SYMPOSIUM O

Materials Computation–Progress Towards Technological Impact

April 24 – 27, 2000

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* Invited paper

SESSION O1: CHEMICAL PROCESSES
Chair: Kenneth C. Hass
Monday Morning, April 24, 2000
Golden Gate C1 (Marriott)

8:30 AM *O1.1
MULTI-LENGTH SCALE MODELING OF ATMOSPHERIC SULFIDATION OF COPPER. J.S. Nelson, H.K. Moffat, K.S. Chen, D.M. Teter, R.T. Cygan, A.F. Wright, J.W. Braithwaite and J.C. Barbour, Sandia National Laboratories Albuquerque, NM.

Understanding and developing predictive models for atmospheric corrosion will require new computational and experimental tools capable of describing coupled chemistry and physics phenomena in heterogeneous gas, liquid, and solid phases. These tools will need to span length-scales ranging from electronic (angstroms) to the continuum (microns), and accurately represent important rate limiting processes, including mass transport limited gas-phase diffusion, cation and anion point defect diffusion in metal-oxide-product layer thin-films, adsorption and transport of corrosion species through thin water layers, and reactive chemistry at the liquid-solid interface. In this presentation, I will describe the on-going work at Sandia to develop such a multi-scale coupled physics and chemistry model of corrosion, focusing on sulfidation of copper. The computational simulations (ab initio electronic structure and continuum calculations) are compared to recent micro-combinatorial experiments of copper sulfidation mechanisms.
Sandia is a multi-program laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under Contract DE-AC04-94AL85000.

9:00 AM *O1.2
THERMODYNAMIC AND KINETIC PROPERTIES OF LITHIUM INTERCALATION COMPOUNDS: A FIRST PRINCIPLES INVESTIGATION. Anton Van der Ven, MIT, Dept of Materials Science and Engineering, Cambridge, MA.

Li_xCoO_2 is an important cathode material for rechargeable lithium batteries. During charging and discharging of a lithium battery, lithium ions are removed and reinserted into the CoO_2 host structure. From first principles, we have calculated the phase diagram of Li_xCoO_2 and the lithium diffusivity within the Li_xCoO_2 crystal structure. The phase diagram exposes a variety of stable phases with changing lithium concentration including staged and ordered phases. Phase transformations within the cathode material are undersirable as they are accompanied by large volume changes. The lithium diffusivity within the oxide host structure determines the rate at which lithium can be removed and reinserted into the cathode material. The present investigation shows that the activation barrier for lithium migration is very sensitive to the local lithium configuration within Li_xCoO_2 aswell as to the overall lithium concentration. This results in a diffusion coefficient that varies several orders of magnitudes over the whole lithium concentration range. Both the calculated thermodynamic and kinetic properties have been combined in continuum diffusion and elasticity models to investigate the macroscopic characteristics of the cathode. Even under reasonable charge and discharge rates of the cathode, the present calculations indicate that the cathode material experiences large strains.

9:30 AM O1.3
TOWARDS 5 V RECHARGEABLE Li-ION BATTERY. R.I. Eglitis, M.R. Philpott, H.J. Lindner, Institute of Materials Research and Engineering, Singapore, SINGAPORE.

The predictive power of first principles quantum electronic structure calculations due to increased speed of computers and recent developments of a new and powerful computational methods allow the rational design of new materials for technology applications on paper. One good example is the recent prediction of an average battery voltage for a series of a cathode materials from first principles calculations by Ceder [1,2]. Currently lithium-ion batteries are the state-of-the-art power sources for consumer electronics operating mainly in the 4 V regime. One frequently discussed direction to improve performance is the development of family of 5 V cathodes materials. We report here a Full Potential Linearized Augmented Plane Wave calculation results for a number of materials including $LiCoMnO_4$, $Li_2CoMn_3O_8$, $LiMn_{2-x}Cu_xO_4$, $LiNi_xMn_{2-x}O_4$, $LiNiVO_4$, $LiCr_xMn_{2-x}O_4$ and discuss prospective cathode materials for future 5 V rechargeable Li-ion batteries.
1. G. Ceder, Science **280**, 1099, 1998.
2. G. Ceder, Y.-M. Chiang, D.R. Sadoway, M.K. Aydinol, Y.-I. Yang, and B. Huang, Nature **392**, 694, 1998.

10:15 AM *O1.4
FAILURE MECHANISMS OF THERMAL BARRIER COATINGS FROM FIRST PRINCIPLES. Emily A. Carter, Emily A.A. Jarvis and A. Christensen, Dept. of Chemistry and Biochemistry, UCLA, Los Angeles, CA.

Thermal barrier coatings (TBC's) are composed of thin ceramic films and are used to protect metal components in gas turbine engines from the extreme environment present in a combustion chamber (high temperature, highly oxidizing conditions). Just 1 mm of such a coating allows the operating temperature of the chamber to increase by up to 300 degrees C! The problem with these TBC's is that, after many heating-cooling cycles, they tend to spall off (deadhere). The failure mechanisms are not well understood. Progress on characterizing the relevant bulk ceramic materials and the ceramic-ceramic and metal-ceramic interfaces from first principles density functional theory calculations will be described. Insight into likely causes of failure of the interface will be discussed, as well as some ideas as to how to prevent spallation by redesign of the coatings involved. If this can be accomplished, the impact for fuel efficiency in both aircraft and stationary power plants would be considerable.

10:45 AM *O1.5
FIRST-PRINCIPLES MODELING OF REACTIONS ON OXIDE SURFACES FOR AUTOMOTIVE EXHAUST AFTERTREATMENT. William F. Schneider, Ford Motor Company, Chemistry Dept., Dearborn, MI.

Metal oxide surface are ubiquitous in automotive catalyst systems, both as a high surface area substrate supporting other active catalytic components and often as an indirect or direct participant in catalytic chemistry. An obstacle to the use of some oxide materials in exhaust aftertreatment is their tendancy to form recalcitrant sulfates which block their participation in more desirable chemistry. In this work, plane wave density functional theory and Car-Parrinello molecular dynamics are used to explore the adsorption of sulfur oxides on alkaline earth oxides common in certain NO_x aftertreatment systems. The sulfur oxides are found to be strongly adsorbed even on the low energy (001) surfaces, and to produce a range of adsorbate structures on higher-energy surface steps and kinks. To examine sulfur oxidation on these surfaces, oxygen vacancies and adatoms are characterized and their reactions with sulfur species explored. The results shed light on the mechanism of sulfur poisoning and its competition with other reactions on these oxide surfaces.

11:15 AM O1.6
QUANTUM CHEMISTRY: A POWERFUL TOOL TO STUDY PALLADIUM CLUSTERS AND THEIR REACTIVITY TOWARDS HYDROCARBONS. Valeria Bertani, Carlo Cavallotti, Maurizio Masi and Sergio Carrà, Dipartimento di Chimica Fisica Applicata, Politecnico di Milano, Milano, ITALY.

Most heterogeneous catalysts present a complex surface constituted of small metallic clusters, behaving as active centers, deposited on a support. Of particular interest is the application of supported transition metal clusters for the reactions concerning hydrocarbon transformations. In order to deepen the mechanisms underlying their behavior, an investigation has been undertaken on how particle size and distribution affect the reactivity of metallic clusters with gases. Experimental investigations on clusters structures and energies are still difficult and not exhaustive, but quantum chemistry methods can make up for this lack in analytical techniques. The attention has been focused on palladium. The calculations were performed through the three parameter Becke-Lee-Yang-Parr hybrid (B3LYP) density functional theory, with the Lanl2DZ basis set (ECP2 on core electrons and Dunning/Huzinaga on outer electrons); these particular functional and basis set were chosen because they gave good agreement with the known experimental bond energies of diatomic molecules containing the analyzed atoms. Clusters containing few Pd atoms were considered. Almost regular polyhedrons were obtained: equilateral triangle for Pd_3, tetrahedron for Pd_4, bipyramid with triangular base for Pd_5, octahedron for Pd_6 and bipyramid with pentagonal base for Pd_7. Then an analysis was developed to describe their chemical evolution in the presence of reacting gaseous species, particularly hydrogen and methane. In this work about 40 different H_x-C_y-Pd_z clusters were considered and their structures and energies were determined. On the basis of such information, the transformation of hydrocarbons chemically adsorbed on the clusters has been investigated, by means of a kinetic model in which the progressive loss of hydrogen atoms to form carbonaceous deposits is accounted for. Its relevance in the preparation of highly diluted supported palladium catalysts is discussed.

11:30 AM *O1.7
MODELING AND SIMULATION APPLICATIONS OF TECHNOLOGICAL IMPORTANCE AT MSC. William A. Goddard, III, Tahir Cagin, Siddharth Dasgupta, Mario Blanco, Richard Muller, Nagarajan Vaidehi, Materials and Process Simulation Center, California Institute of Technology, Pasadena, CA.

The Materials and Process Simulation Center (MSC) at Caltech is established in July 1990 as a major resource center to expand the research activities to better emphasize the application of the new methods of theory to critical problems of technological interest. The bjectives of the MSC are to:

Develop new theory, new methods, and new software essential to describe the properties of chemical, biological, and materials systems directly from first principles (without the necessity of empirical data)

Validate these new methods by application to problems well characterized experimentally

Apply the new methods to critical problems faced by scientistDevelop specific engineering models essential to optimation of new materials for performance, economics, or environment by industry by tackling applied problems of key critical to industry s and engineers in industry and academia

Develop solutions to critical bottlenecks for applications of theory, modeling and simulation to development of advanced materials for society

Transfer this technology to industry through collaborations, publications, and workshops

Assist industrial and government laboratories with support and guidance on theory, modeling, and simulation In this talk we will present the status of research, development and applications of multiscale modeling, from from fundamental quantum mechaninics level to engineering models as applied to materials such as metals, ceramics, semiconductors, polymers and biomaterials catalysts, biological systems and will also address the technological impact of various projects.

SESSION O2: ELECTRONIC AND OPTICAL MATERIALS

Chair: Jeffrey S. Nelson
Monday Afternoon, April 24, 2000
Golden Gate C1 (Marriott)

1:30 PM *O2.1
DESIGNING PHOTONIC CRYSTAL STRUCTURES FOR THE CONTROL OF LIGHT. Shanhui Fan, S.G. Johnson, Attia Mekis, Yoel Fink and J.D. Joannopoulos, Department of Physics, MIT, Cambridge, MA.

Photonic crystals, artificial materials that represent a direct optical analogue of electronic semiconductors, offer a new dimension in controling the flow of light. Their capabilities allow for miniaturization of many optical components down to the single-wavelength scale, and may eventually lead to large-scale integration of optical and optoelectronic devices. In the development of photonic crystal technology, computer simulations play a critical role in elucidating the fundamental physics, and in providing direct guidance for experimental efforts in device development. In this talk, I will give some of the latest examples, where we use large-scale computations to design and develop novel photonic crystal components.

2:00 PM *O2.2
THE INVERSE BAND STRUCTURE APPROACH TO THE DESIGN OF MATERIALS WITH GIVEN PROPERTIES.
Alex Zunger and Alberto Franceschetti, National Renewable Energy Laboratory, Golden, CO.

The historic approach to incorporation of materials into technology has usually involved a discovery of a material with interesting properties, followed by the invention of a technology or device that utilizes these properties. Basic research into the physics of this material then followed. A more logical approach would have been to first specify the property that is needed for a given technology,then to search for a material that has this property. Unfortunatly, the current formulation of Electronic Structure Theory of solids is not condusive to this goal, for band theory requires one to first specify the structure of the solid/molecule before its electronic properties can be predicted. We have invented the inverse process: search for a crystal structure (within a given family) that has a prescribed electronic property. For example, in the context of semiconductor materials, one may ask: Which atomic configuration (alloys, superlattices, ordered compounds, etc.) of $Al_{0.25}Ga_{0.75}As$ has the largest possible band gap? (or a given band gap and effective -mass). This is not a simple question for the number of structural possibilities here is truely astronomic. I will describe a practical way how this can be done by combining linear scaling pseudopotential theory with a simulated annealing search strategy. This opens up the way for theoretical Combinatorial Chemistry and thus a new approach to material design. This work is published in the November 4 1999 issue of Nature.

2:30 PM *O2.3
A THEORETICAL STUDY OF THREADING EDGE DISLOCATIONS IN GALLIUM NITRIDE. A.F. Wright and Kevin Leung, Sandia National Laboratories, Albuquerque, NM.

Group-III nitride films grown on sapphire substrates typically contain between 10^8 and 10^{10} threading dislocations per cm^2 due to the substantial film-substrate chemical and lattice mismatch. Nevertheless, it is possible to fabricate highly efficient light-emitting diodes and even laser diodes on these films, whereas these devices would not work at all if fabricated on equally defective films of other III-V compounds. While group-III nitride films are certainly unique, it is not clear whether the dislocations themselves have unusual properties or some other factor (such as alloy fluctuations) is allowing devices to function in spite of the high dislocation densities. In an attempt to answer these questions, we have employed first-principles electronic structure techniques to identify the most likely core structures for threading edge dislocations in GaN and to examine their electronic properties. We find that both background doping levels and growth stoichiometry can influence the dislocation core structure and the resulting electronic properties. Dislocations in n-type material grown under nitrogen-rich conditions, for example, are predicted to have gallium vacancies at the core and to display deep acceptor-like behavior. Furthermore, these dislocations can accumulate a high enough density of electrons that they have measurable effects on lateral transport in GaN films. We will discuss the dependence of charge accumulation on dislocation density, doping level, and growth conditions, both for n-type and p-type films. In addition, we will describe theoretical results pertaining to how hydrogen contamination can affect dislocation core structures and their electronic properties. This work was supported by the U.S. Deptartment of Energy under Contract No. DE-AC04-94AL85000.

3:00 PM O2.4
FIRST-PRINCIPLES CALCULATION OF ARSENIC DIFFUSION MECHANISMS IN SILICON. Scott A. Centoni, Thomas Lenosky, Babak Sadigh, Tomas Diaz de la Rubia, Charles Musgrave.

Arsenic is the most important n-type dopant in ion implantation of silicon for integrated circuit fabrication. It has been found experimentally (Ural et al. 1999) that both oxidation and nitridation of the wafer surface enhance arsenic diffusion in silicon. This is contrary to the behavior of boron, phosphorus, and antimony, and implies that at the temperatures studied (~1000°C) diffusion proceeds almost equally by means of interstitial and vacancy mechanisms. We have examined this problem by means of state-of-the-art first-principles density functional theory (DFT) calculations employing the generalized gradient approximation (GGA) of Perdew and Wang (PW91). Ultrasoft pseudopotentials were used to increase the efficiency of the calculations. Large supercells (128 to 256 lattice points for geometry optimization, 32 for molecular dynamics) were used to reduce self-interaction between point defects and their periodic images. Quantum chemical calculations were performed on large clusters derived from these supercells to ascertain the accuracy of the methods used.

3:30 PM *O2.5
MICROSCOPIC EFFECTIVE HAMILTONIANS FOR FERROELECTRICS FROM FIRST PRINCIPLES. Karin M. Rabe, Dept of Applied Physics, Yale University, New Haven, CT; Philippe Ghosez, Institut de Physique, Universite de Liege, Liege, BELGIUM.

Ferroelectrics and related materials have great potential for application in a variety of devices, including transducers, field-effect devices, and nonvolatile memory, exploiting their spontaneous polarization and dielectric, piezoelectric and pyroelectric properties. In these applications, the relevant properties are observed to be generally very sensitive to electrical and mechanical boundary conditions, as well as to extrinsic effects such as defects and impurities. We have developed a method for constructing a microscopic effective Hamiltonian with parameters obtained from first-principles calculations, including DFPT calculations of the phonon dispersion relation and corresponding real space interatomic force constants of the unstable high-symmetry prototype phase. In this approach, the boundary conditions can be precisely controlled and the intrinsic behavior of the system determined. Simulations with this Hamiltonian allow us to predict the structures and properties of ferroelectrics in bulk, thin films and other confined geometries, and to investigate the statics and dynamics of these systems under applied fields and stresses and at nonzero temperature. An application of this method to the structure and properties of thin films of $PbTiO_3$ will be presented.

4:00 PM *O2.6
THE Si-SiO_2 and the SiC-SiO_2 INTERFACES. S.T. Pantelides, R. Buczko, M. Di Ventra, G. Duscher and S.J. Pennycook, Department of Physics and Astronomy, Vanderbilt University, Nashville, TN and Oak Ridge National Laboratory, Oak Ridge, TN.

The quality of the Si-SiO_2 interface is one of the reasons Si is the semiconductor of choice for microelectronics. Yet, the SiC-SiO_2 interface is not good enough for SiC-based power devices. First-principles calculations have been used to elucidate the differences and similarities between the two interfaces, the nature of the global bonding arrangements, the identity of intrinsic defects resulting from the thermal oxidation process, and possible mechanisms for defect passivation. Atomic-resolution Z-contrast transmission electron microscopy and electron energy loss spectra provide complementary information. This work was supported in part by grants from DARPA, EPRI, AFOSR, ONR, and NSF.

4:30 PM O2.7
ELECTRONIC PROPERTIES OF THE Si/SiO_2 INTERFACE FROM FIRST PRINCIPLES. J.B. Neaton, Cornell Center for Materials Research and the Laboratory for Atomic and Solid State Physics, Cornell University, Ithaca, NY; D.A. Muller, Bell Laboratories, Lucent Technologies, Murray Hill, NJ; N.W. Ashcroft, Cornell Center for Materials Research and the Laboratory for Atomic and Solid State Physics, Cornell University, Ithaca, NY.

The smallest feature of integrated circuits is the gate oxide, a thin silica (a-SiO_2) layer that can be currently grown less than 1.3 nm thick. As the oxide thickness decreases to scales comparable to atomic bond lengths following Moore's Law, atoms at interfaces will comprise much of the oxide for these thin layers; the oxide can no longer be expected to have the electronic properties of bulk silica. To investigate the extent to which the electronic structure of the oxide at the interface deviates from those in the bulk, density functional calculations were performed within the local density approximation (LDA) for a model Si/SiO_2 interface. Calculated local densities of states (LDOS) of oxygen atoms in the model oxide are compared near and away from the interface; their shape and conduction band-edge onsets are shown to be strongly correlated to the number of neighboring oxygens. From analysis of the fourth moment of the LDOS, the magnitude of local energy gap is shown to be proportional to the number of O nearest neighbors around a given O atom. The energy gap of bulk SiO_2 is only obtained for a fully coordinated O-neighbor shell. The calculated local energy gaps of the oxide becomes considerably smaller within ~ 0.5 nm of the interface, suggesting a gradual transition from the electrical properties of Si to those of SiO_2, even for atomically-abrupt interfaces.

4:45 PM O2.8
SOME OPPORTUNITIES FOR MATERIALS MODELING IN MICROELECTRONICS. David F. Richards, Timothy S. Cale, Rensselaer Polytechnic Institue, Troy, NY.

Modeling and simulation of continuum thin film profile evolution has had a substantial positive impact on process understanding in the microelectronics industry. Several key principles have emerged from our experiences in formulating and applying profile evolution models:

- "Engineering level" models that provide information on semi-quantitative trends are often more valuable than fundamental quantitative models.
- First principles models are intellectually appealing but calibration and fitting are usually more timely and easier to use.
- Industrial process engineers require answers quickly. Technological impact is impossible if we cannot work quickly enough to deliver answers when needed.

We will discuss how working within these principles has helped us develop useful models and simulations. We finish by offering some ideas regarding opportunities in materials modeling. To expand the role of modeling and simulation we will need to develop models that are capable of predicting material properties and performance without loosing sight of the above principles.

SESSION O3: GROWTH PHENOMENA
Chair: Emily A. Carter
Tuesday Morning, April 25, 2000
Golden Gate C1 (Marriott)

8:30 AM O3.1
IMPACTING CRYSTAL GROWTH UNDERSTANDING AND PRACTICE VIA HIGH PERFORMANCE COMPUTATION. Jeffrey J. Derby, Department of Chemical Engineering and Materials Science, Army HPC Research Center and Minnesota Supercomputing Institute, University of Minnesota, Minneapolis, MN.

With the continuing advance of hardware and algorithms, high performance computing is proving to be an increasingly useful approach to understand the growth of crystalline materials. On the continuum level, these processes are characterized by complicated, nonlinear interactions between field and interfacial phenomena, specifically the transport of momentum, heat, and mass coupled with solidification, growth kinetics, capillarity, and interface morphology. The use of realistic theoretical models is extending the traditional paradigm of experimental investigation and process development, thereby accelerating the development and optimization of crystal growth processes. In this seminar, an overview is presented of the models and algorithms employed to simulate melt crystal growth and solution crystal growth, especially the challenges of modeling three-dimensional, transient fluid dynamics and coupled transport. Specific examples of increased process understanding with tangible benefits will be discussed for the Bridgman growth of cadmium zinc telluride (CZT). In addition, ongoing work addressing the coupling of disparate scales, from atomistic behavior to mesoscopic phenomena to the continuum, will be discussed in the context of our crystal growth modeling research.
This work was supported in part by NSF, NASA, and Sandia National Laboratories. Support was also provided by the Minnesota Supercomputer Institute and the US Army, Army Research Laboratory, Army HPC Research Center. No official endorsement should be inferred.

8:45 AM O3.2
PREDICTIONS OF ISLAND NUCLEATION: ETCH PITS ON Si(001). M.C. Bartelt, Sandia National Laboratories, Livermore, CA; J.B. Hannon, Carnegie Mellon Univ, Dept of Physics, Pittsburgh, PA; G.L. Kellogg, Sandia National Laboratories, Albuquerque, NM; J.W. Evans, Iowa State Univ, Dept of Mathematics and Ames Laboratory, Ames, IA.

The nucleation and growth of islands during deposition or etching produces subtle correlations between island size and separation, which control the island size distribution [1]. These correlations develop early on, motivating detailed analysis of the island nucleation regime. Once a few islands exist, subsequent nucleation should occur quasi-deterministically near the dominant maximum in the density, n, of the diffusing species (as the nucleation probability scales like n^{i+1} for critical size i). Subsequently, a more complex density landscape develops, and nucleation often occurs more randomly, often near secondary maxima. Deviations from the dominant maximum were shown to affect the shape of the island size distribution [2]. This follows since they allow for stronger correlations between growth rates of neighboring islands [1]. We can illustrate these features most dramatically for smaller few-island systems. We thus compare LEEM observations of actual nucleation positions of two-dimensional vacancy islands or pits, etched on a finite Si(100) terrace by exposure to O_2, with predictions from solving the appropriate diffusion equation for n, and from kinetic Monte Carlo simulations. Finally, for large systems, we characterize the dependence on island size of the probability for islands to nucleate within the capture area of an existing island. This quantity influences the selection of the size-dependence of the island capture areas, and thus the correlations between island size and separation. This work was supported by the USDOE-OBES under contracts No. DE-AC04-94AL85000 (MCB, JBH, and GLK) and No. W-7405-Eng-82 (JWE). [1] M.C. Bartelt et al., PRL 81 (1998) 1901; PRB 54 (1996) R17359. [2] C. Ratsch, M.F. Gyure, S. Chen, M. Kang, D.D. Vvedensky, unpublished.

9:00 AM O3.3
LARGE-SCALE MOLECULAR DYNAMICS SIMULATIONS OF LIQUID METAL WETTING AND SPREADING. Edmund B. Webb III and Gary S. Grest, Sandia National Laboratories, Albuquerque, NM.

The joining of materials via soldering or brazing is a fundamental step in the construction of many technological components. Inherent in these methods is the phenomenon of liquids wetting and spreading on oxide or metal surfaces. While a detailed understanding of the atomistic behavior during such events would aid in the creation of more reliable joins, such an experimental description is lacking. This is a natural area to which molecular simulations can be applied; however, previous attempts at this have been limited by computational capabilities. This is partly due to the geometry of a wetting simulation requiring very large numbers of atoms and to the absence of interatomic potentials which accurately predict metal surface energetics. In this talk we present a thorough evaluation of embedded atom method (EAM) potential sets with regard to their ability to describe metal surfaces. The EAM incorporates a many-body term into the interatomic potential and, as such, has performed better than two-body potentials in describing bulk metallic properties. Since accurate prediction of surface properties is essential for realistic wetting simulations, we have selected EAM potential sets which perform well in their ability to predict the solid free surface

energies and liquid surface tensions of metals. These potentials, along with massively parallel simulation codes, are employed in large scale molecular dynamics (MD) simulations of liquid metal wetting and spreading. Because of increased computational resources, larger systems and longer time scales than in the past have been studied, thereby removing simulation method artifacts. Results reported from these simulations focus on the dynamics of spreading and compositional heterogeneities which develop during spreading. We conclude our talk with a discussion of the computational challenges inherent in the next phase of our study focusing on the role and behavior of active components in solder or braze alloys.

9:15 AM *O3.4
BRIDGING TIME AND LENGTH SCALES IN SEMICONDUCTOR PROCESS MODEL DEVELOPMENT. Mark Gyure, HRL Laboratories, Malibu, CA.

The development of new device materials is currently dominated by costly and time consuming experimental investigations in which growth parameters are varied and correlated, *ex situ*, to device characteristics. Model-based process development has the potential to assist greatly in reducing development time, optimizing growth parameters and enabling *in situ* process control, but requires robust, predictive models. In this presentation, I will describe two models for the MBE growth of III-V semiconductor materials that form part of an integrated modeling hierarchy describing the growth process on different time and length scales. The first model uses a combination of *ab intio* calculations together with *in situ* STM data as input to detailed kinetic Monte Carlo (KMC) simulations. These simulations capture the effects of surface reconstructions and group V fluxes and provide the insight necessary to construct models valid on much longer length and times scales. The second model is a continuum model for layer by layer growth that utilizes level set methods to provide a description of epitaxial growth that is continuous in lateral directions, but retains the discreteness in the growth direction needed to address atomic scale interface roughness. Detailed comparisons between the results of this model and the results of KMC simulations show excellent agreement between experimentally relevant quantities such as island size distributions and step edge densities.

SESSION O4: MAGNETIC PHENOMENA
Chair: Bruce N. Harmon
Tuesday Morning, April 25, 2000
Golden Gate C1 (Marriott)

10:00 AM *O4.1
METALLIC MAGNETISM: INHOMOGENEITIES, NON-COLLINEARITY, AND SPIN DYNAMICS. G. Malcolm Stocks, Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN.

Although a subject of great venerability, enormous challenges still remain in understanding, calculating, and predicting the properties of magnetic materials. This is particularly true of such technologically important systems as magnetic multi-layers and technically useful properties such as permeability, coercivity, and remenance. In this presentation, I will review recent progress in developing first principles methods for calculating the magnetic properties of homogeneous and non-equilibrium metallic magnets. I will discuss advances in the development of first principles techniques for calculating the properties of large numbers (hundreds to thousands) of atoms using parallel computers that make these studies possible. Finally, I will outline recent progress towards developing a first principles theory of the finite temperature and non-equilibrium properties of metallic magnets based on spin dynamics. Work supported by Office of Basic Energy Sciences, Division of Materials Science and Office of Computational and Technology Research, Mathematical, Information, and Computational Sciences Division, US-DOE, under subcontract DEAC05-96OR22464 with Lockheed-Martin Energy Research Corporation.

10:30 AM *O4.2
SPIN-ORBIT COUPLING INDUCED MAGNETIC PHENOMENA IN ALLOYS, SURFACES AND INTERFACES. Ruqian Wu, Dept of Physics and Astronomy, California State University, Northridge, CA.

First-principles electronic structure studies based on local spin density functional theory and performed on extremely complex simulations of ever increasingly realistic systems, play a very important role in explaining and predicting surface and interface magnetism. This talk will review our progresses on a major issue for first-principles theory, namely the theoretical/computational treatment of the weak spin-orbit coupling (SOC) in magnetic transition metal systems and its important physical consequences: magneto-crystalline anisotropy, magnetostriction, magneto-optical effects. Extensive first-principles calculations and model analyses now provide simple physical insights and guidelines to search and design new magnetic recording and sensor materials.

11:00 AM *O4.3
THEORETICAL SEARCH FOR NEW HIGH PERFORMANCE RECORDING MEDIA. R.H. Victora, University of Minnesota, Dept of Electrical and Computer Engineering, Minneapolis, MN.

Two examples of materials computation directed to materials development are presented. In the first case, ab initio electronic structure calculations, predicting magnetic anisotropy, Kerr effect, and magnetization, were used to search for potential magneto-optic recording media. Results suggested that Tb/Bi/FeCo and Tb/Pb/FeCo superlattices would offer a large Kerr rotation and the necessary perpendicular anisotropy. Subsequent experimental fabrication demonstrated that they offer Kerr rotation and figure of merit (reflectivity times Kerr rotation) substantially exceeding that offered by conventional TbFeCo recording materials, particularly at blue wavelengths. For example, 0.44 nm Tb/0.2nm Bi/0.5nm $Fe_{0.6}Co_{0.4}$ has figures of merit 0.20, 0.18, and 0.14 at wavelengths of 780, 650, and 430 nm respectively. The superlattice displays perpendicular anisotropy and squareness suitable for magneto-optic recording. Dynamic testing with 780 nm and 490 nm light demonstrates that these high figures of merit translate to carriers several dB above those produced by comparably sensitive TbFeCo alloys. Furthermore, very high carrier-to-noise ratios such as 60 dB at long mark lengths (1 MHz) are obtained using 490 nm light and a bandwidth of 30 kHz. In the second example, micromagnetic simulation is used to generate a correspondence between intergranular exchange coupling and demagnetized correlation length for novel Co/Pt perpendicular magnetic recording media. Previous workers have shown a tight correlation between exchange coupling and transition noise: a leading barrier to very high density magnetic recording. Therefore, the theoretical work provides an easily measured quantity (the correlation length) to screen samples before making more difficult recording measurements. Subsequent preliminary measurements suggest that Co/Pt superlattices with short correlation lengths are largely free of transition noise and potentially offer superior performance at very high densities. Portions of this work were supported by the NSIC/EHDR project, the NSIC/ARPA UHDR project, and a U. Minnesota Grant-in-Aid. Parts of this work were performed by collaborations at Eastman Kodak, IBM, and U. Arizona.

11:30 AM *O4.4
MODELING THERMAL DECAY IN MAGNETIC MEDIA WITH LINEARIZED DYNAMICS AND MONTE CARLO METHODS. Byron Lengsfield and Oksana Chubykalo, IBM Almaden Research Center, San Jose, CA.

Micromagnetic simulations employing free-energy minimization and the hybrid-finite element method of Koehler and Fredkin {IEEE 28, 1239, 1992} were used to study the role of exchange in designing high density, thermally stable magnetic media with low SNR. The effect of finite-temperature was included in these calculations by determining energy barriers along Monte Carlo trajectories which are then used to determine rate constant input to Kinetic Monte Carlo calculations {Lu and Charap, J. App. Phys. 75, 5768, 1994}. Magnetization decay curves for times up to a millisecond were also determined with the linearized LLG/Monte Carlo method {Smirnov-Rueda, Chubykalo, Chantrell and Gonzalez, IEEE Trans Magn., 1999} and compared to the Kinetic Monte Carlo results.

SESSION O5: ACCELERATING THE IMPACT OF MATERIALS COMPUTATION
Chair: Ellen B. Stechel
Tuesday Afternoon, April 25, 2000
Golden Gate C1 (Marriott)

1:30 PM O5.1
MODELING AND SIMULATION OF MATERIALS AND PROCESSES: BENEFITS AND NEEDS. Jim Williams, Honda Professor, Department of Materials Science and Engineering, The Ohio State University Columbus, OH.

The mechanical design community has demonstrated the ability to shorten the product design cycle by as much as 100% through the development and use of computational design tools. Today, the development cycle for a new material or process has not benefited in a similar manner that product design has from the use of computational methods. As a result, the rate of introduction of new materials technology into products has not kept pace with the rate of introduction of new product designs in the marketplace. Yet, materials are broadly recognized as a key enabling technology for

competitive products. In an integrated product design and production environment, this technology lapse or disconnect must be addressed and resolved before new products can derive the full benefit of materials technology and the most competitive products can be realized. This talk will discuss the current situation and make some suggestions regarding ways to fix the time lag between new designs and new materials technology. It will be argued that the most promising solution is to compress the materials technology cycle in a similar way to what has been done in design. It also will argued that better, more representative computational methods stand the best chance of achieving the necessary time compression. The talk will close by outlining some challenges to the development and implementation of computational materials technology tools and offer a few examples of areas where major benefit could be derived.

1:45 PM O5.2
SINNERS REPENT! IMPACT IS NEAR. G.B. Olson, Northwestern University, Evanston, IL.

At the end of the 2nd Millenium A.D., industrial materials development laboratories are beset by a flood of massive budget cuts brought by the sin of superficial empiricism which has excluded the materials profession from modern engineering. Meanwhile, the academic materials community has been ravaged by a plague of fad funding and false relevance bringing forth a pestilence of pointless modeling activities which dissipate much-needed resources. Salvation lies in a commitment to true Materials Design, whereby the efficient creation of value motivates the development of purposeful models and the systems to integrate them. The way has been shown by the successful multiscale computational design of advanced alloys, exploiting the predictability of designed systems to accelerate the development cycle. Further acceleration demands integration of process scale in design, and the development of a predictive probabilistic science of materials to further reduce necessary testing.

2:00 PM O5.3
ACCELERATING THE IMPACT OF MATERIALS SIMULATION ON US INDUSTRY. Sharon C. Glotzer, Center for Theoretical and Computational Materials Science and Polymers Division, National Institute of Standards and Technology, Gaithersburg, MD.

Computational materials research has made major advances over the past several years in accelerating the design, processing and performance of real technological materials. As we enter the 21st century, materials simulation promises to play an increasingly important role in guiding materials R&D. Despite great successes, however, substantial barriers to progress remain that must be addressed for materials computation to become a regular part of day-to-day R&D in all industry sectors. We address several of these barriers, focusing on those pertaining to the use of materials simulation by the chemical industries in the US and abroad.[1,2] Several government-sponsored programs have taken a first step towards eliminating these barriers to progress and accelerating advances in computational materials science. Here we describe one such program, the Center for Theoretical and Computational Materials Science at the National Institute of Standards and Technology (NIST)[3], and discuss future opportunities for the computational materials research community.
[1]DOE/OIT Vision 2020 Technology Roadmaps on Computational Chemistry (1999) (http://justice.chem.purdue.edu/v2020/compchem.pdf) and Materials Modeling and Prediction (in preparation), Council for Chemical Research.
[2]International Comparative Study on Applying Molecular and Materials Modeling (http://www.ecs.umass.edu/che/amm99.html).
[3]http://www.ctcms.nist.gov

2:15 PM O5.4
THE COMPUTATIONAL MATERIALS SCIENCES NETWORK. Bruce Harmon, Department of Physics and Astronomy, Iowa State University and Ames Laboratory-USDOE, Ames, IA.

Earlier this year the Materials Sciences Division of the Department of Energy's office of Basic Energy Sciences established the Computational Materials Sciences Network. Besides recognizing the remarkable progress and promise in this field, the network was created to bring together researchers to focus on significant problems requiring intellectural resources not available at any one institution. Modest funding is used to assemble and foster interactions among members of interdisciplinary teams from universities, national laboratories, industry, and other government agencies. A theme in many of the proposed projects is the bridging of length scales from atomistic to mesocopic. Scientists with expertise in first principles calculations are working with those dealing with larger scale phenomena such as dislocations, grain boundaries, or magnetic domain walls. Other projects involve polymer interface simulations, and excited state properties. A discussion of the Network's brief history and its future plans as well an outline of each of the ongoing projects will be presented.

2:30 PM O5.5
EFFECT OF STRESS ON DOPANT DIFFUSION IN SILICON: FROM AB-INITIO CALCULATIONS TO ENGINEERING APPLICATION. Murray Daw, Wolfgang Windl, Matt Laudon and Neil Carlsson, Computational Materials Group, Motorola, Austin, TX and Los Alamos, NM.

The introduction of new back-end materials, as well as the further scaling of silicon device dimensions, has raised the level of stress in device structures. Current engineering simulations of diffusion neglect the direct effect of stress on diffusivity. In this talk, we investigate the effects of stress on diffusion of B in Si. The macroscopic diffusion equation is derived from microscopic transition state theory, relating the diffusivity to the microscopic jump parameters; the approach is useful even for complicated defects. The required microscopic parameters are calculated from first principles. For the latter, special care has been taken to minimize the errors introduced by finite-size supercells and the known band-gap problem in LDA calculations. These numbers are implemented into a continuum model and used to examine diffusion in a stress field.

3:15 PM O5.6
COMPUTATIONAL MATERIALS SCIENCE AS TOOL FOR INNOVATION. Erich Wimmer, Materials Design S.A.R.I. and Institute Supérieur des Matériaux du Mans (ISMANS), Le Mans, FRANCE.

Progress in computational methods combined with the dramatic advances in computer hardware and software architectures offer unprecedented opportunities for materials research. In fact, computational materials science has the potential for enabling true innovation and perhaps even breakthrough discoveries. The impact of these technologies on the design, processing and performance of industrial materials can be accelerated by meeting the following requirements: (1) computer simulations need to be combined with databases and used as such by entire R&D project teams, (2) the capabilities need to be delivered in the form of reliable and well supported software systems which allow automatic calculations on a large number of systems, (3) scientists and engineers need to be trained in applying these tools, (4) theoretical and computational methods should allow tuning of accuracy vs. speed over a wide range. At present, there is a serious imbalance between the potential impact of simulation/information technology in materials research and the actual level of industrial investment. This contribution will conclude by showing how various industrialized nations respond to this challenge.

3:30 PM O5.7
THE IMPACT OF ADVANCED AB-INITIO DENSITY-FUNCTIONAL CODES: FROM GEOPHYSICS TO CATALYSIS. Jürgen Hafner, Institute for Materials Physics and Center for Computational Materials Science, University of Vienna, Wien, AUSTRIA.

The state-of-the-art of advanced codes for performing *ab-initio* density-functional total-energy calculations and molecular dynamics simulations is discussed. It is shown that plane-wave-based calculations using either ultrasoft pseudopotentials or the projector-augmented-wave (PAW) approach allow to treat systems with a few hundreths of atoms in static calculations and with more than hundred atoms in dynamical simulations spanning several dozens of picoseconds, while matching the accuracy of all-electron full potential calculations. Transition-metals, rare-earth elements and first-row elements may be treated with a computational effort comparable to that needed for s,p-bonded elements. Recent applications ranging from catalysis (hydrodesulfurization on MoS_2 based catalysts, alkanes in zeolites), over magnetic materials (thin-film reconstructions driven by magnetic effects) to geophysics (high-pressure phase transitions in silica, melting of iron under ultrahigh pressures) are reviewed.

3:45 PM PANEL DISCUSSION
PANEL:
Jim Williams - Ohio State University
Greg B. Olson - Northwestern University
Sharon C. Glotzer - National Institute of Standards & Technology
Bruce N. Harmon - Iowa State University & Ames Laboratory
Murray S. Daw - Motorola
Erich Wimmer - Materials Design s.a.r.l.
Juergen Hafner - University of Vienna

SESSION O6: ALLOYS
Chair: Gerbrand Ceder
Wednesday Morning, April 26, 2000
Golden Gate C1 (Marriott)

8:30 AM *O6.1
APPLICATION OF COMPUTATIONAL THERMODYNAMICS TO TECHNOLOGICAL MATERIALS. N. Saunders, Thermotech Ltd., Surrey Technology Centre, The Surrey Research Park, Guildford, Surrey, UNITED KINGDOM.

Computational thermodynamics has, in the past, often been seen as a somewhat abstract subject, applciable mainly to simple systems but not to so-called real materials of technological impact. This position may have been true in the past but recent advances in models, development of thermodynamic databases and application software has seen the role of computational thermodynamics increase in practical aplications. The present paper will provide a brief background to what has become known as the CALPHAD (CALculation of PHAse Diagrams) method and give specific examples of how modelling has provided significant impact to practical technological problems. These examples will include alloy development, the use of CALPHAD methods as a substitute for experimental measurement in casting simulations and design of alloy specification limits for materials manufacture.

9:00 AM *O6.2
DILUTE SOLUTION MODEL FOR MULTI-COMPONENT ALLOY SYSTEMS. Chris Woodward*, Air Force Research Laboratory, Wright Patterson AFB OH. *Materials Research Division, UES Inc, Dayton, OH.

Alloy designers manipulate the chemistry and microstructure of alloy systems to achieve a balance of properties. In order to optimize strength, creep resistance and toughness a typical structural material will have more than eight components and incorporate two or more phases. On the other hand, computational methods (e.g. electronic structure, atomistics, continuum methods) developed in the solid state and the materials physics community are best suited for simple metals or stoichiometric single crystals. The dilute solution model offers a way of using the results of these methods to examine the equilibrium defect concentrations of multi-component and multi-phase systems. A number of groups are using this technique to study pacing structure-property relationships in technologically relevant materials. Examples of this method will be presented for the site selection of ternary additions in ordered intermetallics, phase partitioning of ternary additions, segregation to grain boundaries and trends in phase stability. Strengths and weaknesses of the model will be reviewed and strategies for improving the technique will be outlined.

9:30 AM O6.3
AUTOMATING PHASE DIAGRAM CALCULATIONS. Axel van de Walle, Anton van der Ven, Gerbrand Ceder, Dept of Materials Science and Engineering, MIT, Cambridge, MA.

Although the formalism that allows the calculation of solid state phase diagrams from first principles is well established, its practical implementation remains a tedious process. The developement of a fully automated algorithm to perform such calculations serves two purposes. First, it will make this powerful tool available to large number of people outside of the alloy theory community. Second, it frees the calculation process from arbitrary parameters, guaranteeing that the results obtained are truly derived from the underlying first-principles calculations. The proposed algorithm formalizes the most difficult step of phase diagram calculations, namely the determination of the cluster expansion, which is a compact representation of the configurational dependence of the alloy's energy. This is traditionally achieved by a fit of the unknown parameters of the cluster expansion to the energy of a set of structures. We present a statistical basis for the selection of both the number of parameters in the cluster expansion and the number of structures to be included in the fit. The algorithm proceeds by gradually improving the precision of the cluster expansion, either by adding new structures or new parameters, until a given accuracy is reached. Applications to the phase diagram calculation of technologically relevant battery compounds are presented.

9:45 AM *O6.4
A DISCONTINUOUS GALERKIN MODEL FOR PRECIPITATE NUCLEATION AND GROWTH IN ALUMINUM ALLOY QUENCH PROCESSES. N. Sobh, J. Huang, L. Yin, R.B. Haber, D.A. Tortorelli, Center for Process Simulation and Design, University of Illinois at Urbana-Champaign, Urbana, IL; R.W. Hyland Jr. Reading Alloys Inc., Robesonia, PA.

This paper presents a finite element model that treats precipitate nucleation and growth during the quenching phase of aluminum alloy manufacturing processes. A discontinuous Galerkin model for steady advection-diffusion problems predicts the thermal response in a continuous quench process. The thermal history drives a precipiate evolution model, based on a discrete representation of the particle size distribution in each local material neighborhood. This approach can require as many as 10^5 degrees of freedom per spatial location. A second discontinuous Galerkin finite element procedure is presented to solve this seemingly massive problem. The new method scales linearly in both the number of elements and in the number of precipitate degrees of freedom per location. Thus it is feasbile to directly embed the discrete precipitate evolution model in a macroscopic process simulation. Numerical examples demonstrate the effectiveness of the quench model and the feasiblity of studying microstructure development as a function of the quench process employed.

SESSION O7: ADVANCED METHODS
Chair: Ellen B. Stechel
Wednesday Morning, April 26, 2000
Golden Gate C1 (Marriott)

10:30 AM *O7.1
CRYSTALS, DEFECTS AND MICROSTRUCTURES: A MULTISCALE CHALLENGE. Rob Phillips, Brown University, Division of Engineering, Providence, RI.

The modeling of material response in some cases requires insights from several length scales simultaneously. Some recent work has been aimed at constructing models in which degree of freedom elimination has been carried out systematically resulting in a reduction in computational overhead and allowing for accompanying physical insight. Two recent examples of this type of thinking are the quasicontinuum method and dislocation dynamics. In this talk I will describe the use of both quasicontinuum arguments and dislocation dynamics as a way of exploring the properties of defects in solids. Special attention will be given to the interactions of these defects both with each other and with other defects such as precipitates.

11:00 AM *O7.2
ACCELERATED MOLECULAR DYNAMICS SIMULATION OF COPPER FILM GROWTH–PROGRESS TOWARDS ACHIEVING EXPERIMENTAL DEPOSITION RATES. Arthur F. Voter and Timothy C. Germann, Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM.

Applying recently developed methods for accelerating the dynamics of infrequent events, we study the growth of copper on a Cu(100) face. While molecular dynamics simulations are limited to nanoseconds, these methods (hyperdynamics and parallel replica dynamics) allow us to reach the much longer time scales necessary for the activated processes between deposition events, without any presumptions about the microscopic mechanisms. We use an embedded atom method (EAM) interatomic potential that was fit to bulk and diatomic properties, but which has been found to give a good description of surface diffusion barriers. Each depositions event, either thermal or hyperthermal, is simulated using regular molecular dynamics for a few ps. Hyperdynamics combined with parallel replica dynamics are employed for the remainder of the time until the next deposition. With a 72-atom-per-layer simulation cell, we have been able to grow at deposition rates as slow as 1 monolayer per 72 microseconds. We observe rough growth below about 300K, and the onset of reentrant layer-by-layer growth as the temperature is raised. We find the activated events are dominated by nonstandard mechanisms, such as interlayer smoothing via 3-atom exchange and 3- and 4-atom row sliding. As expected, hyperthermal deposition leads to smoother growth.

11:30 AM O7.3
ELECTRONIC STRUCTURE CALCULATIONS USING LARGE SCALE PARALLEL COMPUTERS. Lin-Wang Wang, Andrew Canning, National Energy Research Scientific Computing Center (NERSC), Lawrence Berkeley Lab, Berkeley, CA.

NERSC is the biggest unclassified scientific computing center in the United States, with a 560 processor T3E computer and a SP3 IBM parallel computer. We have developed efficient parallel algorithms and programs to run on such platforms for ab initio (density functional theory) and empirical electronic structure calculations. The capabilities of such massively parallel computations will be presented through a few case studies. The program is implemented in a highly modular way with the intention of being used by other research groups. The ease of use of the programs will be demonstrated. We will also discuss possible impacts of such computations and computational codes in technologies and in material research societies.

11:45 AM O7.4
EFFICIENT DENSITY FUNCTIONAL CALCULATIONS FOR HUNDREDS OF ATOMS. Andrew Horsfield, Fujitsu European Centre for Information Technology, Uxbridge, Middlesex, UNITED KINGDOM; Steven Kenny, Department of Materials, University of Oxford, Oxford, UNITED KINGDOM; Hideaki Fujitani, Fujitsu Laboratories Ltd., Atsugi, JAPAN.

Accurate calculations of phenomena in materials often require more than a few tens of non-equivalent atoms be treated explicitly. For all their success, density functional calculations with a plane-wave basis set become very expensive for systems containing 100 atoms or more. This is a direct consequence of the choice of basis set. Much greater efficiency can be achieved for large systems by moving over to a localised basis set. Here we present a procedure for optimizing atomic like basis sets, and demonstrate the level of convergence and transferability that can be achieved with them in the solid state by applying them to a range of materials (silicon, carbon and aluminum) in a number of configurations. We find that it is straightforward to obtain the level of convergence used in typical plane-wave computations, and that this approach can be applied to systems containing 512 on a single workstation. This allows us to treat a number of systems that are of great interest from a materials science point of view. We will illustrate this by applying this method to the isolated interstitial and to dislocation kinks in silicon.

SESSION O8: MICROSTRUCTURE
Chair: Stephen M. Foiles
Wednesday Afternoon, April 26, 2000
Golden Gate C1 (Marriott)

1:30 PM O8.1
A LATTICE-BASED CONTINUUM FORMULATION FOR COUPLED COMPOSITION-MECHANICS IN POLYCRYSTALLINE SOLIDS-THEORY AND COMPUTATION. Krishna Garikipati, Lori Bassman, Michael Deal, Stanford University, Center for Integrated Systems, Stanford, CA.

This work represents a step toward the development of a continuum field formulation for the coupled phenomena of diffusion and mechanics in polycrystalline solids. The basis of the formulation lies in crystal lattice-level mechanisms that are the building blocks of a continuum thermodynamic description applicable to processes at micron length scales. With self-diffusion in mind, the composition problem is posed in terms of a binary vacancy-atom mixture. The mechanics is based on isotropic linear elasticity and isothermal conditions are assumed. The coupled constitutive relations for composition and mechanics are formally derived from the underlying thermodynamics. When applied to governing partial differential equations for each sub-problem, the fully coupled nature is realized. Under applied tractions or intrinsic stress, the atoms diffuse—in general from regions of compressive normal traction to those with relatively tensile normal tractions. The flow is also mediated by electric fields via the mechanism of electromigration. In the case of metal interconnect lines in integrated circuit devices, the results are phenomena such as diffusional creep, hillock formation, grain growth, grain boundary motion, void formation and void evolution. These phenomena have a significant impact on the function, performance and failure of the metal lines. A computational framework based on the Finite Element Method has been developed to solve the coupled equations. Special finite element techniques have been developed to stabilize the equations, to treat coupling terms and resolve high gradients. Numerical examples of academic and technological interest are presented and comparisons with analytical results are provided where the latter are available.

1:45 PM *O8.2
INTEGRATED KINEMATIC AND KINETIC YIELD SURFACE MODELING. Dierk Raabe, Max-Planck-Institut, Duesseldorf, GERMANY.

The paper presents a novel approach for the integrated modeling of the polycrystal yield surface. The method is based on integrating physically based kinematic and kinetic models for the description of the yield surface. While the kinematic part is based on Taylor-Bishop-Hill type theory the kinetic part is based on orientation-sensitive constitutive dislocation theory.

2:15 PM *O8.3
PROPERTIES OF MICROSTRUCTURES: DIRECT CALCULATIONS USING *OOF*. W. Craig Cater, MIT, Dept. of Materials Science and Engineering, Cambridge, MA; Stephen A. Langer, Edwin R. Fuller, National Institute of Standards and Technology, Gaithersburg, MD.

Many important materials properties depend on microstructure. We describe a public domain program which we have written which allows direct computation of thermoelastic properties from microstructural images, materials properties, as well as other types of spatial data. Especially when properties depend on extreme value interpretations of microstructures, methods of homogenizing microstructures may give misleading results. The code, called OOF, calculates the aggregate response of a microstructure by correlating all possible spatial details with materials property data. Example applications on thermal spray coatings, reliability estimates and quantitative assessment of microstructural response are presented.
http://www.ctcms.nist.gov/oof/

3:15 PM O8.4
THREE-DIMENSIONAL COMPUTER SIMULATIONS OF GRAIN SIZE CONTROL VIA PARTICLE PINNING. Gregory N. Hassold, Elizabeth A. Holm, Sandia National Laboratories, Albuquerque, NM; Mark A. Miodownik, University College, Dublin, IRELAND.

While grain growth inhibition by immobile particles is of great industrial significance for structural metals and ceramics, the fundamental physics of particle pinning has been debated for fifty years. The Smith-Zener relationship between pinned grain size and particle fraction remains the prevailing theory; however, both experiments and computer simulations contradict the Smith-Zener volume fraction scaling. Our recent studies have shown that earlier computer simulations of three-dimensional particle pinning were performed under thermodynamic conditions which induce artificial lattice pinning and yield too small a final grain size. Because the correct thermodynamic conditions mandate very large system sizes, we developed a high performance Monte Carlo simulation for three-dimensional grain growth in the presence of rigid, immobile particles. Results confirm the volume fraction scaling predicted by Smith and Zener. In contradiction with some theories, no change in volume fraction exponent is observed for large particle fractions. Grain growth kinetics and size distributions are also discussed. In light of this confirmation of theory, we offer some explanations for the conflicting experimental results. Application of this model to industrially significant systems, including systems containing mobile particles, will be proposed.

3:30 PM *O8.5
MONTE CARLO SIMULATION OF GRAIN GROWTH WITH A 5-PARAMETER DESCRIPTION OF BOUNDARY PROPERTIES. Anthony D. Rollett, Carnegie Mellon University, Materials Science & Engineering, Pittsburgh, PA.

As progress is being made on the characterization of grain boundary properties as a function of their crystallographic character, so it is important to incorporate these properties into simulation codes. Simulation of microstructural evolution has typically assumed either uniform, isotropic boundary properties or a simplified form of anisotropy. In most materials, however, the boundary properties exhibit a complex dependence on their crystallographic character. To account for this anisotropy the Potts model (Monte Carlo) for simulation of recrystallization and grain growth has been modified to include a five parameter description of grain boundaries. The five parameter description accounts for the known dependence of properties such as energy and mobility on both misorientation and inclination. Quantitative descriptions of boundary properties are drawn from both experimental and theoretical work. Simulations of recrystallization that illustrate the importance of boundary character are described. Issues relating to the numerical representation of complex properties are discussed. For example, the incorporation of inclination results in significant additional computational effort because of the need to fit lines or planes to boundary segments.

4:00 PM O8.6
PHASE FIELD MODELING OF GRAIN GROWTH. James Warren, Center for Theoretical and Computational Materials Science, NIST, Gaithersburg, MD; Ryo Kobayashi, Hokkaido University, Sapporo, JAPAN; W. Craig Carter, MIT, Cambridge, MA.

A phase field model of grain coarsening will be presented which has two novel features: a finite set of physically motivated order parameters (as few as two), and grain boundary evolution not only by motion by curvature but also by rotation. Analytic solutions will be discussed, development from microscopic models described, and simulations in 1 and 2 dimensions presented.

4:15 PM *O8.7
THE EMERGING ROLE OF CRYSTALLOGRAPHY IN MICROSTRUCTURAL ANALYSIS AND CONTROL. Elizabeth A. Holm, Gregory N. Hassold, Corbett C. Battaile, Sandia National Laboratories, Albuquerque, NM; Mark A. Miodownik, University College, Dublin, IRELAND.

In polycrystals, crystallography determines both the grain orientation distribution (texture) and the grain boundary misorientation distribution (boundary character). Traditional metalforming processes, including deformation and annealing, have long been used to influence both texture and boundary character in order to optimize properties. Recent innovations in processing (grain boundary engineering), analysis (orientation imaging microscopy), and physical understanding (boundary property measurements) enable new models for the development and consequence of texture and boundary character. To incorporate crystallography into microstructural scale computer models, we have developed a Monte Carlo procedure that maps experimental crystallographic measurements onto realistic, three-dimensional microstructures. When applied to systems of deformation-induced subgrains, the model not only translates 2D measurements into 3D structures, but also elucidates a physical mechanism for subgrain formation and the observed universal misorientation scaling. In order to evolve these microstructures, we have implemented a cellular automata model for grain boundary motion driven by interfacial and bulk energy minimization. When coupled with boundary energy and mobility measurements from analysis, experiments, and molecular dynamics, the grain growth simulation allows development of texture, boundary character, and grain morphology as a function of crystallography. By incorporating the resulting boundary character in a network analysis algorithm, we examine properties such as corrosion which are mediated by the connectivity of special boundaries formed during grain boundary engineering. By implementing texture into a polycrystalline deformation simulation, we derive relationships between texture, mechanical response, and subsequent microstructural evolution. The relevance of these results to industrial metalforming processes will be discussed.

4:45 PM O8.8
MESOSCALE SIMULATION OF TEXTURE EVOLUTION IN PARTICLE-CONTAINING ALUMINUM ALLOYS. Bala Radhakrishnan, Gorti Sarma, Oak Ridge National Laboratory, Oak Ridge, TN; Hasso Weiland, Paul Baggethun, Alcoa Technical Center, Alcoa Center, PA.

Coarse hard particles significantly influence the development of microtexture during thermo-mechanical processing of industrial alloys through their ability to nucleate recrystallized grains of unique (random or non-preferred) orientations that exist in their deformation zones. The overall recrystallization texture depends on the growth competition between the particle stimulated nuclei and those that form in the particle-free matrix, and at grain boundaries and triple junctions. The recrystallization texture plays a significant role in controlling the forming behavior of aluminum alloys. Although Taylor calculations coupled with "slip-shadowing" concepts can provide estimates of local lattice rotations near hard particles for a given deformation, they cannot quantify the spatial distributions of lattice rotations that exist near the particles. The lattice orientations as well as the orientation gradients existing in the vicinity of the particles determine the evolution of the recrystallization texture. The paper describes a simulation approach that uses finite element calculations based on crystal plasticity theory to predict the evolution of deformation texture during plane strain compression of an aluminum single crystal containing a hard particle when the crystal is initially at Goss orientation. The output of the finite element simulations is used to construct a deformation substructure in the form of subgrains. The heterogeneous evolution of the deformation substructure is then simulated using a Monte Carlo technique to capture the texture evolution during recrystallization. The textures predicted by the simulations compare favorably with experimental results obtained through TEM based on orientation imaging microscopy (OIM). The computational challenges that need to be overcome before the simulations can be applied to the industrial processing of polycrystalline alloys are discussed.

SESSION O9: POSTER SESSION:
MATERIALS COMPUTATION:
PROGRESS TOWARDS TECHNOLOGICAL IMPACT
Chairs: Kenneth C. Hass and Ellen B. Stechel
Wednesday Evening, April 26, 2000
8:00 PM
Salon 1-7 (Marriott)

O9.1
MODELLING OF MECHANICAL PROPERTIES OF TITANIUM ALLOYS USING ARTIFICIAL NEURAL NETWORK. Zhanli Guo, Savko Malinov and Wei Sha, The Queen's University of Belfast, School of Civil Engineering, Belfast, UNITED KINGDOM.

Abstract It has been a long term interest to understand the relationship between the mechanical properties of alloys and their composition together with processing parameters. Statistical analysis shows its advantage when the physical background of property control has not been clarified. Artificial neural network (ANN) data analysis technique has been used in the present work to investigate the connection between alloying elements and ultimate tensile strength (UTS) of conventional titanium alloys. The influence of heat treatment condition on several mechanical property parameters has also been studied with Ti-6Al-6V-2Sn alloy as an example. These models have shown very good performance in property prediction. Moreover, this modelling technique can be extended to optimize the complex material processing route and consequently benefit the design and manufacture of titanium alloys as well as other metallic materials. Advantages, disadvantages and impossibilities of ANN modelling technique are also discussed.

O9.2
STRUCTURE- ENERGETIC CHARACTERISTICS OF PLANAR DEFECTS IN ORDERED ALLOYS ON THE BASIS OF BCC-LATTICE. Veronika V. Romanenko, Evgenya V. Chernyh, Evgenii A. Dubov, Elena A. Golovina, Michail A. Baranov, Michail D. Starostenkov, Altai State Technical Univ, Dept of General Physics, Barnaul, RUSSIA.

It was done exhaustive classification of planar defects, taking place in the superstructures B2, D03, B32, C11b, forming on the basis of BCC lattice: antiphase boundary, conservative and non- conservative types, twins and defects of superstructure and complex type packing, rotation defects. Analytical expressions for determination of energies of planar defects forming in arbitrary crystal graphic plains were received. Structural peculiarities typical for concrete type of planar defect were calculated on the basis of computer simulation. It was of calculated the spectrum of energies of planar defects forming for concrete alloys (FeAl, Fe3Al, NaTl, MoPt2), on which basis possible thermoactivated reactions of intertransformations of conservative APB- non-conservative defect type, superstructural- complex FD were suggested. On the other side non-conservative APB can be the basis for phase transitions between superstructures of B2= D03, B2= B32, B2=C11b type.

O9.3
QUANTUM CHEMICAL MODELING OF PEROVSKITE SOLID SOLUTIONS. R.I. Eglitis, Institute of Materials Research and Engineering, Singapore and Institute of Solid State Physics, Univ of Latvia, Riga, LATVIA; E.A. Kotomin, Max Planck Institut fur Festkorperforschung, Stuttgart, GERMANY and Institute of Solid State Physics, Univ of Latvia, Riga, LATVIA; G. Borstel, Universitat Osnabrueck, Fachbereich Physik, Osnabrueck, GERMANY.

The results of quantum chemical modeling of perovskite solid solutions, $KNb_xTa_{1-x}O_3$, are presented and compared with recent full-potential LMTO calculations. We show that Nb atoms in $KTaO_3$ reveal off-center displacements along both the (100) and (111) axes at as low concentrations as x = 0.04, which is in much better agreement with EXAFS experiments than LMTO calculations (x = 0.75). Analysis of the optimized atomic and electronic structure clearly demonstrates that several nearest Nb impurities in $KTaO_3$ are able for self-ordering, unlike Ta impurities in $KNbO_3$. This is caused by tiny difference in covalency of the two perovskites.

O9.4
HIGH TEMPERATURE/PRESSURE THERMOPHYSICAL PROPERTY PREDICTION. Kai Wang and Robert R. Reeber, Dept. of Materials Science & Engineering, North Carolina State University, Raleigh, NC.

Thermal expansion is approximated by contributions from a perfect crystal and its equilibrium high temperature/pressure thermal defects. We demonstrate that the influence of thermal defects on high temperature/pressure properties can be important. Thermophysical properties are interrelated and results for important metals, and structural ceramics are compared with available data and theoretical calculations.

O9.5
AB INITIO STUDIES ON THE THERMAL PROPERTIES OF ALLOYS. Zhiqiang Li, John S. Tse, Steacie Institute for Molecuar Sciences, National Research Council of Canada, Ottawa, Ontario, CANADA.

A direct method is used to calculate phonon dispersion of Al_3Li and Al based on the Hellmann-Feynman forces obtained from the first-principles plane-wave pseudopotential calculations. The thermodynamic propertis of Al_3Li are calculated by the quasiharmonic approximation in comparison with that of Al. The calcualted quantities, such as phonon dispersions, Gruneisen parameters, heat capacities, thermal expansion coefficient and bulk

modulus as a function of temperature are discussed and compared with the availabe experiments.

O9.6
MODELLING OF COARSENING KINETICS OF MISFITTING PARTICLES. Andrei V. Nazarov, Mariya G. Ganchenkova, Moscow Engineering Physics Institute, Dept. of Materials Science, Moscow, RUSSIA; Alexandr A. Mikheev, I.P. Bardin Central Res. Inst. of Ferrous Metallurgy, Dept. of Metal Physics, Moscow, RUSSIA.

The properties of materials, which harden during the aging process, are in general determined by the sizes, volume fraction and spatial distribution of the strengthening phase. The most optimal structure is formed during the coarsening stage. We examine how elastic stress, arising from precipitate misfit strains, influences the diffusion fluxes and growth rate of precipitates. The elastic stress influence on diffusion flows in binary alloys is came into account using new approach. This approach takes into consideration, that the stress fields can alter the surrounding atom configuration and consequently the local magnitude of the activation barrier. Knowing the change of activation barrier it is possible to calculate the jump rate. The flux expressions are obtained with the help of the hole gas method, using jump rate. The equation system for two components and vacancies in which the influence of elastic stress on flows was taken into account is resolved. The obtained kinetic equation for the growth rate of a precipitate contains the additional terms conditioned by the gradients in both vacancy concentration and misfit strains. The analysis shows that the kinetics of coarsening is altered by these terms, including the possibility of inverse coarsening. Modeling of this process is realized. The temporal dependency of misfiting particle sizes is examined for different system parameter.

O9.7
STRUCTURAL AND ELECTRONIC PROPERTIES OF DEFORMED CARBON NANOTUBES. Mario S.C. Mazzoni, Dept. of Physics, University of California at Berkeley, Berkeley, CA; Helio Chacham, Dept. de Fisica, Universidade Federal de Minas Gerais, MG, Belo Horizonte, BRAZIL.

Carbon nanotubes have been fascinating the materials science community with their potential applications in nanotechnology. In this work, we apply first-principles calculations based on the density functional theory to investigate the structural and electronic properties of deformed single-wall carbon nanotubes. We consider two kinds of deformation, bending and flattening, and their implications in the electronic behaviour. For the first one, we perform a geometry relaxation and find out fourfold carbon rings at the bend region. These fourfold rings, seldomly seen in carbon structures, result from the collapse of carbon hexagons at the highest stressed region of the bend. Concerning electronic properties, localized states are found at this region, making the bend tube behave as a quantum dot. This behaviour is explained by three distinct models which take into account the structural consequences of bending, namely, the flattening of the nanotube cross section, the uniaxial stress at the outer part of the bend, and the rearrangement of the carbon atoms at the bend region. We also find that this bending-induced quantum dot can be charged with one electron or one hole at most, and thus occuring only in three charge states. Concerning the flattening, we show that it can induce a semiconductor-metal transition for an interlayer distance of 3.6 Å. Supposing that the flattening of the nanotube can be produced by a force applied by a scanning microscope tip, we estimate that the force per unit length of the nanotube that is necessary to reach the insulator-metal transition is 14.1 N/m. These results reinforces the potentialities of nanotubes in applications related to nanotechnology.

O9.8
III-V SEMICONDUCTOR MATERIALS IN MINIMOS-NT. Vassil Palankovski, Siegfried Selberherr, Inst. for Microelectronics, TU Vienna, AUSTRIA.

Considerable effort was spent on our two-dimensional device simulator MINIMOS-NT to get it ready for simulation of devices with high complexity and specifity in respect to materials, geometries, etc. Many of the existing physical models (band gap, mobility, thermal conductivity, energy relaxation times, specific heat, etc.) were refined, some of them were replaced by promising new ones, and many new models were added as well. Being an ancestor of the well-known MOS device simulator MINIMOS[1], its experience with Si devices was inherited. Thereby, MINIMOS-NT became a generic device simulator accounting for a variety of materials, including IV group semiconductors, III-V compound semiconductors and their alloys, and non-ideal dielectrics. For example, the temperature dependence of the thermal conductivity is modeled by a simple power law which gives a good agreement with experimental data[2]. In the case of alloy materials $A_{1-x}B_x$ it varies between the values of the basic materials (A and B). The model is used for device simulation with self-heating by solving the lattice heat flow equation self-consistently with the energy transport equations (system of six partial differential equations). Several applications of industrial interest employ devices operating in a wide temperature range. Therefore, our models have been designed to meet this challenge in addition to the conventional Si applications. MINIMOS-NT has been successfully used for simulation of heterostructure devices, e.g. High Electron Mobility Transistors (HEMTs) and Heterojunction Bipolar Transistors (HBTs)[3]. For example, the simulated forward Gummel plots for AlGaAs/GaAs or InGaP/GaAs HBTs compared to experimental data up to 380 K show excellent agreement and demonstrate the ability of MINIMOS-NT to reproduce correctly the thermal device behavior.
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O9.9
SIMULATION OF FERROELECTRIC MATERIALS WITH MINIMOS-NT. Klaus Dragosits, Siegfried Selberherr, Institute for Microelectronis, TU Vienna, AUSTRIA.

During recent years a special type of nonvolatile memory cells became more and more attractive, which takes advantage of the hysteresis properties of ferroelectric materials. To allow rigorous analysis of these devices several models were entered into our simulator MINIMOS-NT, which allow a general transient two-dimensional simulation of arbitrary device structures. MINIMOS-NT provides a rigorous approach to describe the static hysteresis properties of ferroelectric materials including the accurate modeling of subcycles according to Preisach hysteresis[1]. By now two different shape functions are implemented for the locus curves, based on *tanh* and *arctan*, respectively. Increasing clock frequencies lead into a regime where the frequency dependence of basic material parameters like coercive voltage and remanent polarization can no longer be neglected. Simulation in the frequency regime would be numerically cheap, but leads to reduced capabilities in comparison with the time regime. Especially in the context of arbitrarily shaped signals and relaxation effects the rigorous approach of our simulator MINIMOS-NT is mandatory. By introducing three transient terms into the basic material equation, simulation of ferroelectric capacitors in a wide range of frequencies is now possible. This enables the calculation of Q/V characteristics, the extraction of frequency dependent material parameters like e.g. the coercive voltage and the analysis of the signal response in the time regime. Simulation results for a capacitor show good agreement to measurements in a range beginning from 1Hz up to 1MHz. The application of the new simulation tool to circuit simulation is very promising. It can immediately be used for the extraction of specifications for the read and write cycles or the geometry of ferroelectric memory cells.
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O9.10
DYNAMIC EFFECTS AND INTERPARTICLE FORCE IN ELECTRORHEOLOGICAL FLUIDS. Kin-Wah Yu, Jones T.K. Wan, Chinese University of Hong Kong, Dept of Physics, Shatin, New Territories, Hong Kong, CHINA; Guo-Qing Gu, University of Shanghai for Science and Technology, College of Computer Engineering, Shanghai, CHINA.

The prediction of the strength of the electrorheological (ER) effect is the main concern in a theoretical study of ER fluids. Most theories have assumed that the particles are at rest. In realistic situation, the fluid flow exerts force and torque on the particles, setting the particles in both translational and rotational motions under these actions. A recent experiment by Lobry et. al. gave some evidences that the induced forces between rotating particles can be markedly different from the values predicted by existing theories. The gap between theory and experiment further widens rapidly because the technological applications of ER fluids have prompted many experiments which measure directly the interactions between particles of various materials under different experimental conditions. This prompts us to take particle motion into account and examine the interparticle force in ER fluids. In this work, we consider a two-particle system in which a particle is held fixed, and the other one rotates about the axis

perpendicular to the line joining the particles centers. The rotating particle leads to a displacement of polarization charge on the surface. As the polarization charge distribution deviates from the equilibrium one, it tends to restore to the equilibrium polarization within a certain relaxation time τ. We examine two typical cases: (1) a uniformly rotating particle with an angular velocity ω and (2) the particle oscillates with a simple harmonic motion with a frequency ω about its center, which can be realized in an ER fluid under an oscillatory shear. Our results show that the particle rotational motion generally reduces the induced forces between particles. A naive argument based on the point-dipole model (Klingenberg et. al.) showed that the force varies asymptotically as $F \sim 1/\omega\tau$ at large ω. However, a fully multipolar calculation has shown a significant deviation and the dependence of the interparticle force on $\omega\tau$ will be discussed. This work was supported by the Research Grants Council of the Hong Kong SAR Government under grant CUHK4290/98P. G.Q.G. acknowledges a financial support from the Key Project of the National Natural Science Foundation of China, under grant 19834070.

O9.11
NONLINEAR POLARIZATION AND INTERPARTICLE FORCE IN ELECTRORHEOLOGICAL FLUIDS. Jones Tsz-Kai Wan, Kin-Wah Yu, The Chinese University of Hong Kong, Dept of Physics, Hong Kong, CHINA; Guo-Qing Gu, University of Shanghai for Science and Technology, College of Computer Engineering, Shanghai, CHINA.

The prediction of the strength of the electrorheological (ER) effect is the main concern in a theoretical investigation of ER fluids. The applied electric field used in most ER experiments is usually quite high, and important data on nonlinear ER effects induced by a strong electric field were recently unveiled experimentally by Klingenberg and coworkers. However, the impact of a nonlinear characteristics on the interparticle force remains less well known. Naively, if we had ignored the mutual polarization effect between the particles, the force between particles would have been given by the point-dipole expression: $F \approx (\beta E)^2(1 + cE^2 + \cdots)$, where E is the magnitude of the applied field, and β, c are constants. A plot of F/E^2 against E^2 would yield a straight line. A fully multipolar calculation has revealed that the magnitude of the attractive force is generally greater than that of the point-dipole force due to multipole interactions. Moreover, for a stronger nonlinear characteristics, a significant deviation from a linear relationship occurs, which is attributed to when the nonlinear coefficient gets stronger, the force also depends on the higher orders of the nonlinear coefficients. As pointed out by Felici and coworkers, when the applied field is sufficiently strong, F will have an asymptotic E dependence and our results show a similar tendency. The major difference is that while the previous results were based on a conduction model for two touching spheres, here we have extended the considerations to two spheres separated by an arbitrary distance. As we have included all multipole interactions in our calculations, the results will be useful in computer simulation of ER fluids at an intense applied field. This work was supported by the Research Grants Council of the Hong Kong SAR Government under grant CUHK4290/98P. G.Q.G. acknowledges a financial support from the Key Project of the National Natural Science Foundation of China, under grant 19834070.

O9.12
NUMERICAL SIMULATION OF SEMICONDUCTOR MONOCRYSTALS GROWTH FROM A MELT BY A METHOD OF A VERTICAL DIRECTIONAL CRYSTALLIZATION. Alexander Serov, Zelenograd Scientifical Research Center 'Elsovi', Moscow, RUSSIA; Vladimir Maslovsky, Zelenograd State Research Institute Of Physical Problems, Moscow, RUSSIA.

The mathematical model of technological process of monocrystals growth from a melt by a vertical directional crystallization is designed. In a basis of the created numerical method of a Stefan problem solution is the algebraic transformation specifying one-to-one mapping of the solution area from cartesian space in curvilinear settlement space which is taking into account the shape and position of a crystal/melt interface. The application of driving functions allows to realize a required distribution of grid nodes in the field of boundary layers and areas with considerable quantities of grid functions gradients. Numerical scheme includes non-stationary equations of Navier-Stokes, heat conduction equation, diffusions and equation of continuity. Equations of numerical scheme fulfills conservation laws of energy and mass of crystal/melt system, and also geometrical conservation law. The application of methods of split on physical processes and spatial coordinates together with use of an algebraic method of numerical grid generation has allowed to organize effective computing process. The embodying of a designed numerical method was used at examination of HgCdTe monocrystals growth. It has allowed to explore the mutual influence of processes in system volume and processes transiting in boundary layers. The bond between radial and axial impurity segregation processes and inhomogeneities of impurity distribution in solid/liquid interface region was subjected to analysis. Conformities to natural laws of temperature, impurity concentration and melt motion velocity near solid/liquid interface were revealed under presence of interface velocity oscillations. Calculations were accomplished for normal and low gravity, that allowed to perform the investigation of melt velocity influence on the shape and velocity of crystal/melt interface.

O9.13
MODELING OF SILICA ETCHING BY A CF_4 PLASMA: A DENSITY FUNCTIONAL THEORY STUDY. Alessandra Ricca, NASA Ames Research Center, Moffett Field, CA.

The role of CF_3, CF_2 and CF neutrals and their corresponding ions in the etching of silica is studied using Density Functional Theory. The $(HO)_3SiOSi(OH)_3$ cluster is used to model the silica surface. The energetics and the reaction barriers are calculated using the hybrid functional B3LYP. Reaction rates are computed for the most relevant reactions.

O9.14
DEVELOPMENT of LOW-κ METHYLSILOXANE SOG FILMS AIDED BY QUANTUM CHEMICAL CALCULATIONS. Yoichi Matsuzaki, Atsushi Nogami, Nippon Steel Co, Advanced Materials & Technology Research Lab, Chiba, JAPAN.

A new guideline for the Sol-Gel synthesis of low-κ methylsiloxane network polymers has been derived by quantum chemical calculations. The microscopic origin of materials' dielectric response consists of electronic, vibrational, and orientational polarization. Although experiments have indicated that special attention should be paid to the vibrational contribution to ensure low-κ for this class of materials, the structure-property relationship has not been sufficiently elucidated for such polarization. Therefore, we have carried out molecular orbital calculations of vibrational polarizabilities for some oligomers representing typical structures in the methylsiloxane network. It was found that a chain-like structure terminated by Si-OH groups, which is left free from cross-linking, shows significantly enhanced vibrational polarizability relative to those for the "closed" structures such as ring or cube without free Si-OH terminal. The perturbative expression of vibrational polarizability in the chain-like molecule is dominated by the lowest frequency ($\sim$10 cm^{-1}) acoustic mode which is intrinsic to a floppy macromolecular structure. We note such an enhanced contribution is also due to the polarity of terminal OH groups since it increases the dipole change induced by the acoustic mode. Therefore, in order to develop low-κ siloxane network materials, it is essential to optimize the monomer structures as well as the experimental condition such that the number of remaining floppy chains with -OH terminal groups is minimized. The experimental approach along with this guideline has succeeded in developing κ = 2.3 SOG film.

O9.15
MODELLING SILICON SOLID PHASE CRYSTALLIZATION OF NEAR CRYSTAL DENSITY CONTINUOUS RANDOM AMORPHOUS SILICON NETWORKS. E.L. Jaen, State University of New York at Stony Brook, Dept of Applied Mathematics and Statistics, Stony Brook, NY; C.M. Fortmann, Tokyo Institute of Technology, Dept of Innovative and Engineered Materials, Yokohama, JAPAN.

Many groups have successfully developed amorphous silicon continuous random network models capable of describing various properties of this important semiconductor thin film material. Several recent reports give cause to revisit this problem. First, Futako et al. (Futako et. al. to be published in J. Non-Cryst. Solids, in press) reported that the atomic density of hydrogenated amorphous silicon can be as dense as crystal silicon. Second, Fortmann (C.M. Fortmann, Phys. Rev. Lett. Vol. 81, No. 17, 26 Oct. 1998 pp. 3683-3686) developed an optical model that described the hydrogenated amorphous silicon optical band gap in terms of crystal silicon-like indirect transitions modified by forbidden phonon bands interacting with a truncated crystalline Brillouin zone. Therefore, it is possible to decouple the site disorder from the optical band transitions and the measured material random distribution functions. We developed a continuous random amorphous network model using nano scale psuedo crystallytes and an extremely high density of twin boundaries separated by a few Angstroms. This near-crystal continuous random network model replicates the measured amorphous silicon radial distribution function, and the its atomic density. The model predicts that impurity species and hydrogen occupy the micro-twin boundaries nearly strain free. One of the most important aspects of this model is that it renders the difficult problem of nucleation and grain growth in an amorphous silicon network tractable. The model was used to probe several aspects of amorphous silicon solid phase epitaxial growth for vertically integrated circuits, including lateral grain growth; as well as, interface effects, and bulk nucleation effects.

O9.16
A QUANTUM CHEMISTRY INVESTIGATION ON THE FORMATION OF LOW-DIMENSIONAL CARBON MATERIALS. Shaoqing Wang, Shouliang Bu, Hengqiang Ye, Laboratory of Atomic Imaging of Solids, Institute of Metal Research, Chinese Academy of Sciences, Shenyang, PR CHINA.

Since the first discovery of the intriguing gigantic carbon molecular of fullerence in 1995 [1], many theoretical and experimental studies have been performed to explore the specific atomic structure and physical property of low-dimensional carbon materials. The atomic configuration of these materials display in vast varieties from spherical fullerences to columnar nanotubes [2]. In all of these structures the carbon atoms position in the curved surface of two-dimension. An urgent awaiting-answered question lies in front of the researchers is that how these structures form? In the present work, we studied the energetic property of carbon clusters ranging from linear chains, rings, graphite fragments to fullerences and nano-meter tubes by accurate ab initio quantum chemistry and semi-empirical molecular orbital calculations. The essential elementary units were deduced for the construction of low-dimensional carbon materials. A thorough theoretical analysis was accomplished on the basis of the results in present calculations and some early works [3-5]. The preferential relationship of the elementary carbon units, composing fullerence and nano-tube, were investigated, energetically. We explored how these elementary carbon units pile up in different way shaping as different two-dimensional materials and what it is most likely to be formed under specific conditions. References [1] H.W. Krot, J.R. Heath, S.C. O'Brien, R.F. Curl, and R.E. Smalley, Nature 347, 162 (1985). [2] S. Iijima, Nature 354, 56 (1991). [3] C.Z. Wang, B.L. Zhang, K.M. Ho, and X.Q. Wang, Inter. J. Mod. Phys. B7, 4305 (1993). [4] J.C. Niles, X.Q. Wang, J. Chem. Phys. 103, 7040 (1995). [5] F. Jensen, H. Koch, J. Chem. Phys. 108, 3213 (1998).

O9.17
COMPUTATIONAL RESEARCH ON MAGNETIC PROPERTIES OF POWER-SUPPLY FERRITE MATERIALS. Sheng-Chuan Zhu, Hai-Ying Chen, Dept. of Physics, Peking University, Beijing, CHINA; Jun-Bin Zhu, Dept. of Mechanics & Engineering Science, Peking University, Beijing, CHINA.

A series of computational research on the MnZn ferrite power-supply materials have been done. The high frequency characterics of this material are calculated from the basic magnetic parameters and compared with those of experiments. The results show that: 1) The material composition of the MnZn ferrite sample can be determined from the saturation magnetization and the given physical parameters. 2) The Ms (T) and K1 (T) curves can be calculated from the Ms value at room temperature and the material composition (K1 is the magnetocrystilline anisotropy constant). 3)The frequency characteristics of the complex permeability and power loss are calculated by using the basic magnetic parameters and the characteristic parameters. 4) In the ranges of f = 25 - 1000 KHz and T = 25 - 100 C , the calculating and experimental results are compared for a series of MnZn ferrite materials, good agreements are obtained.

O9.18
DETERMINATION OF THIN POLYMER LAYER TENSILE STRENGTH BY NANO-INDENTATION, MICRO-SCRATCH TESTING, AND FINITE ELEMENT METHOD. Timothy C. Ovaert, Byung Ro Kim, Lihui Zhao, Dept. of Mechanical and Nuclear Engineering, Penn State University, University Park, PA.

In a companion paper, thin polymer layer visco-elastic/plastic properties were determined using nano-indentation and the finite element method. The individual layers, once characterized, were then assembled into two- or three-layered structures and subject to micro-scratch tests using a custom designed (with closed loop feedback control) micro-scratch tester, capable of determining a critical normal indentation load (evidenced by visible surface tensile cracks) for the layered structures. Experimental scratch tests and finite element simulations were carried out for three different spherical scratch tip radii. Top-layer tensile strength of the layered structures was then estimated by a finite element scratching model, utilizing the individual layer properties, top layer friction coefficient, and micro-scratch critical load. The method may be used as a basis for coating selection, comparison, and process monitoring.

O9.19
ATOMISTIC MODELING OF TUNGSTEN GRAIN BOUNDARY RESISTANCE WITH RESPECT TO SHIFTS. David Fuks, Gunnar Borstel, FB Physik, Uni Osnabrueck, Osnabrueck, GERMANY; Kleber Mundim, Inst of Physics, FUB, Salvador, BRAZIL; Vlad Liubich, Dept Mater. Eng., BGU, Beer-Sheva, ISRAEL; Simon Dorfman, Joshua Felsteiner, Dept of Physics, Technion, Haifa, ISRAEL.

The purpose of our work was to calculate the elastic properties of $\Sigma_3 < 111 >$ GB in pure tungsten and the same with boron additives, which occupy the interstitial position at the GB. For extremely dilute W-B solid solutions we performed the total energy calculations for the binary disordered alloy with the Linear Muffin-Tin Orbitals (LMTO) method in the framework of the Coherent Potential Approximation (CPA). In this approach the total energies were calculated for varying lattice constants in order to determine the relative stability of different structures and to locate the equilibrium lattice constant. With obtained values of the total energies of the alloy and the total energies of pure compounds we defined the mixing energy as a function of the lattice parameter. We extracted W-B potentials from ab initio calculations with the exact procedure for inverting ab initio energy data. Obtained potentials were applied in Monte Carlo simulations aimed to study the $\Sigma_3 < 111 >$ GB in tungsten with and without boron atom as additive. Our calculations showed that the presence of B on the $\Sigma_3 < 111 >$ GB of tungsten increases sufficiently the value of the energy barrier. Thus boron prevents the sliding along the GB and increases the resistance with respect to the shift in this direction. Experimental investigation of tungsten alloyed with boron was found to result in a significant drop in the ductile-brittle transition temperature. We showed that even a small amount of boron changes the fracture resistance of the $\Sigma_3 < 111 >$ GB in tungsten.

O9.20
ASPERITY SHEAR IN ALUMINUM. Jun Zhong, James B. Adams, Arizona State Univ.- Main Campus, Dept of Chemical, Bio, and Materials Engineering, Tempe, AZ; Hualiang Yu, Arizona Sate Univ., Science and Engineering of Materials Program, Tempe, AZ.

One important wear mechanism involves the shear of asperities by other asperities. We use molecular dynamics to simulate the shearing of aluminum asperities by a hard (Lennard-Jones) asperity. The simulations were repeated for a wide range of conditions, including asperity velocity, temperature, asperity shape, degree of intersection, crystal orientation, and adhesive strength, to determine their effect on the wear process. These simulations involve the use of a reliable EAM potential for Al that was developed by Force Matching Method to a large database of DFT forces.

O9.21
HIGH PERFORMANCE PARALLEL AND DISTRIBUTED SIMULATIONS OF A SIMPLE MODEL FOR DETERMINISTIC AND STOCHASTIC FRAGMENTATION. Alejandro Figueroa and Gonzalo Hernandez, Department of Informatics, Federico Santa Maria Technical University, Valparaiso, CHILE.

In this work we present numerical results of the study of a simple 2 and 3 dimensional model for fragmentation, obtained by large scale parallel and distributed simulations, implemented on a small network of high performance personal computers linked by fast ethernet protocols. Our fragmentation models, which fulfills mass conservation, is an iterative stochastic process running over a 2 or 3 dimensional lattice which represents the material to be broken (discrete materials). At each step of the process the most stressed piece is broken in the direction of the maximum net force, with a probability that depends on its size (area or volume). Also, it is assumed that small pieces are more difficult to be broken than larger ones, depending on another random variable. The analytical complexity of our models allow us to studied them only in a numerical way. For this purpose, we implemented large scale parallel and distributed numerical simulations over a high performance personal computers network, linked by fast ethernet technologies. We determined the fragment or piece size (areas and volumen) distribution, that it is believed that follows a power law behaviour. Despite their simplicity, our models present complex features that reproduce some of the experimental results that have been obtained. For some regimes a log-normal and a power law behavior are obtained for the fragment size histogram. For this reason we propose them as basic models that can be substantially refined to describe the fragmentation process of more realistic model. Finally, it is important to point out that the accuracity and diminution in the finite effects of our results were obtained by large scale numerical results implemented through parallel and distributed simulations over a very cheap network of high performance computers linked by fast ethernet protocols.

O9.22
DEVELOPMENT OF GLUE TYPE POTENTIALS FOR THE Al-Pb SYSTEM: PHASE DIAGRAM CALCULATION. Alex Landa, Paul Wynblatt, Carnegie Mellon University, Dept. of Materials Science and Engineering, Pittsburgh, PA; Donald Siegel, University of Illinois at Urbana-Champaign, Dept. of Physics, Urbana, IL; Jim Adams, Arizona State University, Dept. of Chemical, Bio and Materials Engineering, Tempe, AZ; Oleg Mryasov, Northwestern University, Dept. of Physics and Astronomy, Evanston, IL; Xiang-Yang Liu, Motorola Inc., Comput. Materials Group, Los Alamos, NM.

Empirical many-body potentials of the glue-type have been constructed for the Al-Pb system using the force matching method. Because only very limited experimental information is available for the Al-Pb system (namely the enthalpy of mixing of liquid solutions), the potentials have been fitted to a set of the ground state physical quantities calculated within a full-potential linear-muffin-tin orbitals method, dilute heats of mixing calculated within a first-principles approach using ultrasoft pseudopotentials and a massive quantum mechanical forces database for samples of bulk Al-Pb liquid alloys at different temperatures generated using ultrasoft pseudopotentials in conjunction with a first-principles molecular dynamics program VASP. The constructed potentials were used for computer simulations of the thermodynamic properties of the Al-Pb system over a wide temperature range from 0 K to 2000 K. Monte Carlo simulations using these potentials have been employed to compute an Al-Pb phase diagram which is in fair agreement with experimental data. Support by the DOE under grant DE-FG02-99ER45773 and the NSF under grant DMR9619353.

O9.23
DEVELOPING EAM TANTALUM POTENTIAL USING FORCE MATCHING METHOD. Youhong Li, Department of Chemical, Bio, and Materials Engineering, Arizona State University, Tempe, AZ; Donald J. Siegel, Physics Department, University of Illinois, Urbana, IL; James B. Adams, Department of Chemical, Bio, and Materials Engineering, Arizona State University, Tempe, AZ.

Atomic potentials are useful for molecular dynamics simulations of large systems (millions of atoms). Since the existing potentials for tantalum are of limited reliability, we have attempted to develop an improved EAM potential for tantalum using the Force Matching Method. In addition to fitting the potential to the lattice constant, elastic constants, cohesive energy, and vacancy formation energy, we also fit the potential to thousands of forces calculated via DFT for a wide variety of structures including BCC tantalum, beta-tantalum, surfaces, crystal defects, and liquid tantalum. Initial testing of the potential shows that its predictions are in good agreement with experimental values, and are a substantial improvement over previous models.

O9.24
RELATIVISTIC CLUSTER CALCULATION OF Ti $L_{2,3}$-EDGE X-RAY-ABSORPTION NEAR-EDGE STRUCTURE OF RUTILE. Kazuyoshi Ogasawara, Takahiro Iwata, Takugo Ishii, Hirohiko Adachi, Kyoto Univ, Dept of Materials Science and Engineering, Kyoto, JAPAN; Isao Tanaka, Kyoto Univ, Dept of Energy Science and Technology, Kyoto, JAPAN.

The Ti $L_{2,3}$-edge x-ray-absorption near-edge structure (XANES) of TiO_2 (rutile) has been calculated based on the configuration interaction approach using a relativistic cluster calculation. The relativistic molecular orbitals were obtained by numerically solving the Dirac equation using the 4-component relativistic atomic orbitals as the basis functions. Then the many-electron Hamiltonian was diagonalyzed within the subspace spanned by the Slater determinants constructed from the relavant molecular orbitals. The oscillator strengths of the electric dipole transition between the multiplets were calculated directly using the many-electron wave functions expressed as linear combination of the Slater determinants. The 2p absorption spectra of Ti in rutile was analyzed by de Groot *et al.* based on the atomic multiplet approach. In their analysis, three crystal field parameters were required due to the distortion from the octahedral symmetry and these parameters were determined by fitting to the experimental spectrum. In general, for the analysis of lower-symmetry systems, the number of parameters would increase and the ambiguity in the determination of empirical parameters would also increase. Therefore, first-principle calculation is quite necessary for the analysis of multiplet strucutres in XANES for arbitrary materials. In the present work, we calculated the 2p absorption spectrum of Ti in rutile from first principle. The multiplet structure in XANES was reproduced satisfactorily without referring to any experimental data.

O9.25
GRAIN BOUNDARY AND LATTICE DIFFUSION IN FINE-GRAINED MATERIAL. Irina Belova and Graeme Murch, Univ of Newcastle, Diffusion in Solids Group, Dept of Mechanical Engineering, Callaghan, AUSTRALIA.

In polycrystalline materials the Hart equation provides a relationship between the effective diffusivity and the grain boundary and lattice diffusivities. The principal assumption in the derivation of the Hart equation is that the two diffusion processes (grain and lattice) proceed in parallel. In fine-grained material this assumption is almost certain to be unsustainable since a given atom will sample *both* diffusivities many times in the diffusion time. In this paper we use Monte Carlo methods to calculate the effective diffusivity for the model of parallel slabs (of high diffusivity) representing the grain boundaries embedded in a host of lower diffusivity, up to five orders of magnitude lower. A very fine grain grid overlays this and the grain and lattice diffusivities are represented by particles performing appropriate biassed random walks on the grid. It was found that for a given diffusion time the Hart equation overestimates the effective diffusivity by as much as several orders of magnitude, the overestimation depending on the ratio of the grain boundary diffusivity and lattice diffusivity.

O9.26
DIFFUSION IN Ni_3Al. Graeme Murch, Irina Belova, Univ of Newcastle, Diffusion in Solids Group, Dept of Mechanical Engineering, Callaghan, NSW, AUSTRALIA.

The intermetallic compound Ni_3Al is of substantial interest on account of its valuable high temperature mechanical properties. An understanding of the mechanisms of diffusion in Ni_3Al has immediate benefits in the understanding and prediction of sintering, creep, oxidation and grain growth. In this paper we review first the present understanding of diffusion in Ni_3Al and, specifically, the compatibility of the various proposed diffusion mechanisms with the diffusion data. To obtain further insight into the relative roles of some of the proposed mechanisms we have taken a set of lattice statics migration energies of Bocquet and Schmidt and used them as input into a Monte Carlo calculation of the diffusivities. It is clear that Ni atoms diffuse largely by jumping on the Ni sublattice with only occasional excursions to the Al sublattice. The Ni diffusivity is not sensitive to composition (as found experimentally). Al atoms make frequent jumps to the Ni sublattice where some remain to diffuse. This provides the principal means for Al transport. The Al diffusivity is very sensitive to composition but this is yet to be experimentally verified.

O9.27
SIMULATION OF DIFFERENT DIFFUSION MECHANISMS IN ORDERED STRUCTURES. Maria G. Ganchenkova, Andrei V. Nazarov, Moscow Engineering Physics Institute, Dept. of Materials Science, Moscow, RUSSIA.

The scientific interest in intermetallic compounds arises from basis research aspects of more complex systems and their potentials for technical application as light-weight high-temperature materials. Knowledge of their diffusion behaviour is important for the production of these materials or for their use in practical applications. In this conjunction order-disorder processes, atomic diffusion are of particular relevance and the understanding of these processes requires a profound knowledge of the features of the atomic defects migration. Much theoretical and experimental works has focused on the intermetallics. The diffusion mechanisms in intermetallics, however, are not well established. The aim of our work is taking into account an influence of interaction between the different defects on the defect diffusivity. For the solving of this problem the new model was developed and in the framework of it we have compared the different diffusion mechanisms in B2 and L12 ordered structures. For this aim it was used the static relaxation method for the calculation of the activation barriers for the different atomic jumps and recently developed approach for the calculation of activation volumes of migration. For the migration simulation we used Monte-Carlo method The results allow to infer about the existence of a new diffusion mechanism, which is called Dynamic Pair (DP) mechanism. The analysis of results of our study and different publications on this subject both theoretical and experimental allows us to say, that since starting of diffusion investigations in intermetallics, apart from common monovacancy mechanisms, only two volume mechanisms, to be intrinsic for ordered structures were suggested (six-jump cycle of Huntington and triple-defect mechanism). The DP-mechanism suggested in the research work is the third principally new mechanism in these structures.

O9.28
RAC-MESO CONFIGURATIONAL STABILITY AND STRUCTURAL TRANSITION IN ZIRCONOCENE-BASED ZIEGLER-NATTA CATALYSTS – A HYBRID QM/MM INVESTIGATION. Amitesh Maiti, Jan Andzelm, Molecular Simulations Inc., San Diego, CA; Joe Golab, BPAmoco Naperville Complex, Naperville, IL; Marek Sierka, Joachim Sauer, Humboldt University, Berlin, GERMANY.

Transition metal-based Ziegler-Natta catalysts are extremely important in the synthesis of commercial polymers. The structures of these catalysts determine the rate of synthesis, tacticity, and molecular weight of the polymer, which in turn effects their physical and chemical properties. A typical catalyst can be in one of two configurations - the 'rac' or the 'meso'. Understanding the relative stability of these configurations and structural transformation from one to the other is of great relevance to catalyst design. Due to the large atomic size of these systems, First Principles calculations are often prohibitive. In this work, we circumvent that problem by using a

hybrid QM/MM approach [1]. Such a technique consists of embedding a chemically interesting small 'Quantum Mechanical cluster' inside a 'classical surrounding' or 'host'. A typical application involves structural and electronic changes in the 'QM cluster' while the inert classical host exerts long-range electrostatic and strain effects on the QM cluster. Contrary to the typical scenario described above, the present application involves significant changes in atomic positions in the classical 'host' region as one goes from the 'rac' structure to the 'meso'. This leads to a very strong dependence of results on the choice of the QM cluster. In particular, a bigger QM cluster does not necessarily yield better results. With the concrete example of a commercially important Zirconium-based catalyst, the present work illustrates that in spite of these difficulties, one can prudently choose a QM cluster for such a system, such that hybrid QM/MM techniques can yield meaningful results when compared with accurate first-principles DFT calculations. The talk will discuss: (1) rac-meso energetic stability, (2) activation barrier for the rotational transition, and (3) effects of structural substitution of the basic catalyst with organic functional groups. [1] Eichler, Kolmel, and Sauer, J. Comp. Chem 18, 463 (1997)

O9.29
DESIGNING ADSORBENT MATERIALS FOR GAS SEPARATION. Anthony Malanoski, The University of New Mexico, Department of Chemical and Nuclear Engineering, Albuquerque, NM; Frank van Swol, Sandia National Laboratories, Catalytic and Porous Materials Dept., Albuquerque, NM.

Recent advances in the development and application of self-assembly techniques have opened up the possibility of tailoring membranes for specific gas separation problems. The new self-assembly processing route has made it possible to finely control both the three-dimensional (3D) porosity as well as the chemical nature of the adsorbing structures. To provide guidance to the optimized use of these designer membranes we have developed a substantial modeling program that focuses on permeation through porous materials. The key issues that need to be modeled concern 1) the equilibrium adsorption behavior in a variety of 3D porous structures, ranging from straight pore channels to fractal structures, 2) the transport (i.e. diffusion) behavior in these structures. Complicating the problem is the presence of reactive groups that may be present on the surface. An important part of the design of actual membranes is to optimize these reactive sites with respect to their strength as characterized by the equilibrium constant, and the positioning of these sites on the adsorbing surface. What makes the technological problem challenging is that the industrial application requires both high flux and high selectivity. What makes the modeling challenging is the smallness of the length scale (molecular) that characterizes the surface reaction and the confinement in the pores. This precludes the use of traditional continuum engineering methods. However, we must also capture the 3D connectivity of the porous structure which is characterized by a larger than molecular length scale. This paper focuses on how we have used lattice models and both Monte Carlo and 3D density functional theory methods tackle the modeling problems.

O9.30
AB INITIO STUDY OF ADSORPTION AND DECOMPOSITION OF NH_3 ON Si(100)-(2×1). Yuniarto Widjaja, Michael M. Mysinger, Charles B. Musgrave, Stanford Univ, Dept of Chemical Engineering, Stanford, CA.

In this study, we investigate the atomistic mechanisms for NH_3 adsorption and initial decomposition on the (2×1) reconstructed Si(100) surface using B3LYP density functional theory (DFT) and by utilizing cluster approximations. We present a detailed investigation of the effects of cluster size on reaction energetics. We find that the Si_9H_{12} (1-dimer) cluster model does not describe the NH_3(a) adsorption state well due to the delocalized nature of the electron transfer in forming the NH_3(a) dative bond. We find that $Si_{21}H_{20}$ (3-dimer) cluster is necessary to capture these non-local effects. The 3-dimer model is able to satisfactorily reproduce the results of larger cluster models. We then use the 3-dimer cluster model to study the adsorption and decomposition reactions of NH_3 on the Si(100)-(2×1) surface. Ammonia is found to adsorb on the down atom of buckled silicon dimers with no activation barrier. This adsorption is then followed by dissociation to form NH_2(a) + H(a) with a low activation energy. Our calculated recombination desorption energy of 51 kcal/mol is found to be in good agreement with the TPD experimental result of 47 kcal/mol. In addition, we also calculate the vibrational spectra of the dissociated species. We find our results to be in good agreement with the experimental HREELS spectra.

O9.31
DYNAMICS OF SMALL MOLECULES AT GaS/ZEOLITE INTERFACES. M. Chandross, E.B. Webb III, G.S. Grest, Sandia National Laboratories, Albuquerque, NM.

Zeolites are microporous materials that are ideally suited to a number of industrial applications including the separation of small molecules. In order to better understand this technologically important process, we have performed molecular dynamics simulations in which interfaces are formed between the 010 surface of silicalite and dilute gases of linear and branched hydrocarbons. After equilibration, we examine both the structure of the adsorbed surface layer and the dynamics of exchange between the gases and the zeolite. We present results for both single gas species and binary mixtures to address the roles of preferential adsorption and diffusion in separations processes.

SESSION O10: INTERFACES
Chair: Sokrates T. Pantelides
Thursday Morning, April 27, 2000
Golden Gate C1 (Marriott)

8:30 AM O10.1
FIRST PRINCIPLES MODELING OF ADHESION AT METAL-CERAMIC INTERFACES. Donald J. Siegel, University of Illinois at Urbana-Champaign, Department of Physics, Urbana, IL; Louis G. Hector, Jr., ALCOA Technical Center, ALCOA Center, PA; James B. Adams, Arizona State University, Department of Chem, Bio, and Materials Engineering, Tempe, AZ.

One of the most common tribological problems is that of adhesion and adhesive metal transfer, in which one material transfers to the surface of another at an interface. Although the structure of such interfaces is generally very complex–possibly involving lubricants and oxides of various composition–much can be learned from a simplified model in which a metal surface is placed in coherent contact with a ceramic surface. Until recently, there have been no successful theoretical models capable of predicting the nature of the adhesive bonding at such an interface. With the advent of first principles calculations based on Density Functional Theory (DFT), such predictions are now possible and have been made for a small number of systems. Along these lines, we discuss our recent DFT-LDA/GGA calculations of the equilibrium structure, bonding, and adhesion energetics of various Al/ceramic interfaces, focusing on the industrially relevant Al(111)/α-Al_2O_3(0001) and Al(111)/WC(0001) systems. We comment on our ultimate goal of an *ab initio* simulation depicting adhesive metal transfer.

8:45 AM O10.2
DESIGN GUIDELINES FOR THIN FILM LAYERED SYSTEMS. W.M. Ashmawi, K. Wang*, R.R. Reeber* and M.A. Zikry, Department of Mechanical Engineering, North Carolina State University, Raleigh, NC. *Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC.

Deformation and failure modes of thin film layered systems have been investigated. Analytical and computational methodologies have been developed and used to characterize the effects of film thickness, interfacial stress and strain evolution, temperature, crystalline structure, and orientation on the growth, deformation, and failure of thin films in GaN/Al_2O_3, GaN/SiC, and GaN/AlN. New property estimates for thermal coefficients, which have been obtained for a broad range of temperatures, are used in conjunction, with specialized finite-element computational techniques to obtain accurate predictions of interfacial shear delaminations and tensile debonding on physical scales commensurate with thin film layered structures. It is shown that these computational methodologies, predictions, and material properties can be used as design guidelines to determine optimal materials and architectures for layered thin film systems.

9:00 AM *O10.3
CATALYSIS FROM FIRST PRINCIPLES. J. K. Norskov, Center for Atomic-scale Materials Physics, Department of Physics, Technical University of Denmark, Lyngby, DENMARK.

Recent progress in the theoretical description of elementary reactions on transition metal surfaces is discussed. Calculations based on density functional theory and a non-local description of exchange and correlation effects can now be used to predict changes in reactivity from one system to the next. On the basis of the calculations, models can be developed elucidating the electronic factor in catalysis. The calculations and concepts developed on their basis provide new tools for catalyst development.

SESSION O11: SOLIDIFICATION AND SINTERING
Chair: Gerbrand Ceder
Thursday Morning, April 27, 2000
Golden Gate C1 (Marriott)

10:00 AM *O11.1
FROM CONSTRAINED TO UNCONSTRAINED GROWTH DURING DIRECTIONAL SOLIDIFICATION. Ch.-A. Gandin, Ecole des Mines de Nancy, Laboratoire de Science et Génie des Matériaux Métalliques, Nancy, FRANCE.

A one-dimensional solidification model is developed for the study of directional solidification of dendritic alloys. It is based on the resolution of the heat flow equation using a two-interface front tracking technique. The two interfaces are defined by fictitious limits assumed macroscopically flat which correspond to the position of the growing dendritic and eutectic interfaces. They delimit the three regions that are considered: the liquid, the mush and the solid. Growth kinetics laws are applied on the interfaces via velocity versus temperature relationships. It is found that, if a complete solidification is carried out directionally up to the top of the ingot (i.e., formation of a fully columnar structure), the velocity of the dendrite tips first increases during the stage of the superheat loss, then decreases when no substantial thermal gradient remains in the liquid ahead of the growing dendritic interface. Applied to directional solidification experiments carried out in aluminum-silicon alloys, the model shows that the maximum velocity is reached when the top position of the mushy zone (i.e., the dendritic interface) reaches about two-thirds of the ingots length. This position being in the vicinity of the columnar-to-equiaxed transition (CET) observed in the longitudinal cross section of the ingots, a CET scenario is proposed based on a constrained-to-unconstrained growth transition, leading to a breakdown of the columnar dendritic front.

10:30 AM *O11.2
MULTI-PHYSICS MODELLING OF MATERIALS PROCESSING DURING MANUFACTURE. Mark Cross, Chris Bailey, Koulis Pericleous, Nick Croft, Gary Taylor, Kevin McManus, Daniel Wheeler and Alison Williams, Centre for Numerical Modelling and Process Analysis, University of Greenwich, London, UNITED KINGDOM.

The increasing demand for higher fidelity components and products in the market place is matched by the need for control over manufacturing processes and performance prediction in service. Manufacturing processes, especially, often involve a range of complex physical phenomena that interact. To analyse these proceses requires the simulation of these phenomena and their interactions - multi-physics. A problem with many of these processes is that they are flow dominated and so the modelling tools require high fidelity 'flow' solvers with extensive 'physics' capabilities, as well as solid mechanics, heat transfer and phase change, etc. In the paper we will outline these issues and describe the approach taken at Greenwich to address them. The approach is based upon finite volume discretisation methods, but uses unstructured (i.e. finite element like) meshes in 3D. The demanding numerical computation requires parallel computing systems and so this issuse will also be briefly reflected upon. Finally, we shall show the application of multi-physics simulation to electronics component manufacture and shape casting of metals, to illustrate some of the key ideas and challenges.

11:00 AM *O11.3
AN ATTEMPT OF PREDICTION OF THERMODYNAMIC PROPERTIES BY THE COMBINATION OF FIRST PRINCIPLE CALCULATION AND MONTE CARLO STIMULATION. Tooru Matsumiya, Advanced Technology Research Laboratories, Nippon Steel Corporation, Futtsu, JAPAN.

As the first approximation, activity coefficients and interaction parameters of solutes in dilute silicon solution was estimated by total energy calculations based on local density functiobal approximation neglecting excess entropy of mixing. The energies of pure silicon with diamond structure and various pure elements, which were considerd as solutes in silicon, were calculated. The energy of the silicon solution with a solute atom were calculated and subtraced by the sum of the energy value of solute atom in its pure state and silicon atoms in its pure sate with diamond structure calculated above. This gave the excess energy of the solute atom in the silicon solution, from which activity coefficient of the solute was deriverd. The energy of silicon solution with an atom of solute A and an atom of solute B was also calculated and the interaction parameter of solute A and B was estimated. Grand canonical Monte Carlo simulations were conducted to improve the estimated values of activity coefficients and interaction parameters by taking into consideration of the excess entropy of mixing. In the simulations, chemical potentials of solutes were given as input values and the chemical compositions of the simulated system were obtained as the output. Based on the derived relation among mole fractions and chemical potentials of solute elements in silicon, the estimations of above thermodynamic parameters were improved. Finally, the free energy of silicon-oxygen solution was calculated as a function of oxygen concentration by the integration of the relation between the gradient of the free energy curve given as the input, i.e. chemical potential difference, and chemical compositions resulted as the output in Monte Carlo simulations. On the other hand total energy on pure tridymite SiO_2 was calculated. From the cotangent line of the above obtained free energy curve of silicon-oxygen solution and the tridymite energy, solution limit of oxygen in silicon was estimated.

11:30 AM O11.4
A STATISTICAL-MICROSTRUCTURAL MODEL FOR SIMULATION OF SINTERING. Michael Braginsky and Veena Tikare, Sandia National Laboratories, Albuquerque, NM.

The concepts of sintering theory have been developed either based on the application of complex diffusion mechanisms to a simple geometry, or in the framework of continuum mechanics (i.e., plastic or nonlinear-viscous compaction of porous bodies). We present a model for simulation of sintering which incorporates the concept of deformation into the mesoscale statistical Monte-Carlo (Potts) approach. This model is capable of simulating vacancy diffusion along grain boundaries, annihilation of vacancies at grain boundaries resulting in densification, and coarsening of microstructural features. The incorporation of deformation into the model is needed to generate, based on the evolution of microstructure and sintering mechanisms, parameters of macroscopic constitutive equations used in continuum simulations of sintering of complex parts. Thus, the model establishes an important link between mesoscopic mechanisms of sintering and the macroscopic mechanical behavior of the sintered material.

11:45 AM O11.5
RELATING INITIAL MICROSTRUCTURE AND CONSTRAINTS TO REARRANGEMENT AND INSTABILITITIES DURING SINTERING. Jeffrey W. Bullard, University of Illinois at Urbana-Champaign, Department of Materials Science and Engineering, Urbana, IL.

The inital-stage sintering of a porous powder compact is an enormously complex dynamic phenomenon that often involves massive rearrangement of particles and the development of undesirable flaws in the microstructure. Traditional models of initial-stage sintering has involved either rigorous treatment of a few particles or constitutive models of macroscopic behavior that involves few if any links to the underlying microstructure. In this talk a simple numerical model will be presented that simulates microstructural rearrangement during initial-stage sintering of hundreds of particles. The model distills the complexity of the sintering phenomena into a non-uniform finite element framework. Applications and comparisons will be made to constrained sintering of particulate coatings on rigid substrates for electrical and thermal applications.

SESSION O12: MECHANICAL PROPERTIES
Chair: James A. Warren
Thursday Afternoon, April 27, 2000
Golden Gate C1 (Marriott)

1:30 PM O12.1
MOLECULAR DYNAMICS SIMULATIONS OF WEAR PROCESSES. Hualiang Yu, Arizona State Univ, Science and Engineering of Materials Program, Tempe, AZ; James B. Adams, Arizona State Univ, Dept of Chemical, Bio and Materials Engineering, Tempe, AZ.

Wear processes are important in many industries. Relatively little work has been done on determining atomic-scale wear mechanisms and the factors that control them. We have begun studies of two wear processes, indentation and asperity shearing. For nanoindentation, the effects of many process variables are investigated, including temperature, tip-substrate bonding, indentation force, orientation and alloy composition. We discuss the deformation mechanisms which occur during indentation and pull-out. Similar studies are presented for asperity shear.

1:45 PM *O12.2
COMPUTATIONAL MODELING OF CONTACT AT SURFACES: MICROMECHANICS, NEW CHARACTERIZATION TOOLS, DESIGN OF SURFACES AND APPLICATIONS. S. Suresh, A.E. Giannakopoulos, Massachusetts Institute of Technology, Department of Materials Science and Engineering, Cambridge, MA.

This presentation will provide a description of recent advances in the computational modeling of normal and sliding contact, and depth-sensing indentation at surfaces with homogeneous and graded compositions, or with electrical and mechanical coupling. The computational results, along with analytical models and systematic and controlled experiments, will be used to demonstrate how quantitative analyses of the contact phenomena can be used to (i)

assess the onset and progression of contact damage at surfaces, (ii) provide a means for the quantitative characterization of composition and mechanical properties at surfaces and thin films, (iii) derive guidelines for the design of processing parameters and compositional profiles which impart extraordinary resistance to contact damage at surfaces, and (iv) extract certain electrical and mechanical properties as well as activation energies for depolarization in coupled systems such as piezoelectric ceramics and composites. The size scale of experimental observations and analyses span the nanometer to the macroscopic dimensions. A number of immediate practical applications of the analyses for materials characterization, surface design, quality control in large volume commercial production, as well as failure analysis will be discussed for situations ranging from structural coatings, thin films, biomechanical and dental implants, and armors. Needs and challenges in the computational modeling of such problems will also be addressed.

2:15 PM O12.3
ANALYSIS OF DISLOCATION PROPERTIES FROM AB-INITIO GENERALIZED STACKING FAULT ENERGETICS. O.N. Mryasov[1], Yu N. Gornostyrev[2], M. van Schilfgaarde[3], A.J. Freeman[4] and D. de Fontaine[4]. [1]Univ. of California, Dept. of Material Science, Berkeley, CA, [2]Inst. of Metal Physics, Ekaterinburg, RUSSIA, [3]Sandia Natl. Labs, CA, [4]Northwestern Univ., Dept. of Phys. and Astronomy, Evanston, IL.

A method [1] to solve the two dimensional Peierls-Nabarro (2D PN) model with a generalized restoring force law was used to determine the dissociation splitting path for ordinary and super dislocations in B2 NiAl, $L1_0$ TiAl, CuAu and $L1_2$ Ni_3Al, Ni_3Ge and Ni_3Si intermetallics. In this scheme, ab-initio generalized stacking fault energetics (ie., the so-called γ-surfaces) are rigorously related to dislocation structure characteristics: (i) partials separation and (ii) partials width and "splitting" path (dependence of the screw on the edge component of displacements). Mechanisms of multi-valley Peierls relief formation and fundamental characteristics controlling the low-temperature dislocation motion are identified on the basis of γ-surfaces (calculated within the local-density approximation using the full-potential linear muffin-tin orbital method) and with a solution of the modified PN model with a discrete representation of the misfit energy. We find that (i) the splitting of dislocations may deviate significantly from what is usually assumed in the elasticity theory of dislocations, primarily because of the complex character of interatomic interactions; (ii) the role of stable and unstable faults can be identified; (iii) the relation between stacking fault energy and splitting distances may be significantly modified for narrow dislocations; (iv) the effect of sublattices on the Peierls stress (σ_P) can be large enough to reverse the usual relation $\sigma_P^{(s)} > \sigma_P^{(e)}$ (within isotropic elasticity theory) between mobilities of screw and edge dislocations.
Supported by the Office of Basic Energy Science, Division of Materials Science, of the U.S. Department of Energy under contract No. DE-AC04-94AL85000, and at NWU by the AFOSR (Grant No. F49620-98-1-0321).
1. O. N. Mryasov, Y. N. Gornostyrev and A. J. Freeman, Phys. Rev. B. **58**, 11927 (1998).

2:30 PM *O12.4
RETHINKING PLASTICITY: THE DEFORMATION DYNAMICS OF NON-CRYSTALLINE SOLIDS. Michael L. Falk, Harvard University, Division of Engineering and Applied Sciences, Cambridge, MA.

The microscopic processes that underlie plasticity in crystalline solids are relatively well understood in terms of dislocations. Nonetheless, making a connection between these dislocation dynamics and theories of macroscopic plasticity in order to understand phenomena such as the brittle/ductile transition in fracture remains elusive. Meanwhile, the micromechanics that give rise to a nearly identical range of phenomena in noncrystalline materials are relatively poorly understood. This route to understanding plasticity may prove simpler and could shed light on phenomena in both types of solids. Molecular dynamics investigations of fracture in model noncrystalline systems reveal that the relative ductility, ie fracture toughness, is sensitive to the particulars of the interatomic potential. Similar investigations of the inelastic shear response of this model system reveal analogous time and history dependent behavior to that seen in actual materials. Examination of the rearrangements underlying these phenomena leads to the conclusion that the basic unit of inelastic shear can be understood as a microscopic two-state system. A theory of the dynamics of inelastic deformation under shear is developed from this assumption and compared to the simulation results.

3:30 PM *O12.5
MECHANICAL DEFORMATIONS AND ELECTRONIC TRANSPORT IN CARBON NANOTUBES. Marco Buongiorno Nardelli, J.L. Fattebert, D. Orlikowski, C. Roland, B. Yakobson, Q. Zhao and J. Bernholc, North Carolina State University, Dept. of Physics, Raleigh, NC.

Carbon nanotubes are potentially the strongest materials known. They also have unusual electronic properties that may enable quantum nanodevices. Using state-of-the-art classical and quantum simulations, we have evaluated the mechanical, electronic and electrical response of carbon nanotubes to external deformations, such as strain and bending, and investigated how mechanically-induced transformations can lead to the formation of nanotube-based electronic devices. In strained nanotubes, the spontaneous formation of topological defects in an originally perfect graphitic network is observed. Following the formation of these defects, plastic or brittle behaviors can occur depending upon the external conditions and tube symmetry. The presence of addimers on strained carbon nanotubes leads to the formation of a novel class of defects that wrap themselves about the circumference of the nanotube. These defects modify the geometrical structure and induce the formation of nanotube-based quantum dots. Turning to electronic and electrical properties, we have evaluated the quantum conductance of various ideal and mechanically deformed nanotube structures. The calculations have been performed in the framework of a general scheme for computing the transport properties of extended systems that we have recently developed. It is based on the Landauer theory of coherent conductance and is applicable to any general Hamiltonian that can be described with a local orbital basis, including orthogonal and non-orthogonal tight-binding methods and full DFT approaches. Among the most significant results, we find that bent and/or stretched armchair tubes keep their metallic character for most practical purposes, while metallic chiral nanotubes undergo a bending-induced metal-semiconductor transition, which manifests itself in the occurrence of effective barriers for transmission. High defect densities affect substantially the coherent transport in individual nanotubes, while single point defects have only a limited effect. The role of nanotube-substrate interaction in modifying the electrical response has also been investigated.

4:00 PM O12.6
VISCO-ELASTIC/PLASTIC MECHANICAL MODEL OF NANOINDENTATION OF POLYMER FILMS USING A SPHERICAL INDENTER AND FINITE ELEMENT METHOD. Timothy C. Ovaert, Byung Ro Kim, Lihui Zhao, Dept. of Mechanical and Nuclear Engineering, Penn State University, University Park, PA.

Nanoindentation techniques are utilized extensively to characterize a wide variety of coatings, both hard and soft. In this investigation, a five-parameter axisymmetric visco-elastic/plastic finite element model has been developed to explain the behavior of polymer coatings subject to nanoindentation tests utilizing a 1 micron radius spherical indenter. The five parameters account for both visco-elastic and plastic response in the coating, and are determined by a process that matches the experimental load vs. indentation depth plot from the nanoindentation test with the load vs. indentation depth plot from the finite element model. The model is very useful for characterizing coatings that exhibit time dependent behavior and neither a distinct yield strength nor typical linear plastic behavior. The five parameters computed become a unique "thumbprint" for a particular coating, providing more unique information than that obtained by an ordinary elastic (unloading) approximation or visco-elastic (sinusoidal load) tests.

4:15 PM O12.7
RELAXATION MECHANISMS OF RESIDUAL THERMO-MECHANICAL STRESSES IN DUCTILE METALLIC THIN FILMS: ATOMISTIC AND CONTINUUM MODELING. M. Rauf Gungor and Dimitrios Maroudas, Department of Chemical Engineering, University of California, Santa Barbara, CA.

The reliability of ULSI microelectronic devices can be limited severely by failure of metallic thin-film components in complex multi-level configurations. Failure is driven by thermomechanical stresses induced in the films during thermal processing steps of device fabrication and by electric currents conducted through the metal during device service. The dynamics of failure is mediated by various defect formation and propagation phenomena involving point defects, dislocations, and larger-scale defects such as intergranular and transgranular voids in the metallic films. In this study, possible failure mechanisms are analyzed of ductile metallic thin films under applied tensile strain conditions based on a multi-scale approach that combines atomistic and continuum-scale computational methods. Our atomistic simulations are based on an EAM parametrization for copper and employ large million-atom supercells with cylindrical voids that extend throughout the thickness of the film. Results of molecular-dynamics simulations are presented over a range of applied biaxial tensile strains and thin-film temperatures for given crystallographic orientation of the film surface. Relaxation of the tensile stress in the films is found to occur through void growth by plastic deformation and self-diffusion mechanisms. Dislocation

emission from void surfaces and propagation through the film are demonstrated and their role in void growth and stress relaxation is analyzed in detail. In addition, mechanisms of nanovoid nucleation ahead of larger voids that can lead to film rupture at large applied strains are analyzed. A possible delamination mechanism also is suggested that can be initiated at metal/dielectric interfaces in devices due to interface roughening induced by plastic deformation in the film. The atomic-scale results are used to parametrize phenomenological kinetic models of strain relaxation. The results of the kinetic analysis are incorporated into mesoscopic self-consistent dynamical models of electromechanically-induced thin-film failure.