White-noise quantum heat bath for MD and SLD simulations in magnetic materials

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In both MD and SLD models, phase-space trajectories of activation processes are obtained by integrating classical equations of motion. Thermodynamic equilibrium/quasiequilibrium conditions among different parts of the system are maintained through temperature, which is controlled through the use of classical Boltzmann statistics. This route cannot produce sufficiently accurate results unless the atoms under consideration have wavelengths shorter than the interatomic spacing, and have vibrating frequencies corresponding to a Debye temperature much lower than the temperature of interest. In the former case, the classical particle picture breaks down and transition via quantum tunneling must be considered. In the latter case, quantum statistics have to be used to ensure that the correct thermodynamics conditions are kept in the simulations. Well known cases in MD simulations where quantum effects must be considered include light atoms such as hydrogen and helium. A similar, but even more serious, problem exists in SLD simulations of irradiation-generated lattice defects in magnetic materials. The quantization of the energy spectrum of the spin system is about an order of magnitude larger than that of the lattice and its effects cannot be neglected even for temperatures higher than the melting point of iron. Under such conditions the heat reservoir designed to control the spin temperature based on classical statistics fails. A quantum-physics based methodology is required to remove the restriction of the classical thermostat to solve this problem. In this paper, we will show that a white-noise temperature control can be reformulated based on the Langevin thermostat methodology. By recasting the scheme in an alternate form, using the Bose-Einstein statistics instead of the Boltzmann statistics, a quantum heat reservoir may be constructed from a simple modification of the classical fluctuation-dissipation relation. Development of the theory and associated numerical techniques for the quantum heat reservoir is considered in this presentation.

Effects of magnetism and strain relaxation on the phase stability of multi-component alloys

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Fe-Cr-Ni alloys and austenitic steels form an important class of high temperature structural materials developed for nuclear and related applications. Fe-Cr-Ni alloys are among the most extensively studied ternary alloy systems, still the relation between their phase diagram, still the effect of magnetism on their phase stability and defect structures formed under irradiation is not known. We investigate Fe-Cr-Ni alloys by using a combination of several simulation methods. Density functional theory (DFT) and Cluster Expansion methods are combined in order to derive the cluster expansion parameters characterizing bcc and fcc crystal structures. DFT-based Monte Carlo (MC) simulations are then used for predicting the relative stability of fcc and bcc phases in Fe-Cr-Ni at elevated temperatures. Enthalpies of formation obtained from MC simulations at finite temperatures, as well as order-disorder temperatures and short-range order parameters predicted for a broad range of temperatures are compared with experimental data. Magnetic Cluster Expansion, a new method that extends the conventional Cluster Expansion to magnetic alloys, is applied to modelling magnetic phase transitions, with the emphasis on non-collinear magnetic configurations. We apply the above methodology to multi-component high-entropy alloys, exploring the effect of fluctuating microscopic strains and stresses, associated with the difference between volumes and electro-negativities of the alloying elements, on the phase stability of crystalline versus amorphous phases of the alloys.

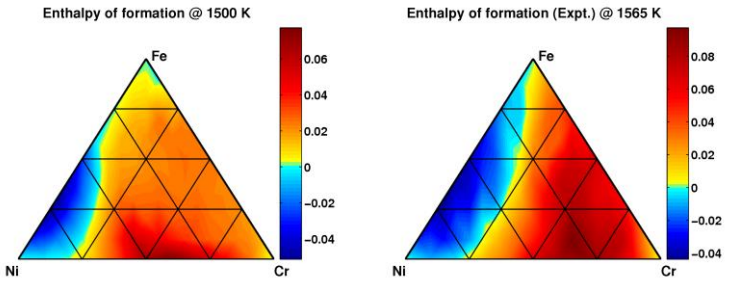

Figure 1: Enthalpies of formation (in eV) for the most stable crystal structures of Fe-Cr-Ni alloys computed using MC simulations at 1500 K (left). Experimental data from Ref. [1] are shown on the right.

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Effect of point defect sinks on irradiation-induced compositional patterning

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The dynamical competition between the chemical mixing forced by energetic particle irradiation and thermally activated decomposition can lead to the stabilization of self-organized steady states in alloy systems comprised of immiscible elements. Continuum modeling and atomistic simulations predicted the stabilization of steady-state nanoscale compositional patterns for a well-defined range of ballistic mixing frequency normalized by the irradiation-enhanced thermal atomic jump frequency. While irradiation-induced compositional patterning has been observed experimentally, a quantitative comparison has been lacking because models and simulations had relied on a simplified treatment with a fixed point defect concentration. We have developed a novel kinetic Monte Carlo code that treats vacancies and interstitials as non-conserved species, including their production, recombination, and elimination on sinks. By controlling the sink density for the point defects to be in the sink dominated elimination regime, steady-state compositional patterns can be stabilized over a much larger temperature range than predicted before. These predictions are tested by comparing the characteristics of patterning in Cu-Ag and Cu-Ag-W alloys, and in particular the maximum irradiation temperature for Ag precipitates to maintain a stable size under irradiation.
In addition, KMC simulations performed on systems with two planar sinks with different sink efficiency reveal that segregation and precipitation under irradiation offer a direct way to measure the relative sink efficiency of these two sinks.

Radiation induced grain boundary flow – Effects of grain boundary segregation

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Irradiation of crystalline structure in conjunction with stress causes the materials to slowly deform ("creep"), resulting in changes to the material's dimensions and strength. We recently demonstrated a new mechanism for irradiation-enhanced creep in nanocrystalline materials. The deformation is due to local atomic relaxations within the grain boundaries as they absorb point defects e.g., interstitials and vacancies, produced in the grain interior during irradiation. Similarities between these relaxation events and "strings" observed in flow of amorphous systems are discussed. We then use atomistic and analytic mean field model to establish the role of immiscible atoms on the grain boundary flow, discussing its effect on the localized relaxation events as well the integral macroscopic flow. We analyze the effects of various segregation scenarios, from isolated dilute gb solution to formation of immiscible precipitates, through their effect on the characteristics of the relaxation events.

Ion-Irradiation Induced Vacancy and Interstitials Clusters in Fe Investigated by X-Ray Diffuse Scattering and by Continuum and Molecular Dynamics Simulations

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We have performed fundamental investigations of vacancy and interstitial defect clusters generated during room-temperature15-MeV Ni-ion irradiation of Fe and Fe-Cr single crystals using x-ray diffuse scattering measurements near Bragg reflections. The measurements were analyzed using continuum anisotropic elasticity simulations of lattice distortions for vacancy and interstitial loops and by molecular dynamics simulations for voids and vacancy clusters as well as for vacancy and interstitial loops. Synchrotron x-ray diffuse scattering measurements were performed near the (002) reflection of <001> oriented Fe and Fe-Cr single crystals and analyzed within the so-called asymptotic regime using continuum and MD based scattering cross-sections. Measurements for irradiations corresponding to one displacement per atom (DPA) at ambient temperature will be presented and discussed in connection with determinations of the presence of vacancy and interstitial type clusters, determinations of the cluster types and orientations, and size distributions using the so-called local Bragg scattering interpretation. The method for calculating diffuse scattering cross sections directly from simulations of the atomic displacements around a/2(111) and a(100) vacancy and interstitial loops and around 3D vacancy clusters and voids.

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Modeling of swelling under electron, ion, and neutron irradiation

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Development of radiation resistance materials requires understanding the controlling degradation mechanisms for structural materials, which can guide the development of a strategy for creating a new generation of the materials. To obtain these data using existing nuclear reactors requires keeping the materials in reactors for ~10-20 years, which is unfeasible. The only timely way to obtain the required data is to use charged particle irradiation since the high doses required can be reached in orders of magnitude smaller times because damage production rates are much higher than those in reactor irradiations. However, the utilization of such data to predict behavior under neutron irradiation, which is the main goal, is not straight forward. High dose rate ion irradiation conditions are qualitatively different from in-reactor irradiation. This situation has been considered in many papers but the analysis used there has been rather qualitative. The main objective of this presentation is to provide a quantitative comparison of damage accumulation in the form of swelling taking place under different types of irradiation. The computational method used here is based on a numerical integration of the master equation for the void size distribution. The calculations have been done using the RIME code developed at ORNL, which is based on a grouping method developed at ORNL as well. The implications of results for comparing data between ion and neutron irradiation is discussed.

Ductility and work hardening in nano-sized and irradiated metallic glasses

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We report results of in-situ nano-tensile experiments performed on 70nm-diameter free-standing NiP metallic glass nanostructures made using a template electroplating technique. These samples exhibit tensile true strains of ~18%, an amount comparable to compositionally-identical 100nm-diameter samples made via focused ion beam (FIB) and ~3 times greater than 100nm-diameter samples produced via the same methodology. Simultaneous in-situ observations and stress-strain data during post-elastic deformation reveal necking and work hardening, features vastly uncharacteristic for metallic glasses. The evolution of free volume distribution within the sample during elastic and plastic deformation revealed by Molecular Dynamics simulations on two 30nm NiAl samples, irradiated and as-cast, shows evidence of a free surface-mediated free volume relaxation mechanism, leading to the observation of a new plasticity mechanism for nano-sized metallic glasses.

Modelling self trapping and trap mutation in tungsten using DFT and Molecular Dynamics with an empirical potential based on DFT

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Being virtually insoluble in metals, but very mobile, He atoms can be trapped, associate with vacancies, forming platelets and bubbles. They have been shown to contribute to swelling, cause intergranular embrittlement and produce roughening and blistering at metal surfaces. Being repelled by the metal atoms, He atoms form stable clusters, which are also mobile. This tendency to form clusters is so strong that when too many He atoms are aggregated together it can be less costly to relieve the strain created by the interstitial elements by the ejection of one or more matrix atoms leading to the formation of one or more Frenkel Pairs (FP), i.e. vacancies and Self Interstitial Atoms (SIAs). When no vacancy is initially present, the He cluster will be trapped by the vacancy it created, in a self-trapping (ST) event; whereas when one or more vacancies are already associated with the He cluster, the same mechanism is referred to as trap mutation (TM) or loop punching, if more than one SIA is created. The metal studied in this work is tungsten, candidate for the divertor and currently under heavy investigations experimentally and theoretically. We have thus investigated the thermodynamics and kinetics of ST and TM using Density Functional Theory (DFT) calculations and Molecular Dynamics with a recently developed potential for W-He adjusted on DFT calculations. The stability of helium-vacancy clusters (He_nV_m) as well as pure interstitial helium clusters in tungsten results from a competitive process between thermal emission of vacancies, self interstitial atoms (SIAs) and helium, depending on the helium-to-vacancy ratio in mixed clusters or helium number in pure interstitial helium clusters and will be presented in this work.

Inert-gas defects and trapping of helium in bcc transition metals: *Ab initio* predictions and thermal desorption spectroscopy validation

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Modelling radiation effects formed through the bombardment of metals and alloys by inert gas ions is important because inert gas defects contribute to embrittlement and swelling of structural and plasma-facing materials, an issue of critical significance for fission and fusion power generation technologies. So far, experimental and theoretical effort has been focused primarily on the combined synergetic effects associated with the simultaneous accumulation of helium and hydrogen, and the effect associated with the incorporation of inert gases has not been systematically studied even though the agglomeration of noble gas atoms in metals and alloys is a well known phenomenon observed in multi-beam ion implantation irradiation experiments.

We investigate the structure and properties of defects resulting from the incorporation of noble-gas atoms (He, Ne, Ar, Kr, Xe) into all the bcc transition metals, including iron, using first-principles density functional theory (DFT) calculations. Helium is a relatively small atom and the scale of He defect energies is smaller than that corresponding to other noble gas atoms. The atom size effect changes the relative stability of tetrahedral and octahedral defects for Ne, Ar, Kr and Xe in comparison with He. There is a remarkable trend exhibited by the binding energy associated with interaction between inert-gas atoms and vacancies, where a pronounced and colossal size effect is observed when going from He to Ne, Ar, Kr, Xe. The origin of this trend can be explained by electronic structure calculations that show that p-orbitals play an important part in distinguishing the last four inert-gas elements from helium, which contains only $1s^2$ electrons in the outer shell. Figure 1 shows DFT predicted binding energies, validated by experimental thermal desorption spectroscopy measurements, of a helium atom trapped by five different defects (the substitutional He atom defect HeV, Ne, Ar, Kr, Xe) in Tungsten [1]

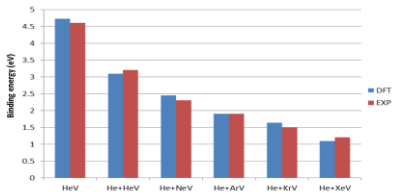

Figure 1: Comparison of predicted values of binding energies between He and various defects (HeV, Ne, Ar, Kr and Xe) with experimental measurements derived from thermal desorption spectrometry of bcc tungsten.

We have also investigated the attachment of He clusters to inert gas impurity atom traps in all the bcc transition metals. The results form a trapping energy database required for modeling helium bubble nucleation in bcc metals and alloys.

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