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Modelling radiation effects in solids with two-temperature molecular dynamics

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ABSTRACT

The ability to predict the structural modifications of materials resulting from a broad range of irradiation scenarios would have a positive impact on many fields of science and technology. Established techniques for modelling large atomic systems, such as classical molecular dynamics, are limited by the neglect of the electronic degrees of freedom which restricts their application to irradiation events that primarily interact with atomic nuclei. Ab initio methods, on the other hand, include electronic degrees of freedom, but the requisite computational costs restrict their application to relatively small systems. Recent methodological developments aimed at overcoming some of these limitations are based on methods that couple atomistic models to a continuum model for the electronic energy, where energy is exchanged between the nuclei and electrons via electronic stopping and electron-phonon coupling mechanisms. Such two-temperature molecular dynamics models, as they are known, make it practicable to simulate the effects of electronic excitations on systems with millions, or even hundreds of millions, of atoms. They have been used to study laser irradiation of metallic films, swift heavy ion irradiation of metals and semiconductors, and moderately high ion irradiation of metals.

In this review we describe the two-temperature molecular dynamics methodology and the various practical considerations required for its implementation. We provide example applications of the model to multiple irradiation scenarios that accommodate electronic excitations. We also describe the challenges of including the effects of the modification of the interatomic interactions, due to the excitation of electrons, in the simulations and how these challenges can be overcome.

1. Introduction

The effects of radiation on materials impacts many scientific and technological fields, ranging from nanotechnology and microelectronics to the nuclear industry and space technology. Radiation has many forms, but in terms of its effects on materials it can be broadly classified into three types: radiation that primarily excites electrons, radiation that primarily excites the nuclei, and radiation that deposits proportionately to both the electrons and the nuclei. Photons and very high energy ions belong to the first class, neutrons and low energy ions belong to the second class, and moderate to high energy ions belong to the third.

Predicting the effect of radiation on materials requires knowledge that spans an enormous range of length and time scales. Defect configurations created on the nanoscale and in the first few picoseconds following an irradiation event continue to evolve over many years, resulting in potentially catastrophic degradation of the material properties. It is, therefore, important to understand and characterise defect creation and evolution at all stages of the ageing process, to assess whether a material is fit for purpose in a radiation environment.

Classical molecular dynamics (MD) is a powerful method for modelling radiation effects in the early stages following a radiation event, as it can simulate appropriate length (nm) and time (ns) scales. It has enabled many notable advances in the understanding of radiation damage over the past three decades. There are, however, well-known limitations to the method and these are briefly discussed in Section 2 of this review. The limitation that is most relevant to the current review is the inability of the conventional MD method to incorporate the effects of electronic excitations, which restricts the class of radiation that can be studied by this method to that which interacts primarily with the nuclei in a material, such as low energy ions. Indeed, some types of radiation only excite electrons, in which case the standard MD method is entirely inappropriate. An example of radiation that interacts solely with electrons is laser irradiation, whereby the photons are absorbed by the material via the excitation of electrons to higher energy

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levels. Very energetic ions, generally referred to as swift heavy ions (SHI), also interact strongly with the electrons in a material. When an ion moves through a material it gradually loses energy and the energy loss per unit distance is known as the stopping power. There are two main mechanisms for this energy loss. One is the elastic collisions of the moving ion with the nuclei of the material, referred to as nuclear stopping power, and the second is energy loss due to the inelastic scattering of electrons, known as electronic stopping power. The relative amounts of energy loss via the two mechanisms is strongly dependent on the energy of the moving ion, with the typical relationship illustrated in Fig. 1. For very energetic projectiles, it can be seen that the energy loss is primarily due to electronic stopping power and the cross section for nuclear collisions is extremely low. It follows that conventional MD simulations are not appropriate for modelling the effects of swift heavy ions, which are very high energy projectiles.

To address this limitation, augmented MD methods have been developed that couple classical MD to a model for the transfer and diffusion of electronic energy. These efforts were initially developed to simulate laser irradiation of metal films [1–3] and electron ejection from metal surfaces [4–6]. A later development extended the model for cascade simulations that enabled the effect of electronic energy loss and electron-phonon coupling to be included in simulations of irradiation with moderate to high energy ions [7].

In this paper we describe an augmented MD methodology, known as two-temperature molecular dynamics (2T-MD). We give examples of how the method has been used successfully to model laser irradiation, swift heavy ion irradiation and moderate energy ion irradiation. We also show evidence that, for very highly excited electrons, coupling the lattice and the electrons via electron-phonon coupling is not sufficient to capture the full effects of the electronic excitations on the dynamics of the system. Rather, intense excitations affect the interactions between the atoms themselves, in which case it is necessary to employ dynamical interatomic potentials that are parametrized by the electronic temperature. We discuss how such interatomic potentials can be developed by fitting to finite temperature density function theory (DFT) calculations and how they can be employed in classical MD simulations. We conclude with a summary of progress in this research area to date and future challenges.

2. Simulating radiation effects

2.1. Molecular dynamics

The most widely used methodology for simulating radiation effects in materials is molecular dynamics in the form of cascade simulations. Cascade simulations model the effects of a particle interaction with the atoms in a solid by imparting a high velocity to one atom in a simulation cell. This atom, known as the primary knock-on atom or PKA, moves through the crystal and collides with other atoms, also setting them in motion. The resulting collision cascade, as it is known, continues creating an increasing number of mobile atoms until the energies of all atoms have fallen below the threshold required to knock an atom from its lattice site. At this stage the energy of the mobile atoms is shared thermally amongst their neighbours, resulting in a localised region of liquid-like material with a high energy density encapsulated within a cooler crystal structure. As time passes, the energy diffuses through the crystal and the molten region cools, fully or partially recrystallising. Partial recrystallisation leaves behind an amorphous volume embedded in the crystal, but even full recrystallisation generally concludes with many defects that may take the form of isolated vacancies and interstitials, small clusters, or dislocation loops. Cascade simulations, as reviewed in [8], have made an enormous contribution to the understanding of the fundamental mechanisms involved in radiation damage.

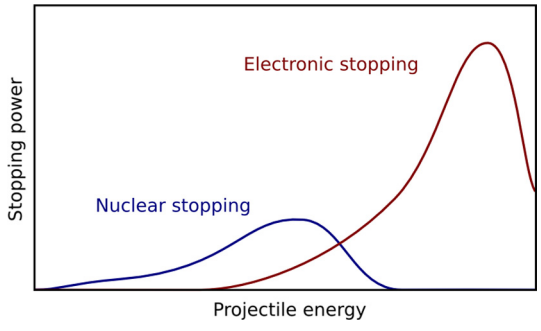

Fig. 1. A schematic representation of the nuclear (blue) and electronic (red) stopping power as a function of the ion projectile energy. For low energy ions most energy is lost to the nuclei in the material whereas for high energy ions the inelastic electronic interactions dominate the energy loss. At intermediate energies both mechanisms are important. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The success of cascade simulations has, of course, been tempered by the usual limitations of MD methods. One limitation is that the size of the simulation cell required to contain the cascade effects increases with increasing PKA energy, with approximately 100 million atoms required to simulate a PKA energy of 500 keV. Another limitation is the use of interatomic potentials that have been fitted to equilibrium conditions, which deviates strongly from the far-from-equilibrium conditions experienced in high energy collisions. This is overcome to some extent by the employment of a universal potential which takes the form of a Coulomb potential times a screening function, such as the Ziegler-Biersack Littmark (ZBL) potential [9]. The ZBL potential is utilised at very close interatomic separations, being merged with the interatomic potential of the material at intermediate separations, and deactivated at larger separations, leaving only the unmodified interatomic potential. It has, however, been noted that the residual defect numbers and defect configurations are quite sensitive to the details of the joining procedure, and so the procedure must be performed with care [10].

The third limitation, and the one most relevant to the current review, is that classical cascade simulations are restricted to modelling radiation events that transfer energy solely to the nuclei since they neglect the effects of excited electrons. As an ion moves through a solid, the proportion of its energy lost to inelastic electronic scattering increases as the cross-section for nuclear collisions decreases (Fig. 1). These effects attracted significant interest in the late 1980s and early 1990s when a few seminal papers [11–13] were published. Thereafter, it became standard practice to include the effects of electronic stopping by introducing a friction term to the MD equations of motion. In these classical simulations there is no facility for describing the electronic energy therefore the energy dissipated by electronic stopping is lost by the system. Consequently, such methods neglect the transport and redistribution of the electronic energy, which is one of the issues that the 2T-MD model aims to resolve.

2.2. The continuum two-temperature model

High energy radiation events generally drive the electrons in a solid far from equilibrium. Shortly after the radiation event, usually within the femtosecond time-scale, the electrons thermalise and adopt a well-defined temperature. However, the thermalized electronic temperature will initially differ from that of the nuclei. This state of quasi-equilibrium will evolve, usually on a picosecond time-scale, to a state of equilibrium whereby the electrons and nuclei converge to the same temperature. The initial heating of