Coarse Grained Approach to First Principles Modeling of Radiation Cascade in Large Fe Supercells

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Abstract. Classical Molecular Dynamics (MD) simulations characterizing dislocations and radiation damage typically treat $10^5$-$10^7$ atoms. First principles techniques employed to understand systems at an atomistic level are not practical for such large systems consisting of millions of atoms. We present an efficient coarse grained (CG) approach to calculate local electronic and magnetic properties of large MD-generated structures from the first principles.

Local atomic magnetic moments in crystalline Fe are perturbed by the presence of radiation generated vacancies and interstitials. The effects are most pronounced near the defect cores and decay slowly as the strain field of the defects decrease with distance. We develop the CG technique based on the Locally Self-consistent Multiple Scattering (LSMS) method that exploits the near-sightedness of the electron Green function. The atomic positions were determined by MD with an embedded atom force field.

The local moments in the neighborhood of the defect cores are calculated with first-principles based on full local structure information. Atoms in the rest of the system are modeled by representative atoms with approximated properties. The calculations result in local moments near the defect centers with first-principles accuracy, while capturing coarse-grained details of local moments at greater length scales. This CG approach makes these large scale structures amenable to first principles study.

Introduction
Radiation induced damage has substantial impact on the longevity of structural materials used in fission and fusion reactors. The primary radiation damage such as displacement defects initiate a build up of defect aggregates at the micro scale, and eventually lead to the altering of the material’s mechanical properties [1, 2]. Fundamental properties of point defects and defect accumulations in bcc-Fe under irradiations have been investigated experimentally [3, 4, 5, 6]. The radiation induced damage in structural metals, and its evolution dynamics are highly complex processes and extremely difficult to investigate in-situ. Therefore, efficient numerical modeling at multiple scales represents a valuable tool in understanding the underlying physical processes associated with defects.
Classical Molecular Dynamics simulations have been the most prolific tool for investigating time evolution of defects within nanosecond range [7, 8, 9]. Recent developments in classical MD simulations of defects utilize classical force fields, e.g. embedded atom method (EAM) [10], and incorporate interatomic potentials computed from first-principles calculations. A magnetic EAM force fields that include the local volume effect on local magnetic moments has been developed by Dudarev et al [11]. Using Kinetic Monte Carlo based technique is an approach to investigate long term temporal and spatial evolution of defects [12, 13].

In recent years, there have been extensive efforts underway in coupling the classical and quantum simulation techniques along with continuum mechanical simulations [16, 14, 15]. The so called multi-scale Coarse-Grained (CG) technique used mostly for biological molecules represents one or more atoms by one CG site and computes interactions between the CG regions from atomistic configurations [16, 17, 18, 19, 20].

These approaches focus on the evolution dynamics of defects or complex molecules. The CG approach presented here looks at radiation damage from a slightly different point of view: that is local electronic and magnetic properties at time snapshots during a classical MD simulation. As it has been shown in MD simulations [21], radiation in the bulk iron produces only a few highly localized defect cascades, and transfers most of its energy as thermal displacements of atoms in the bulk. The thermal displacements change the local electronic and magnetic states, but can still be reasonably averaged in a manner that reflects the local strain.

The CG approach presented here exploits this contrast between the defect cores and rest of the system by treating the the defect core atoms explicitly while representing the rest of the system with CG average parameters. In addition, we employ the LSMS method that exploits the near-sightedness of the electronic Green function. This technique enables us to investigate local properties of radiation damage samples from first principles using very large MD simulation data and is applicable to two broad categories of simulation: relaxation of atomic positions to study the structure of static dislocation cores and molecular dynamics (MD) simulations of the defects.

Computational Details

**LSMS**

The core of our computational tool is the Locally Self-consistent Multiple Scattering method (LSMS) [22], an all-electron, real space multiple scattering theory based approach within the framework of density functional theory (DFT) with local spin density approximation (LSDA) [23].

Being implicitly parallel, the key feature of the LSMS is to associate a processor with every atom and vacant site. Each processor solves the multiple scattering equations for the Green function of a cluster (local interaction zone), of which it is the center and calculates the electron density (as a function of the maximum occupied energy) in the Voronoi polyhedron directly from the Green functions. The union of the site densities computed up to the Fermi energy gives the electron density of the system, and is set to satisfy global charge neutrality constraint. The potential is reconstructed by solving Poisson’s equation and adding the exchange-correlation potential in the usual manner of LSDA and the self-consistent cycle proceeds to convergence in the familiar fashion. The long range electrostatic contribution to the potential is evaluated by the Madelung procedure assuming periodic boundary conditions.

**Coarse Grained Approach**

There are several issues to be resolved in constructing a local portion of a large MD configuration in first principles calculations. The CG approach takes all of the structural information as input, this includes explicit coordinates and boundary conditions that imply, for example, periodic repetition in some directions and termination in others.
The CG scheme used in iron radiation damage sample. The system is represented by fewer selected atoms (magenta). The defect core atoms are given explicitly (cluster in magenta), and the rest of the system is represented by atoms selected on the nodes of simple mesh. As an example, a set of pseudo-atoms represented by one atom is shown gold colored.

The coordinates of atoms in the desired first principles region are given explicitly. In addition, we choose representative atoms spread throughout the simulation volume by some prescription and give their coordinates explicitly. The multiple scattering equations are solved self consistently on these atoms only. The remaining atoms are assigned charges (higher multipoles in a full potential calculation) and scattering t-matrices from the nearest representative atom. The t-matrices of the periodic image sites are dealt with as in the original LSMS. Given these associations the LSMS calculates self-consistently the density and potential appropriate to the local strain in the regions surrounding each representative atom and transfers this information to its associated pseudo-atoms. This scheme converts most of the original MD structure into spatially coarse grained structure, while leaving the region under investigation accessible from first principles. The schematics of this CG approach is shown in Fig.1.

**Results**

To validate the CG approach, we first investigated a 9826 atom radiation damaged bulk iron sample using both full atom first principles calculation and the CG calculation using just 629 representative atoms. The primary damage state of the sample and following cascade states
were obtained through MD simulations using empirical, embedded-atom inter-atomic potentials [24, 25]. The MD simulation started as a 500 eV radiation event at constant pressure and was terminated when all atomic motions have thermalized at $t = 3.6$ ps.

In the full atom first principles calculation, we treated all 9826 atoms explicitly and converged the LSMS self consistently to obtain moments on each atom. Local volumes for each site have been calculated directly from the structural data. The local volumes change by as much as 12.5% from the average and this change is distributed throughout the simulation volume consistent with a reflected shock wave under periodic boundary conditions. A closer inspection of the structure reveals the compact nature of defects left by the radiation.

In order to investigate the defect regions from the first principles, the atoms at the defect cores have been identified from the structural data. Equipped with this information, atoms in the region encompassing the defects are given explicitly. The rest of the system is modeled by a set of representative atoms that are chosen on the nodes of 8x8x8 mesh in the simulation volume. As a result, the 9826 atom structure at $t = 3.6$ ps is now reduced to 629 atom CG model, in which atoms in the defect region are given explicitly.

The CG calculation captures the spatial distribution of local volumes in the full atom calculation as expected. Atom by atom comparison between the calculations shows that the local moments near the defect cores closely match. For the rest of the system, the CG model captures the thermal fluctuation range of the full atom case (Fig.2, left). At $t = 3.6$ ps, site moments calculated by the CG technique (red) matches the full atom calculation (blue) closely. Despite treating the defect regions explicitly, moments at some of the atoms in these region (red)
Figure 3. Local moments in the 9826 atom radiation damaged bulk iron sample at different time snapshots. The color and size of the arrows shown here represent deviation from an average value of moments, and their directions indicate local magnetization directions in the system. Blue colors correspond to decreased local moments, and increased moments are shown in the red of the color scale. Earlier time step at 1170 (t=3.48 ps, left) contains more point defects with greater magnetic disturbance than time step 1197 (t=3.6 ps, right). In particular, there is one anti-ferromagnetic coupling formed in the defect region (the largest blue arrow pointing down).

differ from that of full atom calculation (blue). Further analysis revealed that this difference is caused by different convergence behavior of the two calculations. The convergence value set for the Root Mean Square (RMS), (Definition: square root of average over atomic volume of the square of the difference between input and output electron densities) is achieved at different rates since the two calculations use vastly different number of atoms (Fig.2, right).

The evolution of radiation cascades can be investigated by comparing snapshots of the system at different time steps. CG model of the system at t=3.48 ps is constructed using the same procedure described above. Being earlier stage in the cascade, the system now contains more atoms in the defect regions, resulting in 657 atoms for the CG model. The calculation reveals more pronounced perturbations in the system than the t=3.6 ps (Fig 3).

After validation on a 9826 atom system, we extended the CG calculation to a larger system consisting of 54K atoms. The classical MD simulations of 1 keV radiation damage has been performed using the same techniques as before. Treating a system of this size from first principles requires significant computational resources and poses challenges associated with numerical convergence. We constructed a CG model of this system using 1241 representative atoms only. Classical MD simulations show that the number of atoms involved in point defects has no direct correlation with the system size and depends only on the energy introduced during the irradiation. Since the number of atoms in the defect regions is comparable to that of the 9826 atom system, increase in the number of representative atoms is due mostly to the number of mesh points that represent the larger simulation volume. Changes in local volumes due to the irradiation is available from the structural data and are closely captured in the CG model (Fig.4, left). The damage is concentrated in a very compact region near the center of the simulation volume, and is mostly compressed local volumes (blue). The rest of the system exhibits atomic
Figure 4. Local volumes (left) and moments (right) for 54K atom radiation damaged bulk iron sample (1 keV, pka-1, T=100 K, t=3.947 ps). The color and size of the arrows shown in the right represent deviation from an average value of moments, and their directions indicate local magnetization directions in the system. Blue colors correspond to decreased local moments, and increased moments are shown in the red of the color scale. The blue arrows pointing down correspond to locally anti-ferromagnetic couplings formed in the defect cores. The calculation is performed atomistically in the defect regions while treating rest of the system using the CG technique.

It should be noted that there is no comparable first-principles data currently available for systems of this size. The radiation induces compact damage aggregates consisting of point like defects, some of which form local environments that favor an anti-ferromagnetic state. The locally anti-ferromagnetic coupling between iron atoms has been observed before, and are signature of an early stage of radiation damage [21].

Summary
We developed a coarse-grained scheme for solving multiple scattering equations by representing a system with fewer sites. This approach allows us to treat part of a system from the first principles, while substituting the rest of the system with CG model. Comparison with the full atom calculation of 9826 atom system shows that the technique calculates magnetic properties of the selected region accurately from the first principles, while capturing CG distribution of the properties for the rest of the system. Calculation of local properties for 54K atom system demonstrates that this CG technique can efficiently handle larger systems. We also observed that the convergence behavior of the self-consistent calculation improves due to the reduced number of sites. This CG technique substantially reduces the computational resources needed.

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