Recent developments in IMD: Interactions for covalent and metallic systems

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Abstract. We describe the recent developments of IMD (**I**TAP **M**olecular **D**ynamics), a general purpose program for classical molecular dynamics simulations on workstations and massively parallel supercomputers. As pair potentials are not entirely suitable for many classes of materials, several further types of interactions with many body forces have been implemented, thereby extending the range of applicability of IMD. IMD now supports, in particular, also EAM (Embedded Atom Method) potentials for the simulation of metals, and Stillinger-Weber and Tersoff potentials for the simulation of covalent systems, such as ceramics and semiconductors.

1 Introduction

IMD (ITAP Molecular Dynamics) [1] is a program package for classical molecular dynamics simulations [2]. It is designed to run efficiently both on single processor workstations and massively parallel supercomputers. IMD supports a large number of different thermodynamic ensembles, and a rich set of simulation options, in particular for the study of all kinds of mechanical properties. As the architecture of the program and its many options and possibilities have been described in previous papers [1,3], we report here only on the most significant recent developments. We also refer to the IMD Home Page [4], where information on the most recent version can be found.

Originally, IMD supported only tabulated pair potentials. For many classes of materials, the restriction to two-body forces is not adequate, however. One such class of materials are covalent systems like ceramics and semiconductors, where, in addition to two-body forces, at least three-body terms depending on the angle between bonds must be taken into account. Potentials frequently used for such systems are the Stillinger-Weber potential [5], and the Tersoff potentials [6]. These potentials, described in more detail in Section 2, are now implemented in IMD, and extend its range of applicability considerably.

Metals are another class of materials where the restriction to two-body forces is not adequate. For the description of metallic bonds many-body effects have to be taken into account. The Embedded Atom Method (EAM) [7], a semi-empirical, many-atom potential, provides a way to realistically model the mechanical properties of metallic systems with empty or filled d-bands. EAM potentials have been implemented in IMD as described in Section 3. With these, IMD is now suitable also for the simulation of metals.

All these new types of interaction are of short range, so that there was no need to change the general structure of the program. The parallelization and the parallel performance of the new interactions is therefore very similar to that of the pair potential case [1]. The parallelization is described in Section 4.

Besides the implementation and testing of new types of interaction, IMD has been used in production runs for the study of the physical properties of quasicrystals. In particular, fracture propagation, dislocation mobility under applied shear stress, and shock waves have been studied extensively (Section 5). For some of these simulations, a new kind of force boundary conditions had to be implemented, where certain atoms in boundary layers experience an additional force.

Very recently, IMD has been tested successfully on a new type of supercomputer architecture. It has been ported to the new HLRS Hitachi SR8000 machine using the OpenMP library. In contrast to the Cray T3E with its single processor nodes and distributed memory, each node of the SR8000 consists of eight CPUs with shared memory. The nodes on the other hand have distributed memory. Thus within one node communication is carried out via OpenMP and between the nodes via MPI. Although IMD has not been optimized completely up to now, we find very encouraging results already. The computing performance on the SR8000 is about half the performance on the T3E, whereas the communication performance is comparable. The lower computing performance is mainly due to the slower processors of the SR8000.

2 Simulations of covalent solids

A large class of materials of technological importance consist of covalently bonded atoms. Among these materials are structural ceramics, for example silicon carbide and silicon nitride, and semiconductors. In the case of structural ceramics, the covalent nature of the atomic bonds leads to the specific properties of these materials, that is, their high strength, their high melting point, and their low thermal conductivity. Grain boundaries in such materials are of great importance since their structure determines the mechanical properties. The presence of interfaces increases the fracture toughness of the inherently brittle ceramics.

A further example of covalent solids are semiconductors. Their mechanical properties play a role in the self-assembled epitaxial growth of superlattices of three dimensional islands. These quantum dot structures are important in laser technology.

2.1 Many-body potentials

Since covalent solids contain directed bonds that stabilize open atomic configurations, the usual pair potentials cannot be used because they lead to close-packed crystal structures. Instead, many-body potentials have to be used. There are mainly two classes of such potentials, potentials that can be split in two-body and three-body contributions, and the so-called bond order potentials. The most often used potential of the first class is the Stillinger-Weber potential [5]. For the second class, the Tersoff potential [6] is the best known. Both potentials have been implemented in the IMD program.

The Stillinger-Weber potential has the following form:

$$\Phi = \sum_{i<j} \Phi_2(r_{ij}) + \sum_{\substack{i\neq j,k \\ j<k}} \Phi_3(r_{ij}, r_{ik}, r_{jk})$$

where

$$\Phi_3(r_{ij}, r_{ik}, r_{jk}) = C_{ijk} f_c(r_{ij}) f_c(r_{ik}) \left(\cos\theta_{jik} + \frac{1}{3}\right)^2$$

Φ_2 is a pair potential, usually of Morse type, which depends only on the radial distance r_{ij} between the atoms i and j. Φ_3 is a three particle term which depends on the angle θ_{ijk} that is formed by three atoms with labels i,j,k and where j denotes the center atom. f_c is a cutoff function which restricts the interaction to the nearest neighbours and C_{ijk} is a force constant. In our implementation of the Stillinger-Weber potential, the pair potential and the cutoff function are read in from function tables. In order to compute the three particle contribution a neighbor list for each particle is generated at each time step which subsequently is used to compute the angle θ_{ijk}.

The Tersoff potential can be considered as a sum of pair potential contributions associated with each directed bond where the attractive part of the pair potential depends on the local environment of the atoms, that is, the bond order. The form of the Tersoff potential that has been implemented in IMD is the following:

$$\Phi = \frac{1}{2} \sum_{i\neq j} \Phi_{ij}$$

$$\Phi_{ij} = f_c(r_{ij}) \left[A_{ij} \exp(-\lambda_{ij} r_{ij}) - b_{ij} B_{ij} \exp(-\mu_{ij} r_{ij})\right]$$

$$\begin{aligned} b_{ij} &= \chi_{ij} \left(1 + \gamma_i^{n_i} \zeta_{ij}^{n_i}\right)^{-\frac{1}{2n_i}} \\ \zeta_{ij} &= \sum_{k\neq i,j} f_c(r_{ik}) \omega_{ik} g(\theta_{kij}) \\ g(\theta_{kij}) &= 1 + \frac{c_i^2}{d_i^2} - \frac{c_i^2}{d_i^2 + (h_i - \cos\theta_{kij})^2}. \end{aligned}$$

f_c is a cutoff function which is computed according to

$$f_c(r_{ij}) = \begin{cases} 1, r_{ij} < R_{ij} \\ \frac{1}{2} + \frac{1}{2}\cos\left(\frac{\pi(r_{ij}-R_{ij})}{S_{ij}-R_{ij}}\right), R_{ij} < r_{ij} < S_{ij} \\ 0, r_{ij} > S_{ij} \end{cases}$$

The indices of the parameters refer to the types of atoms.

In contrast to the implementation of the Stillinger-Weber potential there are no potential tables to be read in. The use of the neighbor tables is the same as in the case of the Stillinger-Weber potential.

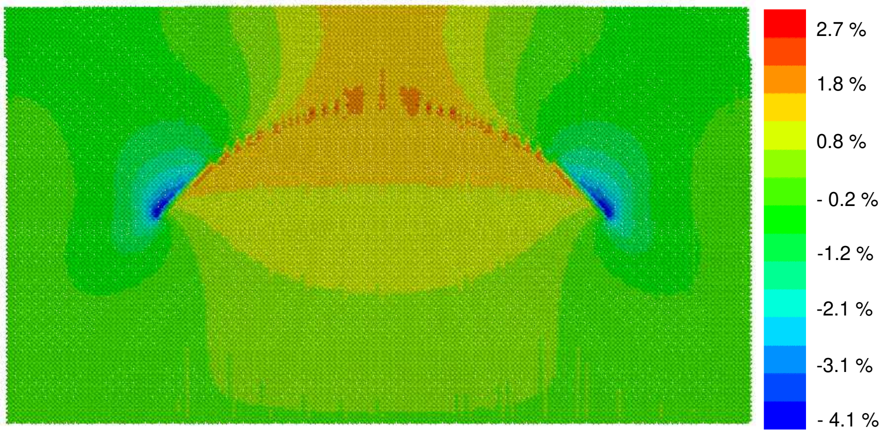

Fig. 1. Horizontal component of the strain field of a quantum dot consisting of a silicon substrate, a layer of germanium atoms, a lens shaped inclusion of germanium, and a covering of silicon atoms. Shown is a vertical section through the middle of the sample. The size is 43.44 nm × 24.44 nm.

2.2 Applications

The many-body potentials are presently used to determine the mechanical properties of grain boundaries in silicon carbide and silicon nitride. Furthermore, they are employed to study quantum dot heterostructures of semiconductors. From the point of view of molecular dynamics, there are at least two phenomena of interest in this respect.

First, the knowledge of the strain and stress fields at the atomic level of a single quantum dot is important for the determination of the electronic and optical properties, that is, the band structure, of quantum dots. Figure 1 shows the horizontal component of the strain tensor in a vertical section through a lens shaped germanium quantum dot embedded in silicon. The entire system consists of approximately 2.3 million atoms and has been relaxed with periodic boundary conditions applied horizontally, the first bottom layers fixed, and the top of the sample allowed to expand freely. For the simulation, 128 processors of the Cray T3E have been used and the computation time was 44 hours. The strain tensor was computed from the local atomic displacements with respect to an ideal silicon crystal (diamond lattice). The horizontal expansion of the quantum dot consisting of germanium atoms, which have a lattice constant 1.042 times larger than the one of silicon, as well as the compressive strain at the border of the lens can be seen clearly in the figure.

The second application of molecular dynamics simulations to quantum dots concerns the vertically self-organized growth of quantum dot superlattices during the epitaxial growth. This ordering is supposed to be driven by the strain field of the dots. Figure 2 shows the strain field of two vertically arranged quantum dots consisting of indium phosphide embedded in gallium indium phosphide. The position of the upper dot directly above the lower dot is energetically preferred compared to quantum dots laterally displaced.

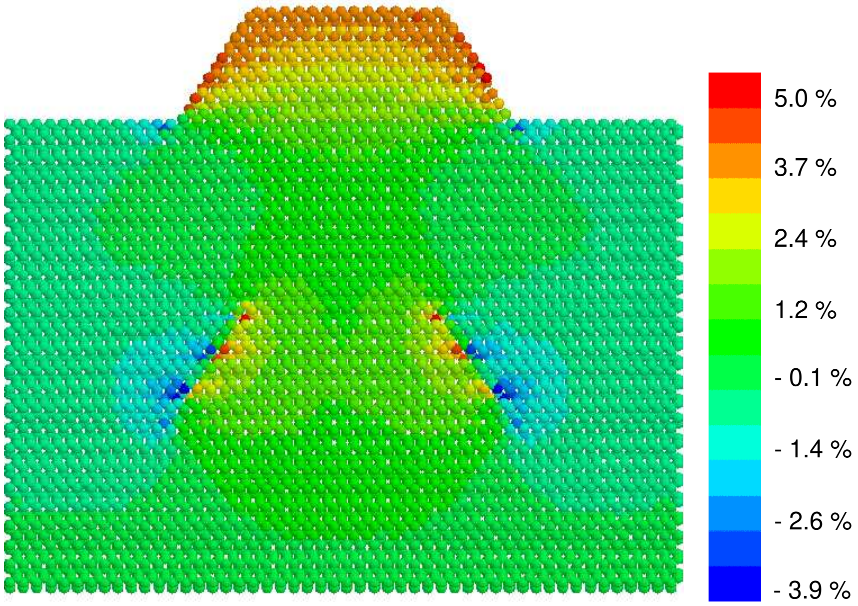

Fig. 2. Horizontal component of the strain field of two quantum dots consisting of truncated pyramids of InP atoms embedded in GaInP. Shown is a vertical section through the middle of the sample. The size is 20 nm $\times$ 17 nm.

3 Simulations of metals

3.1 Limitations of two-body potentials

Many-atom effects are neglected if only pairwise interaction of atoms is taken into account. Especially the consequence of the Pauli principle, that bonds become weaker when the environment gets "crowded", is not accounted for.

By comparing experimental data that might indicate many-atom effects with predictions of pair-potential models, the importance of many-body effects becomes clearly visible:

- The ratio E_c/k_BT_m between the cohesive energy and the melting temperature is about three times higher in metals than in two-body systems. This indicates that metals have some "extra cohesion" with respect to pair interaction systems.
- E_c/E_v, the ratio between vacancy formation energy and the cohesive energy is between 1/4 and 1/3 for metals, but 1 in two-body systems, because there the bonds have — independent from each other — a fixed energy contribution.
- The ratio of the elastic constants C_{12}/C_{44} in a cubic crystal is exactly 1 in two-body systems, but deviations in metals are very common.

These findings indicate that for a more realistic description of metals — in particular when dealing with non-equilibrium situations — one should incorporate many-body effects in the potentials.

3.2 The Embedded Atom Method

The Embedded Atom Method (EAM) was suggested by Daw and Baskes [8,7] as a way to overcome the coordination independence of the bond strength, while still being acceptably fast.

Ideas from the Density Functional Theory or the Tight Binding formalism suggest the following form for the total energy:

$$
\begin{aligned}
E_{tot} &= \frac{1}{2}\sum_i \sum_{j(\neq i)} \Phi_{ij}(r_{ij}) + \sum_i F(\rho_i^h) \\
\rho_i^h &= \sum_{j(\neq i)} \rho_{ij}^{at}(r_{ij}) \ .
\end{aligned}
$$

The pair potential term $\Phi_{ij}(r_{ij})$ can be interpreted as the electrostatic core-core repulsion, the cohesive term (the "embedding" energy) $F(\rho_i^h)$ can be seen as the energy the atom i gets when it is "embedded" in the local electron gas of density ρ_i^h. This local electron density is constructed as a superposition of contributions $\rho_{ij}^{at}(r_{ij})$ from neighbouring atoms j.

Also belonging to this analytical form are models like the glue model [9] and the Finnis-Sinclair potentials [10]. They vary in the functional form of F, Φ, and ρ^{at}. The potentials are obtained by fitting these functions to basic material properties like lattice constant, cohesive energy, elastic constants, vacancy formation energy, and sometimes also to the dilute heats of alloying or stacking fault energy. There are many reliable and well tested potentials for noble metals, nickel and their alloys, and for aluminium and some aluminium alloys.

With EAM potentials, the materials community has investigated many problems of interest [11] like point defects, grain boundaries, fracture, dislocations, alloying, etc.

3.3 Implementation in IMD

The functions F, Φ, and ρ^{at} are read from function tables. For greater flexibility they can have different step widths, starting and end points. There is no restriction on the number of different atom types.

The forces are calculated in two steps. In the first step, the forces and energy due to the pair potential $\Phi_{ij}(r_{ij})$ are computed, and the host electron density ρ_i^h of every atom is determined. In a second step, the embedding energy and its derivative are calculated from this density.

3.4 Applications

The EAM implementation in IMD is currently used to study the interaction of dislocations in fcc metals with surfaces (including surface defects like steps and grooves) and other interfaces.

The plastic deformation of metals is mainly carried by dislocations, line defects of the regular crystal lattice. In engineering simulations (e.g. crash tests) however, the material is treated as a continuum, described by constitutive equations. Currently the necessary parameters are empirically adjusted to experiments, but simulations could in the future allow the incorporation of information about atomistic processes in these constitutive equations [12]. The interaction between dislocations and surfaces becomes important if one considers the mechanical properties of small structures, e.g. ultra-thin metal films.

On the long range, interactions of dislocations are governed by their elastic field $\sim 1/r$. The short range interaction, however, is crucial when dislocations interact with defects that have an atomistic structure, like other dislocations or interfaces. In order to simulate dislocation-obstacle interactions by molecular dynamics, large simulation boxes have to be used to minimize boundary-effects.

Perfect surfaces act as weak pinning and dragging points for dislocation motion. The pinning force is increased by 2 orders of magnitude by defects on the surfaces. Figure 3 shows a monoatomic step on a (111)-surface of a nickel cristal acting as barrier for the motion of a split edge dislocation.

For the future, a detailed analysis of crack-dynamics is planned.

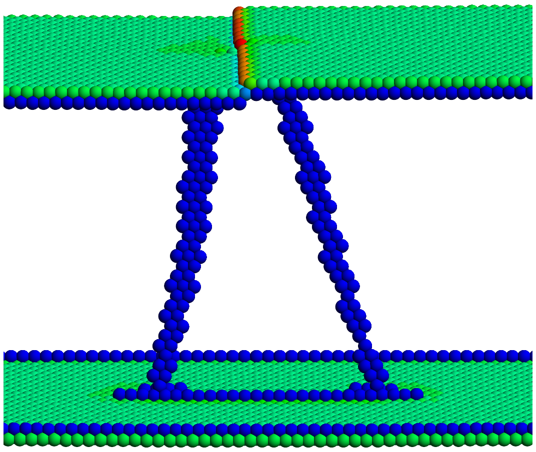

Fig. 3. A monoatomic step on the (111)-surface of a nickel cristal acting as obstacle to dislocation motion. View on the glide plane, only atoms with potential energy above a threshold are shown (red atoms have a high potential energy). Upon relaxation the initial perfect edge dislocation has split into two partial dislocations. At an applied shear stress of 75 MPa these dislocations can not overcome the surface step. The total size of the sample is 31 nm $\times$ 8 nm $\times$ 7 nm.

4 Parallelization and parallel performance

The data structure of IMD has been described previously [1]. The system is divided into cells whose diameter is the interaction radius, so that only particles in the same and in neighboring cells interact. Each processor deals with a rectangular block of cells, whose data is stored in the memory of that processor. In addition, each processor stores also a layer of buffer cells around its own block of cells. These buffer cells contain copies of neighboring cells located on different processors. Before the force computation, the buffer cells are filled with up-to-date copies of the original cells.

For pair potentials, the force between two particles is usually computed only once, and added with different signs to the force variables of the two particles (actio=reactio, AR). If the two particles reside on different processors, one of the force contributions gets stored in a buffer cell. One then has either of two choices: send back forces in buffer cells to the processor with the original cell, and add them there (AR across processor boundaries), or compute the forces for inter-processor pairs twice, once on each processor. On computers with fast communication hardware like the Cray T3E, AR across processor boundaries is faster.

AR across processor boundaries is used also for covalent potentials (both for Stillinger-Weber and Tersoff potentials). The force for each pair or triple of atoms is computed only once, and in the end the forces in buffer cells are added to those in the original cells. However, for the computation of the many-body interactions also a neighbor table has to be built up first. This poses the same problem as with the forces: they may reside partly in real cells, and partly in buffer cells. In order to avoid the communication of neighbor tables, we compute them completely locally, by running over inter-processor pairs of atoms a second time.

For EAM potentials, there is less to be gained from AR across processor boundaries. Except for the core-core potential, the force contribution is not symmetric in the pair of atoms, at least if the atom types are different, so that one has to compute it twice anyway. Therefore, at the moment no use of AR is made, but it is planned to be added later where apropriate. The force computation is done in two steps. In the first step, the forces of the core-core potential and the host electron density are computed. The host electron density is then sent to the buffer cells on the neighboring processors, so that in the second step the forces from the host electron density can be computed.

The communication requirements for the force computation of covalent and EAM potentials are essentially the same as for pair potentials. For the latter, the communication time is is typically a few percent of the total simulation time [1]. It depends on the amount of work each processor has to carry out. The more cells (or atoms) a processor has to deal with, the less important is the communication overhead. The computation requirements of the new interactions may differ from that of pair potentials, however, so that the relative importance of communications will also differ. The computation of EAM potential forces is considerably more complicated, while the number of interaction neighbors is comparable to that of typical pair potentials. This means that the communication overhead is smaller than for pair potentials, typically by a factor 2. For covalent potentials, in particular Tersoff potentials, the force computation is also more complicated, but there are much fewer interaction neighbors, so that the the force computation is still relatively fast. The communication overhead is therefore comparable, or slightly bigger than for pair potentials.

5 Further applications of IMD: fracture and shear simulations, shock waves

Besides the implementation and testing of new interactions, IMD has been used (with classical pair potentials) for the study of physical properties of quasicrystals. The fracture propagation studies in two-dimensional decagonal quasicrystals have been extended to higher temperatures. It is now possible to study crack propagation in a more realistic context. At higher temperatures the crack tip gets blunted, and the paths of the dislocations which are emitted from the crack tip are no longer straight lines but get disturbed rather often. The mobility of edge dislocations in icosahedral quasicrystals under applied shear stress have been simulated [13,14]. It turned out that dislocations are pinned at cluster centers. Moreover, the dislocations leave a clearly visible stacking fault behind them. This is different from an ordinary crystal, where no visible defect remains after a dislocation has passed. Finally, shock waves have been simulated in icosahedral quasicrystals and structurally similar crystals [15]. The behavior of these materials is much more complicated than that of simple crystals, where stacking faults are generated, but the structure itself remains undestroyed. In the materials studied here the structure is increasingly fragmented with the growing strength of the shock waves, until the structure finally turns completely amorphous at very strong shock wave intensity.

6 Acknowlegdments

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