Chapter 2

ACCELERATED MOLECULAR DYNAMICS METHODS

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1 INTRODUCTION

The evolution of radiation damage in materials is a classic example of a problem that spans many time and length scales. The initial production of damage occurs on the atomic scale via collision cascades that take place on the picosecond time scale. However, this damage ultimately manifests itself macroscopically in the form of swelling or cracking which can take years to develop. There is a wide range of phenomena that bridge these two extremes, including defect diffusion, annihilation and aggregation, the formation of interstitial loops and voids, and the development of more complex microstructure. As a result, no one simulation method can be employed to study the problem of radiation damage on all relevant time and length scales. Rather, a combination of many techniques must be used to address this problem.

Molecular dynamics (MD) simulation, in which atom positions are evolved by integrating the classical equations of motion in time, is ideally suited to studying the collision cascade. MD simulations can probe timescales of ps to ns, which is the time typically required to initiate a cascade and to allow it to evolve until the thermal spike of the collision has dissipated. Thus, the damage produced in the collision cascade can be directly simulated using MD.

However, once that damage has been formed, diffusion and subsequent annihilation or aggregation of those defects can occur on much longer time scales, perhaps even seconds or beyond, depending on the conditions (temperature, pressure, etc.). Such phenomena must be accounted for in order to accurately predict larger scale features – including interstitial dislocation loops and vacancy voids – that comprise the overall microstructure of the system and lead to macroscopic response to radiation damage such as swelling and cracking. However, MD will not allow us to study defect behavior on these longer timescales.

Recently, methods based on a new concept have been developed for circumventing this time scale problem. For systems in which the long-time dynamical evolution is characterized by a sequence of activated events – typically the case for defect
diffusion – these “accelerated molecular dynamics” methods [1] can extend the accessible time scale by orders of magnitude relative to direct MD, while retaining full atomistic detail. These methods – hyperdynamics, parallel replica dynamics, and temperature accelerated dynamics (TAD) – have already been demonstrated on problems in surface and bulk diffusion, surface growth, and molecular problems. With more development they will become useful for a broad range of key materials problems, including grain growth, dislocation climb and dislocation kink nucleation. Here we give an introduction to these methods, discuss their current strengths and limitations, and demonstrate their use in problems involving radiation damage.

While the treatment given here is similar to that presented previously [2], the examples used to illustrate the accelerated dynamics methods were chosen because of their relevance to studies of radiation damage.

2 BACKGROUND

2.1 Infrequent Event Systems

We begin by defining an “infrequent-event” system, as this is the type of system for which the accelerated dynamics methods are ideal. The dynamical evolution of such a system is characterized by the occasional activated event that takes the system from basin to basin, events that are separated by possibly millions of thermal vibrations within one basin. A simple example of an infrequent-event system is an adatom on a metal surface at a temperature that is low relative to the diffusive jump barrier. We will exclusively consider thermal systems, characterized by a temperature $T$, a fixed number of atoms $N$, and a fixed volume $V$; i.e., the canonical ensemble. Typically, there is a large number of possible paths for escape from any given basin. As a trajectory in the $3N$-dimensional coordinate space in which the system resides passes from one basin to another, it crosses a $(3N-1)$-dimensional “dividing surface” at the ridgetop separating the two basins. While on average these crossings are infrequent, successive crossings can sometimes occur within just a few vibrational periods; these are termed “correlated dynamical events” (e.g., see [3–5]). An example would be a double jump of the adatom on the surface. For this discussion it is sufficient, but important, to realize that such events can occur. In two of the methods presented below, we will assume that these correlated events do not occur. This is the primary assumption of transition state theory, which is actually a very good approximation for many solid-state diffusive processes. We define the “correlation time” ($\tau_{\text{corr}}$) of the system as the duration of the system memory. A trajectory that has resided in a particular basin for longer than $\tau_{\text{corr}}$ has no memory of its history and, consequently, how it got to that basin, in the sense that when it later escapes from the basin, the probability for escape is independent of how it entered the state. The relative probability for escape to a