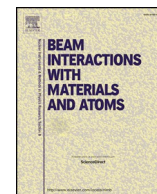




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Radiation defect dynamics in solids studied by pulsed ion beams

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ABSTRACT

The buildup of radiation damage is often a dynamic phenomenon proceeding via migration and interaction of point defects ballistically generated in collision cascades. Here, we describe how the dynamic aspects of damage formation via inter-cascade defect interaction can be studied by pulsed ion beam bombardment. We illustrate the pulsed beam method by an example for Si bombarded at 100 °C with 1.35 MeV Xe ions and compare with previously reported data for 0.5 MeV Xe ion irradiation. Results reveal that the time constant of defect relaxation strongly depends on both ion energy and the depth from the sample surface. This observation cannot be entirely explained as an effect of the collision cascade density on defect interaction dynamics, based on depth dependencies of cascade densities calculated with a recently developed model that accounts for the fractal nature of cascades. These results call for future systematic studies to better understand ion energy and cascade density effects on defect relaxation dynamics in Si.

1. Introduction

Understanding and controlling radiation damage is important for ion-beam processing of semiconductor devices such as doping, dry-etching, and electrical isolation. It is also a limiting factor in the performance of materials for service in nuclear energy systems and satellite electronics. Defect interaction dynamics is one of the most complex and least understood aspects of radiation damage [1,2]. The understanding of defect interaction dynamics has direct and important practical implications. Indeed, for most semiconductors and metals, point defects, generated in ballistic collisions, are mobile at process and/or service temperatures, which are typically room temperature and above. These mobile defects experience interaction processes, including the annihilation of vacancies and interstitials, defect clustering (into either interstitial or vacancies-based complexes), or defect trapping at pre-existing disorder, including surfaces, interfaces, extended defects, and impurity complexes. Such point defect migration and interaction processes occurring during irradiation are often referred to as *dynamic annealing* (DA) [1,2].

Our limited understanding of DA is related to the fact that both theoretical and experimental studies of DA are challenging. In particular, since the lifetime of defects dominating DA is often on time scales of $\geq 10^{-6}$ s [2,3], the powerful molecular dynamics method cannot be readily applied [4], and kinetic Monte-Carlo and rate theory approaches are often used instead [4,5]. However, these latter techniques

require explicit assumptions about the defect interaction processes involved and are challenging due to our limited understanding of the actual (both stable and metastable) defect configurations and reaction pathways in most materials. This applies to even crystalline Si, which is perhaps the most extensively studied material [2].

There is also limited experimental data available that can be used to critically assess and benchmark DA models. For example, damage buildup curves (i.e., the accumulation of a certain type or a group of lattice defects as a function of ion fluence) often cannot differentiate between physically different models [6]. Direct experimental studies of defect interaction dynamics are challenging. In fact, most DA studies in the past have not directly studied defect relaxation time constants. They have instead involved measurements of the dependence of damage on beam flux (see, for example, [7–9]). However, the separation of the temporal (defect lifetime) and spatial (defect diffusion length) information in such beam-flux-effect studies is not straightforward and typically requires explicit assumptions about defect interaction processes.

To circumvent these difficulties, our group at Lawrence Livermore National Laboratory (LLNL) has been developing a pulsed beam method to study defect interaction dynamics. Here, we illustrate the pulsed beam method with new data for Si bombarded at 100 °C with heavy (Xe) ions with an energy of 1.35 MeV and compare to previous pulsed-beam data for 0.5 MeV Xe ions. A larger ion energy of 1.35 MeV places the bulk damage peak deeper into the crystal bulk, away from the

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sample surface. We also calculate average collision cascade densities with a recently developed model that accounts for the fractal nature of collision cascades. Our results reveal a pronounced influence of ion energy on defect interaction dynamics that cannot be explained entirely by the depth dependence of the collision cascade density. These results reveal additional complexity of defect dynamics and call for further systematic studies.

2. Experimental

The 4 MV ion accelerator (National Electrostatics Corporation, model 4UH) at LLNL was used for both ion irradiation and ion beam analysis. Float-zone grown (100) Si single crystals (with a resistivity of $\sim 5 \Omega \text{ cm}$) were bombarded at 100°C with 1.35 MeV $^{129}\text{Xe}^+$ ions at 7° off the [100] direction to minimize channeling. For comparison, we also include data for pulsed beam bombardment of Si with 0.5 MeV Xe ions reported in [10]. To improve thermal contact, all samples were attached to a Cu sample holder with conductive Ag paste. All irradiations were performed in a broad beam mode (rather than with rastered beams) [3]. The total ion fluence (which was $6.2 \times 10^{13} \text{ cm}^{-2}$ for both 0.5 and 1.35 MeV Xe ions) was split into a train of equal square pulses with an instantaneous beam flux value (F_{on}) of $4 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$. The pulse duration (t_{on}) was 1 ms. The adjacent pulses were separated by time t_{off} , which was varied between 1 and 50 ms. The inset in Fig. 1(c) shows a schematic of the time dependence of instantaneous beam flux and defines pulsing parameters t_{on} , t_{off} , and F_{on} .

The dependence of lattice damage on t_{off} was studied *ex-situ* at room temperature by ion channeling with 2 MeV $^4\text{He}^+$ ions incident along the [100] direction and backscattered into a detector at 164° relative to the incident beam direction. Raw channeling spectra were analyzed with one of the conventional algorithms [11] for extracting depth profiles of relative disorder. Note that a relative disorder level of unity corresponds to full amorphization. Values of average relative disorder at different depths were obtained by averaging depth profiles of relative disorder over $\sim 40 \text{ nm}$. Error bars are standard deviations. Note that this method of calculating error bars reflects both statistical fluctuations of the ion channeling yield and any gradients in the depth distribution of damage that are pronounced particularly at depths on both sides adjacent to the bulk damage peak.

Three-dimensional distributions of ballistically-generated lattice vacancies were calculated with the TRIM code (version SRIM-2013.00, full cascade calculations) [12] with a Si atomic concentration of cm^{-3} and a threshold energy for atomic displacement of 15 eV. The average collision cascade density (ρ) was calculated with a fractal model from [13] by averaging over ≥ 600 individual cascades. As in [13], ρ is defined as the average local density of lattice vacancies within individual cascades with an averaging radius of 20 nm, which was chosen based on our recent estimates of the effective diffusion length of the mobile defects dominating DA in Si [13].

3. Results and discussion

In the pulsed-beam method developed in [2,3,10,14–19], the total ion fluence is delivered as a train of equal square pulses. The measurement of the effective time constant of DA (τ) involves ion bombardment of a series of specimens with all the irradiation parameters fixed except for t_{off} . Irradiation is followed by an *ex-situ* measurement of the level of stable disorder by, for example, ion channeling, as in the present work. An analysis of experimental dependencies of the level of stable disorder on t_{off} yields τ . A depth dependence of τ can be also obtained in cases of depth-resolved damage measurements, as in the present ion channeling study.

Fig. 1(a) depicts depth profiles of concentrations of vacancies, implanted Xe atoms, and ρ for Si irradiated with 0.5 and 1.35 MeV Xe ions. It reveals that the profiles of ballistically-generated vacancies are uni-modal, peaking at depths shallower than the maxima of implanted

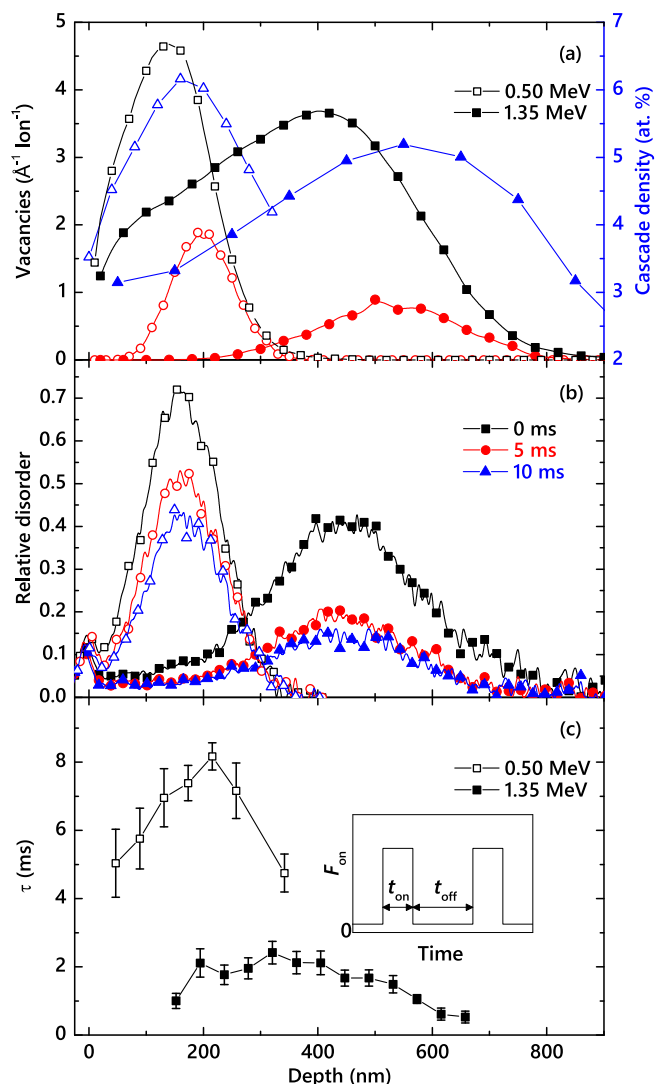


Fig. 1. Depth profiles of (a) concentrations of vacancies (squares, left axis), the average cascade density (triangles, right axis), and implanted Xe atoms (circles, with the linear vertical scale in arbitrary units), (b) relative bulk disorder, and (c) the effective defect relaxation time constant (τ) for Si bombarded at 100°C with pulsed beams of (open symbols) 0.5 and (closed symbols) 1.35 MeV Xe ions with $F_{\text{on}} = 4 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$, $t_{\text{on}} = 1 \text{ ms}$, and different t_{off} values given in the legend of panel (b). For clarity, only every 5th experimental point is depicted in panel (b). Data for 0.5 MeV Xe ions is taken from [10]. The inset in (c) shows a schematic of the time dependence of instantaneous beam flux, defining t_{on} , t_{off} , and F_{on} .

Xe atoms. This is expected based on ion ballistics. A less obvious observation from Fig. 1(a) is that, for both ion energies, depth profiles of ρ have maxima deeper than those of vacancies.

Ion-channeling-derived profiles of lattice disorder for Si bombarded with Xe ions with these two energies are shown in Fig. 1(b). Data for irradiation with different t_{off} values is given, with all the other irradiation conditions kept constant for each ion energy. It is seen from Fig. 1(b) that, for both ion energies used, bulk damage decreases with increasing t_{off} , while the damage level at the sample surface remains unchanged, suggesting different dynamic mechanisms of bulk and surface disordering. This behavior is qualitatively similar to that found in our previous studies (with pulsed beams of 0.5 MeV Ar ions) of 3C-SiC, 4H-SiC, Si, and Ge [3,15,16,18,20,21].

Fig. 2 summarizes dependencies of the damage level at the bulk peak on t_{off} for the two energies used (i.e., at ~ 160 and $\sim 450 \text{ nm}$ from the sample surface for cases of 0.5 and 1.35 MeV Xe ions, respectively).

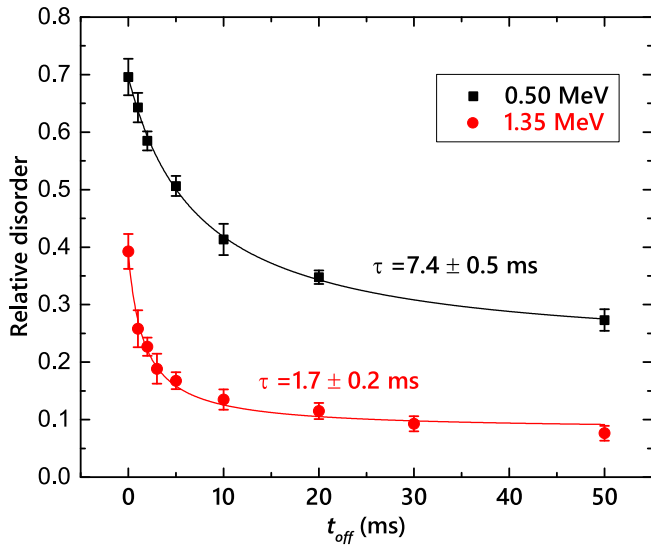


Fig. 2. Average relative bulk disorder in Si bombarded at 100 °C with a pulsed beam of 0.5 and 1.35 MeV Xe ions with $F_{\text{on}} = 4 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ and $t_{\text{on}} = 1 \text{ ms}$ as a function of the passive portion of the beam duty cycle (t_{off}). Fitting curves with the second order decay equation are shown by solid lines.

Solid lines in Fig. 2 are fits of the data via the Marquardt–Levenberg algorithm [22] with the second order decay equation: $n(t_{\text{off}}) = n_{\infty} + \frac{n(0) - n_{\infty}}{1 + \frac{t_{\text{off}}}{\tau}}$. Here, τ is the effective time constant of DA, and n_{∞} is relative disorder for $t_{\text{off}} \gg \tau$. Both dependencies from Fig. 2 obey the second order decay better than the first order (i.e., exponential) decay. Fig. 2 reveals a clear dependence of τ on ion energy.

To better understand the origin of such a dependence, we have analyzed damage levels at different depths. The resultant depth dependencies of τ for the two ion energies are plotted in Fig. 1(c). It reveals a pronounced depth dependence of τ . For both energies, the depth dependence of τ in Fig. 1(c) follows the depth dependence of ρ from Fig. 1(a). However, it is also clear that τ is determined not only by ρ but by some other parameters. Indeed, for 1.35 MeV Xe ions, at a depth of $\sim 500 \text{ nm}$ from the sample surface, ρ is the same as for 0.5 MeV Xe ions at a depth of $\sim 70 \text{ nm}$ [Fig. 1(a)], while τ values at these two depths differ by more than a factor of 2. Hence, the average cascade density (ρ) is not the only parameter defining τ for different irradiation conditions. This is somewhat unexpected given that a recent study [10] of Si bombarded with different mass ions has shown that ρ determines the behavior of both the activation energy of DA and the critical transition temperature between two regimes where different DA processes dominate. Values of τ could also be affected by collision cascade parameters other than ρ , effects of the instantaneous defect generation rate [2,23], electronic energy loss effects, or some DA processes that are specific for cascades created by high-energy ions. It is clear that more work is currently needed to better understand defect interaction dynamics in Si.

4. Summary

In summary, we have described a pulsed ion beam method to study the dynamics of defect interaction in solids. To illustrate the method, we have used examples for Si bombarded at 100 °C with 0.5 and 1.35 MeV Xe ions, with depth profiles of disorder measured by ion channeling. Our results have demonstrated that, for these irradiation conditions, the defect relaxation time constant (τ) in Si depends strongly on both ion mass and depth from the sample surface. Moreover, at least for these conditions, the average collision cascade density is not the sole predictor of τ , and more work is needed to better understand defect interaction dynamics in Si. These experimental results can be used to benchmark models of radiation damage in Si.

Acknowledgments

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