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Coupled electronic and atomic effects on defect evolution in silicon carbide under ion irradiation ☆

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ABSTRACT

Understanding energy dissipation processes in electronic/atomic subsystems and subsequent non-equilibrium defect evolution is a long-standing challenge in materials science. In the intermediate energy regime, energetic particles simultaneously deposit a significant amount of energy to both electronic and atomic subsystems of silicon carbide (SiC). Here we show that defect evolution in SiC closely depends on the electronic-to-nuclear energy loss ratio (S_e/S_n), nuclear stopping powers (dE/dx_{nucl}), electronic stopping powers (dE/dx_{ele}), and the temporal and spatial coupling of electronic and atomic subsystem for energy dissipation. The integrated experiments and simulations reveal that: (1) increasing S_e/S_n slows damage accumulation; (2) the transient temperatures during the ionization-induced thermal spike increase with dE/dx_{ele}, which causes efficient damage annealing along the ion trajectory; and (3) for more condensed displacement damage within the thermal spike, damage production is suppressed due to the coupled electronic and atomic dynamics. Ionization effects are expected to be more significant in materials with covalent/ionic bonding involving predominantly well-localized electrons. Insights into the complex electronic and atomic correlations may pave the way to better control and predict SiC response to extreme energy deposition.

1. Introduction

Understanding, predictively modeling and ultimately controlling the material response to energy deposition and dissipation from energetic particles at the level of electrons and atoms are a grand challenge in material science across diverse fields. Extreme energy deposition and dissipation are non-equilibrium processes. Material responses to these processes are the foundation for a wide range of research and applications, including laser-assisted additive manufacturing, semiconductor ion doping technology, nuclear material performance in both fission and fusion reactors, long-term performance of nuclear waste forms, radiation therapy, as well as materials and device performance in space exploration. Material response to energy deposition, especially by energetic ions, has been under intensive investigation for many decades due to the unique control of material modification [1–19]. An ion penetrating a target material deposits its energy to both the atomic and electronic subsystems through nuclear (dE/dx_{nucl} or S_n) and electronic (dE/dx_{ele} or S_e) stopping powers, respectively. It is well established experimentally and in the recent rapid development of modeling work that the response of materials to energy deposition in both subsystems is inherently connected through a disturbance of ordered atomic arrangements and electronic structure [1–10]. Such energy deposition and subsequent dissipation initiate rich sets of electronic/atomic-level interactions and non-equilibrium atomic processes on energy landscapes. The energy dissipation within the electronic and atomic subsystems and the exchange of energy between the two subsystems can act to

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dampen [2,3] or accelerate ionic motion [20], to inhibit or enhance defect production [1–10,15,16], to create kinetically stable and metastable defects and phases [3,21], or to quench damage accumulation [2,7,9,10,15–18].

Irradiation effects in materials have been advanced mainly under two extreme conditions: collision cascade processes dominated at low energies and ionization processes dominated at high energies [1–3,22]. This generally means that, while ion energy deposition into the electronic and atomic subsystems are coupled in both time and space, the nuclear and electronic energy deposition and dissipation are often simplified as independent and non-correlated defect dynamic processes, which are considered separately. In the low-energy regime, basic processes of defect and damage production by ballistic collisions on the atomic structure are reasonably well understood. Atomic collision processes due to energy transfer from incident ions to target atoms through nuclear energy loss [3,11–14,23,24] cause displacement events and lead to defect formation, migration and recombination, as shown by experimentally observable microstructure changes. The collision and energy transfer processes between atoms are also investigated using screened empirical interatomic potentials in classical molecular dynamics (MD), and the entire collisional energy cascade can be described with temporal and spatial characteristics. In the high-energy regime, such as for swift heavy ions (generally defined as ion energy >0.5 MeV amu^{-1}, ion mass >20 amu, and dE/dx_{ele} much larger than 10 keV nm^{-1}), the subsequent thermal spikes (hot electrons created from extreme electronic energy deposition transfer much of their energy to atoms along the ion path, causing local heating) are believed to reach much higher temperatures (more than ten thousand K) [3], resulting in the formation of amorphous tracks in many ceramic oxides [3,7,25–27] or annealing of pre-existing defects in silicon carbide [2,22,28,29].

In many studies employing ion-beam irradiation, the ion energies are above the low-energy (nuclear-dominated) regime and below the high-energy (electronic-dominated) regime, and the damage evolution in this intermediate energy regime is a consequence of coupled ionization effects and displacement events occurring at the same time in the same space. While interactions between energetic ions and target nuclei lead to atomic displacements through elastic collisions, target electrons can absorb a significant amount of the incident ion energy through inelastic interactions, from a few percent up to more than 90%. In the case of ions with energy ranging from a few keV amu^{-1} to a few hundred keV amu^{-1} (the intermediate energy regime), dE/dx_{ele} can be much larger than dE/dx_{nucl}. Electronic energy deposition in semiconductors and insulators can cause ionization that produces localized electronic excitations and local thermal spikes (highly localized heating in the atomic subsystem along the ion paths, due to electron–phonon coupling), which can modify migration barriers and cause local atomic rearrangements, respectively [3,30–32]. This means that the electronic energy loss can affect defect dynamics and damage accumulation processes. Due to the common assumption that this energy is largely dissipated away without noticeable effects on the atomic structure, the effects of electronic energy loss have been essentially ignored in many studies related to ion–solid interactions in the past. While energy is thought to dissipate through the electronic subsystem rapidly in metals and conventional alloys (metallic bonding in the form of an electron cloud of delocalized electrons) and consequently have less impact on atomic-level defect dynamics, new discoveries have shown that extreme compositional complexity in concentrated solid-solution alloys can substantially reduce electron, phonon, and magnon mean–free paths, alter coupling strengths, and therefore greatly affect defect production and microstructural evolution [9,11,33–38]. Recent work [3] has demonstrated that ionization effects in SiC, a ceramic material with predominantly covalent bonding [2,39], are very significant.

In this work, we address the coupled dynamics of electronic and atomic processes induced by charged-particle irradiation in the intermediate energy regime in SiC. Energy deposition and dissipation in electronic and atomic subsystems are explored through processes coupled temporally and spatially. Defect production and damage accumulation are investigated as a function of both dE/dx_{nucl} and dE/dx_{ele}, as well as the electronic-to-nuclear energy loss ratio, S_e/S_n, by varying ion energy, ion mass, and the incident angle. Building on the existing knowledge in the low- and high-energy regimes [24,30,32], from separate effects studies [2,3,28,29] and new information derived from this work, we demonstrate conclusively here that ionization can effectively anneal radiation damage and suppress damage accumulation in SiC through temporal and spatial coupling of ionization with displacement events.

2. Materials and methods

2.1. Ion irradiation and ion beam analysis

Single crystal SiC used in this study were 4H- and 6H-SiC single crystal wafers from Cree, Inc. Highly ionizing irradiations using 4.5 MeV C, 6.5 MeV O, 21 MeV Si, 21 MeV Ni and 25 MeV Au were conducted at room temperature [50]. Au irradiations were performed on 6H-SiC, both along and a few degrees (~7°) off the ⟨0001⟩ direction (in-channeling and off-channeling directions, respectively), while all the other irradiations were performed using 4H-SiC. Damage production and accumulation due to displacement damage in different polytypes (3C, 4H or 6H) have been studied previously, and no clear difference is observed.

To better understand interactions of Au ions with pre-existing defects, a separate effects study was conducted. Prior to the Au irradiations, low-energy bombardments were performed using 900 keV Si with fluences of 6.12×10^{14} and 6.2×10^{14} cm^{-2} on 6H-SiC, respectively, to produce a shallow pre-damage profile peaked at 650 nm.

Backscattering measurements along the ⟨0001⟩ direction were utilized to quantify the irradiation-induced damage. Helium ions of 3.5 MeV were employed to probe the entire damage profile. Compared with 2.0 MeV He typically used in Rutherford backscattering spectrometry (RBS) measurements, the lower dE/dx_{ele} of 3.5 MeV He results in deeper depth analysis. Moreover, 3.5 MeV He with a scattering angle of 155° induces non-Rutherford backscattering (NRBS) with the C atoms and results in much higher non-Rutherford cross sections, as shown by the enhanced backscattering yield and clear surface peak (Fig. 1). The utilization of RBS on Si atoms and NRBS on C atoms enables the disorder on both sublattices to be quantified from a single channeling backscattering measurement [50]. These channeling spectra were analyzed using an iterative procedure to achieve the relative disorder level as a function of depth [51].

2.2. Scanning transmission electron microscope characterization

High angle annular dark field (HAADF) imaging was performed on various samples using a 5th order aberration corrected scanning transmission electron microscope (STEM, Nion UltraSTEM200) operating at 200 kV. A detector with an inner semi-angle of 65 mrad was used to collect electrons for HAADF imaging. The electron probe current used in the measurement was 28 pA. The samples for the STEM characterization were prepared by focus ion beam in a scanning electron microscope, followed by a low energy

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