize channeling. After implantation, samples were characterized *ex situ* at room temperature by RBS/C using an ANU 1.7 MV tandem accelerator (NEC, 5SDH) with 2 MeV ⁴He⁺ ions incident along the [100] direction and backscattered into a detector at 98° relative to the incident beam direction. This glancing-angle detector geometry was used to provide enhanced depth resolution for examining near-surface damage accumulation. All RBS/C spectra have been

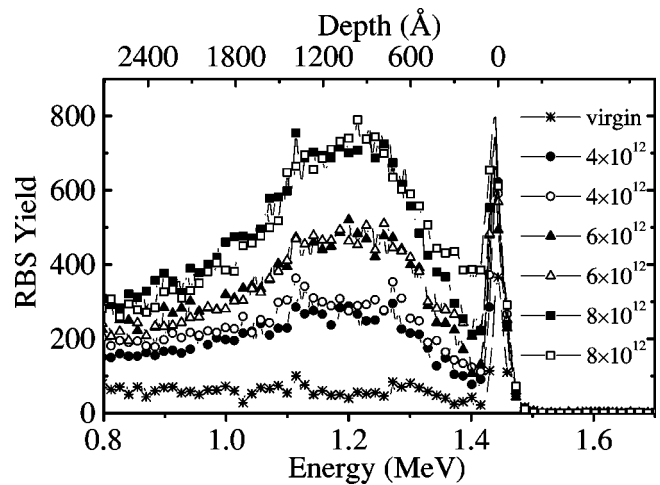


FIG. 1. RBS/C spectra showing the damage buildup for 0.5 MeV Bi₁ (closed symbols) and 1 MeV Bi₂ (open symbols) ion bombardment of Si at -196°C with a beam flux of $\sim 6 \times 10^9$ Bi atoms/(cm²s). Implantation doses (in Bi atoms/cm²) are indicated in the legend. The random yield corresponds to ~ 1980 counts.

analyzed using one of the conventional algorithms⁸ for extracting depth profiles of the effective number of scattering centers. For brevity, the number of scattering centers extracted from RBS/C data, normalized to the atomic concentration of Si, will be referred to below as “relative disorder.”

III. RESULTS AND DISCUSSION

Figure 1 shows selected RBS/C spectra which illustrate the damage buildup in Si with increasing dose of 0.5 MeV Bi₁ (closed symbols) and 1 MeV Bi₂ (open symbols) ions at -196°C . It is seen from this figure that, with increasing ion dose, implantation-produced lattice damage accumulates both in the bulk and apparently at the surface. Indeed, a relatively large surface defect peak is seen in addition to the expected bulk defect peak, which is close to the maximum of the nuclear energy loss profile. Similar strong surface disordering, observed by RBS/C, is typical for bombardment of Si with light ions at room temperature.^{9–14} However, Fig. 1 shows that a relatively large surface defect peak is also present in Si bombarded with MeV heavy ions at low temperature. Preferential surface disordering is generally attributed to the trapping of ion-beam-generated point defects by the surface or the amorphous/crystalline interface.¹⁵ However, dynamic annealing processes (i.e., defect interaction processes) in Si during ion bombardment at liquid nitrogen temperature are suppressed and long-range migration of ion-beam-generated point defects is negligible.¹⁶ Hence, other mechanisms of the formation of surface defect peaks, observed by RBS/C, should be considered, as has recently been discussed in Ref. 15. In the present article, we will not further discuss this issue but, rather, focus on the ME and the behavior of the damage buildup in the crystal bulk.

Figure 1 also shows that, in the region between surface and bulk peaks of disorder, bombardment with ²⁰⁹Bi₂ ions produces a larger level of lattice damage than irradiation with ²⁰⁹Bi₁ ions to the same dose (in Bi atoms/cm²). Hence, in the near-surface region of Si, the ME takes place (with

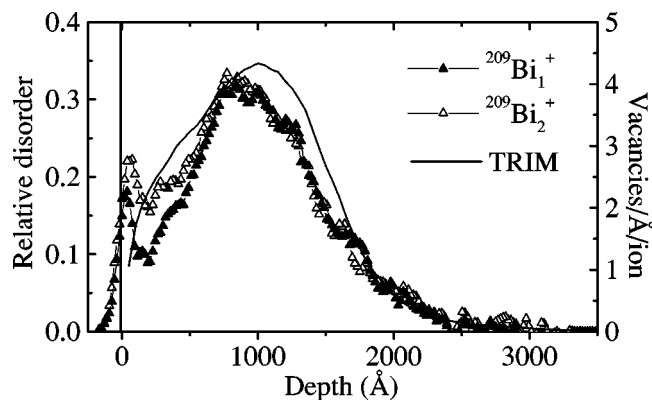


FIG. 2. Depth profiles of relative disorder in Si bombarded at -196°C with 0.5 MeV Bi_1 (closed symbols) and 1 MeV Bi_2 (open symbols) ions with a beam flux of $\sim 6 \times 10^9$ Bi atoms/($\text{cm}^2 \text{s}$) to a dose of $\sim 8 \times 10^{12}$ Bi atoms/ cm^2 . A solid line shows a depth profile of lattice vacancies generated in Si by bombardment with 0.5 MeV $^{209}\text{Bi}_1$ ions, as calculated using the TRIM code.

$\gamma > 1$). The ME has been observed for all ion doses used in this study (from 2×10^{12} up to 1.5×10^{13} Bi atoms/ cm^2). However, we will discuss ME data only for relatively low ion doses (up to 8×10^{12} cm^{-2}) when lattice disorder is significantly below the amorphization level. Indeed, as has previously been discussed in Refs. 17 and 18, the ME should be studied only in the case of relatively low levels of lattice disorder when the interaction of defects generated by an impinging ion with preexisting lattice disorder produced by previous ions can be neglected.

The ME is better illustrated by depth profiles of relative disorder extracted from RBS/C spectra. For example, Fig. 2 shows such disorder depth profiles in Si bombarded with 0.5 MeV $^{209}\text{Bi}_1$ and 1 MeV $^{209}\text{Bi}_2$ ions at -196°C to a dose of 8×10^{12} Bi atoms/ cm^2 . It is seen from Fig. 2 that the magnitude of the ME decreases as ions penetrate deeper into the Si crystal. Also shown in Fig. 2 by a solid line is a depth profile of lattice vacancies generated in Si by bombardment with 0.5 MeV $^{209}\text{Bi}_1$ ions, as calculated using the Transport of Ions in Matter (TRIM) code with a threshold displacement energy of 13 eV.¹⁹ Figure 2 shows that shapes of the defect generation function and the disorder depth profile, measured by RBS/C, are close to each other. This supports the above assertion that long-range migration of ion-beam-generated point defects in Si is negligible for the implant conditions of this study.

Figure 3 summarizes ME data, showing a depth profile of γ obtained by averaging data for depth profiles of relative disorder produced by atomic and molecular ion bombardment to different doses (2, 3, 4, 5, 6, and 8×10^{12} cm^{-2}). This figure again clearly illustrates that the magnitude of the ME decreases as ions penetrate deeper into the crystal. Such a behavior is expected. Indeed, the ME is due to a spatial overlap of collision cascades produced by atoms which form a molecular ion (and, hence, simultaneously impact the surface). Such a spatial overlap of collision cascades takes place only in the near-surface region, at the beginning of ion trajectories, before the atoms forming a molecular ion are scattered to large distances compared with the lateral size of collision cascades. This behavior of ion propagation through

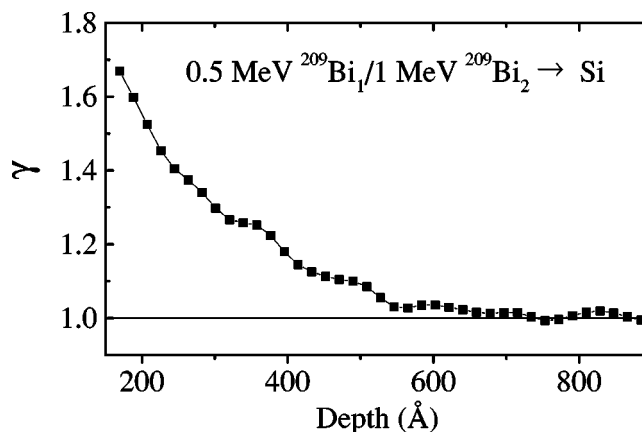


FIG. 3. Depth profile of the effectiveness of the molecular effect (γ) in Si bombarded at -196°C with 0.5 MeV Bi_1 and 1 MeV Bi_2 ions with a beam flux of $\sim 6 \times 10^9$ Bi atoms/($\text{cm}^2 \text{s}$), after averaging RBS/C data for several ion doses.

a Si crystal is illustrated in Fig. 4, which shows atomic displacements generated in Si by one randomly chosen 0.5 MeV $^{209}\text{Bi}_1$ atom, as calculated using the TRIM code.¹⁹ Note that the depth at which the Bi atom starts to significantly depart from its initial direction (see Fig. 4) correlates with the depth at which γ becomes close to unity (see Fig. 3).

It is clear that the average parameters of individual collision cascades produced by the components of a molecular ion (such as maximum relative disorder within an individual collision cascade or the depth dependence of the average distance between the centers of two collision cascades) cannot be correctly determined based on data for only one col-

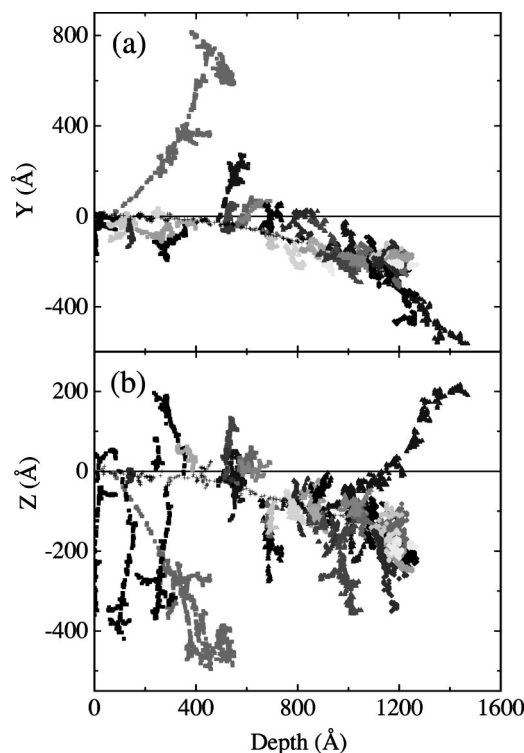


FIG. 4. Two projections of the distribution of atomic displacements generated in Si by one randomly chosen $^{209}\text{Bi}_1$ ion with an energy of 0.5 MeV, as calculated using the TRIM code.

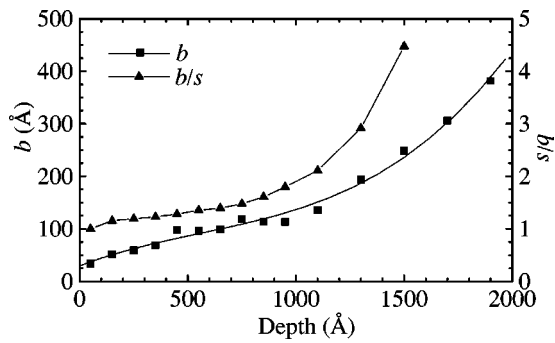


FIG. 5. Depth profiles of a half of the average distance between the centers of collision cascades (b) produced in Si by bombardment with 0.5 MeV $^{209}\text{Bi}_1$ ions. Also shown is the ratio of b to the standard deviation (s) of the lateral distribution of displacements within collision cascades.

lision cascade, such as illustrated in Fig. 4. Hence, we have performed an analysis of data obtained using the TRIM code¹⁹ for 33 different collision cascades produced in Si by 0.5 MeV $^{209}\text{Bi}_1$ ions. In such an analysis, each collision cascade was sliced by planes parallel to the sample surface. In each slice, a uniform depth distribution of atomic displacements was assumed, while the lateral distribution was assumed to be Gaussian

$$f_d = f_{dm} \exp[-(y^2 + z^2)/2s^2], \quad (1)$$

$$f_{dm} = N/(2\pi s^2 n_0 \Delta x), \quad (2)$$

where y and z are Cartesian coordinates perpendicular to x ($y=z=0$ corresponds to the cascade center at depth x); s is the standard deviation; N is the total number of displaced atoms (produced by all recoils) in the collision cascade in the slice under consideration; $\Delta x = 100 \text{ \AA}$ is the thickness of slices; and n_0 is the atomic density of Si. For each slice, f_{dm} , s , and the average distance between the centers of individual collision cascades ($2b$) were determined by averaging data for all 33 cascades.²⁰ The total number of displaced atoms (N) in the slice under consideration and the nuclear energy deposition profile were separately calculated (using the TRIM code¹⁹) for a relatively large number of ions (> 1000) to ensure good statistics. Assuming a Gaussian distribution for nuclear energy deposition in the lateral direction with the same value of s as for atomic displacements, we also calculated the maximum value of nuclear energy deposition in averaged individual collision cascades, normalized to the atomic concentration of Si (Θ_m).

Results of the above analysis are shown in Figs. 5 and 6, which illustrate depth profiles of b and b/s (Fig. 5) and profiles of f_{dm} and Θ_m (Fig. 6). It is seen from Fig. 5 that the average distance between the centers of individual collision cascades increases with depth. For depths of 500–700 Å, where γ is close to unity (see Fig. 3), $2b$ is $\sim 200 \text{ \AA}$. In this case, the ratio b/s of the average distance between the centers of individual collision cascades ($2b$) to the characteristic diameter of collision cascades ($2s$) is ~ 1.4 (see Fig. 5). Hence, results of calculations shown in Fig. 5 are in very good agreement with experimental data from Figs. 1–3.

However, results from Fig. 6 are in contrast to expectations that nonlinear energy spike processes take place within

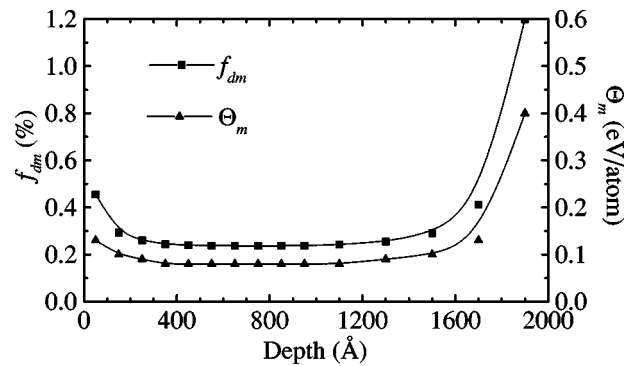


FIG. 6. Depth profiles of average relative disorder (f_{dm}) and nuclear energy loss (Θ_m) in the center of individual collision cascades produced in Si by bombardment with 0.5 MeV $^{209}\text{Bi}_1$ ions.

collision cascades produced by MeV heavy ions. Indeed, Fig. 6 shows that, for most of the ion penetration depth (except for the ion end-of-range part), f_{dm} and Θ_m are significantly lower than the threshold values for these parameters required for nonlinear energy spike processes to occur. Such threshold values for f_{dm} and Θ_m for Si are generally accepted to be $\sim 10\%$ for displacement spikes and 0.7 eV/atom for thermal spikes, respectively,^{1,2} while Fig. 6 gives $f_{dm} < 0.5\%$ and $\Theta_m < 0.2 \text{ eV/atom}$ in the near-surface region. Hence, even in the case of a complete spatial overlap of individual collision cascades produced by the atoms which form a molecular ion, the ME should not take place. This is in contrast to experimental data shown in Figs. 1–3, which clearly illustrate the ME with $\gamma > 1$. Therefore, averaging nuclear energy deposition over a relatively large volume of individual collision cascades may not be giving a true picture in the case of bombardment with MeV heavy ions, as will be discussed more fully below.

Unexpectedly low values of f_{dm} and Θ_m (see Fig. 6) also suggest that bombardment of Si with 0.5 MeV $^{209}\text{Bi}_1$ or 1 MeV $^{209}\text{Bi}_2$ ions does not produce completely disordered regions within the total volume of collision cascades. This indeed appears to be the case, as is supported by data from Fig. 7, which clearly illustrates that the curve of maximum rela-

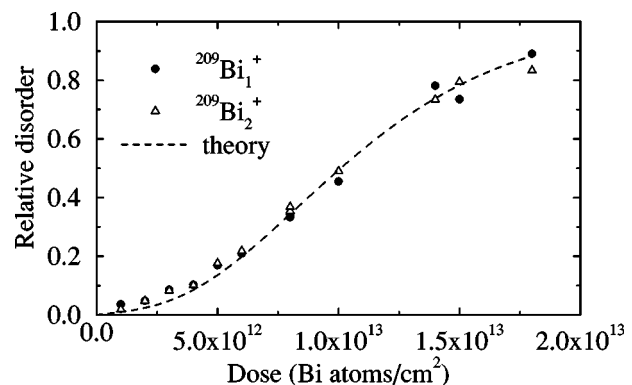


FIG. 7. Dose dependence of maximum relative disorder in the bulk defect peak for bombardment of Si at $-196 \text{ }^\circ\text{C}$ with 0.5 MeV Bi_1 (closed symbols) and 1 MeV Bi_2 (open symbols) ions with a beam flux of $\sim 6 \times 10^9 \text{ Bi atoms/(cm}^2 \text{ s)}$. Also shown by a dashed curve is a result of theoretical calculations.

tive disorder in the bulk defect peak as a function of ion dose is sigmodal. If the damage buildup proceeded via the accumulation of completely disordered zones produced by single ion impact, the gross level of lattice disorder would be an exponential function, rather than a sigmodal one (shown in Fig. 7), as we discuss more fully below.

Indeed, in the first approximation, sigmodal damage-dose curves, such as shown in Fig. 7, can be described by a model which takes into account a spatial overlap of regions with an incompletely disordered crystal structure.^{21–23} This quantitative model can be applied for the case of the damage buildup in Si during heavy-ion bombardment at liquid nitrogen temperature because the rate of dynamic annealing in this case is considerably lower than the defect production rate.¹⁶ It is usually assumed in such cascade-overlap models that an m -fold spatial overlap of incompletely disordered regions is required for complete lattice disordering. The standpoint of these models is the fact that the part of the area of a layer at specific depth taken by a spatial overlap of $(k-1)$ disordered regions can be described by the Poisson equation

$$S_k = (a\Phi)^k \exp(-a\Phi)/k!, \quad (3)$$

where a is the area (projected on the surface of the crystal) of the disordered region at specific depth, Φ is ion dose, and k is an integer number. In the case of $k=0$, S_0 determines the relative area of undamaged material. Based on Eq. (3), relative disorder in the crystal at specific depth as a function of ion dose can be written as

$$n_d = 1 - \exp(-a\Phi) \left\{ 1 + \sum_{k=1}^{\infty} [\alpha_k (a\Phi)^k / k!] \right\}. \quad (4)$$

Here, $\alpha_k = 1 - f_k$, where f_k is the level of relative lattice damage in a disordered region after a $(k-1)$ -fold spatial overlap. We have assumed that f_k linearly increases with increasing number of overlaps; i.e., $f_k = ck$, where c is the level of relative damage in a disordered region formed by single ion impact. In contrast to earlier models,^{21–23} we have assumed that f_k increases until the level of relative disorder within a disordered region at specific depth equals the critical level f_c when a catastrophic collapse into an amorphous phase occurs. After such a collapse, $f_k = 1$ and $\alpha_k = 0$. The critical level of relative disorder for such a collapse to occur has been chosen to be $f_c = 0.1$, based on experimental data from Refs. 24 and 25.

Results of such calculations are shown in Fig. 7 by a dashed curve. The best fit to experimental data was found with parameters a and c of 3500 \AA^2 and 0.027 , respectively. The value of $c = 0.027$ suggests that, on average, a threefold spatial overlap ($k=4$) of collision cascades is necessary for complete lattice disordering in the region of the maximum of the nuclear energy deposition profile in the case of bombardment with $0.5 \text{ MeV } ^{209}\text{Bi}_1$ ions at liquid nitrogen temperature. Note that the value of a is in reasonably good agreement with parameters of collision cascades at the maximum of the nuclear energy loss profile discussed above. Indeed, a characteristic radius of disordered regions with an area of $a = 3500 \text{ \AA}^2$, obtained from a fitting of experimental data shown in Fig. 7, is only about two times larger than the value of s obtained using ballistic calculations discussed above.

However, the above analysis of collision cascades revealed that $f_{dm} \cong 0.0024$ at the maximum of the nuclear energy loss profile (see Fig. 6). This value of f_{dm} is an order of magnitude less than the value of $c = 0.027$ obtained from a fitting of experimental data shown in Fig. 7. This result, however, is consistent with previous reports²⁶ where it has been found that the disorder level, as measured by RBS/C, after low temperature ion bombardment (which does not produce completely disordered regions by single ion impact) is about an order of magnitude higher than the level of atomic displacements predicted by ballistic calculations. The fact that lattice damage measured experimentally is significantly larger than that predicted using ballistic calculations is also supported by disorder depth profiles reported in this study. For example, Fig. 2 shows that relative disorder in the maximum of the nuclear energy loss profile is ~ 5 times larger than the total number of lattice vacancies calculated using the TRIM code.

Hence, both experimental data and theoretical estimations decidedly show that, during bombardment with MeV heavy ions (such as $0.5 \text{ MeV } ^{209}\text{Bi}_1$) at liquid nitrogen temperature, for most of the ion penetration depth (except for very end of range, where ions have relatively low energies), the damage buildup in Si cannot be considered as an accumulation of completely disordered regions. This is in contrast to the situation in Si under keV heavy ion bombardment at low temperatures.^{1,21} Moreover, collision cascades produced by MeV heavy ions are not nonlinear. In this case, what is the nature of the ME in the accumulation of stable lattice disorder experimentally observed in Figs. 1–3? A possible explanation for the ME observed is that, similar to the explanation currently accepted for the ME observed in the case of light-ion bombardment of Si,^{1–4} the ME may be attributed to nonlinear energy spike processes resulting from a spatial overlap of (relatively dense) collision subcascades. Such subcascades, which are clearly seen from Fig. 4, are formed in localized regions of high energy density within the larger volume of an individual collision cascade. The density of atomic displacements and/or nuclear energy deposition within some of these subcascades may be sufficient for the formation of completely disordered regions due to nonlinear energy spike processes. In addition, a spatial overlap of such subcascades produced by the atoms which form a molecular ion may result in the ME experimentally observed in this study.

Alternatively, the ME can be attributed to an increase in the defect clustering efficiency with increasing density of collision cascades in the case of implantation with molecular ions as compared with the case of atomic ion bombardment. It should be noted that, although the flux effect is usually not observed in Si bombarded with heavy ions at liquid nitrogen temperature,^{16,21} this cannot eliminate such a mechanism for the ME. Indeed, an increase in the density of collision cascades should enhance the formation of stable defects even for implant conditions with a low level of dynamic annealing when the flux effect is weak. This is due to the fact that interaction processes between point defects from the same collision cascade may occur in much shorter time intervals than possible migration and interaction of defects produced

in different collision cascades. However, at this stage, additional studies would be desirable to better understand damage processes in Si during MeV heavy-ion bombardment.

IV. CONCLUSIONS

In conclusion, damage buildup in Si bombarded at -196°C with MeV heavy atomic and molecular ions (the so-called molecular effect) has been studied by RBS/C. Results strongly suggest that, even for low temperature MeV heavy-ion bombardment, the buildup of lattice disorder cannot be considered as an accumulation of completely disordered regions, and collision cascades are not nonlinear. Results also show that, in the near-surface region, Bi_2 ions produce a larger level of lattice disorder than Bi_1 ions implanted to the same dose; i.e., the molecular effect with $\gamma > 1$ takes place. Such a molecular effect has been observed only in the near-surface region, where collision cascades produced by the atoms which form a molecular ion spatially overlap. The molecular effect has been explained based on (i) nonlinear energy spike processes resulting from a spatial overlap of (relatively dense) collision subcascades and/or (ii) an increase in the defect clustering efficiency with increasing density of ion-beam-produced defects.

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