Simulation of dislocations in icosahedral quasicrystals with IMD

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Abstract. We report on recent investigations performed with IMD (**I**TAP **M**olecular **D**ynamics), a general purpose program for classical molecular dynamics simulations on workstations and massively parallel supercomputers. Especially the simulations of dislocations in icosahedral quasicrystals are described. The quasiperiodic structure leads to new interesting properties. The visualization of a dislocation is much more complicated than in periodic crystals and is presented in detail. An overview of the software used is also provided.

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, among which is the microcanonical or NVE ensemble, the canonical ensemble (NVT) and the NPT ensemble where the pressure instead of the volume of the sample is fixed. IMD offers several options for the study of all kinds of mechanical properties. These enable various deformations of the sample like shear deformations, or loading the sample for the study of cracks. For further details we refer to the IMD home page [3], where more information on the most recent version can be found.

Several forms of atomic interactions are supported, which are suitable for different types of materials. The simplest interactions are given by central pair potentials. Such two-body potentials have mainly been used for quasicrystals, where more refined potentials are still lacking. For an adequate description of most materials, it is necessary to use more complicated potentials, which include also three-body or many-body terms. For covalently bound materials, like ceramics and semiconductors, it is essential to include also three-body terms in the interactions, which depend on the angle between adjacent bonds. Such interactions are provided in the form of Stillinger-Weber potentials [4], and Tersoff potentials [5]. Adequate interactions for metals must take into account the delocalized conduction electrons and the consequences of the Pauli principle. This can be achieved with the Embedded Atom Method (EAM) [6], which has been implemented in IMD. In EAM potentials, in addition to the pair force a cohesive force term is calculated for each atom, which depends on the local electron density into which the atom is "embedded". This density is calculated from contributions of neighbouring atoms.

The parallelization method and the parallel performance of the pair interactions has been described in [1]. More details on the implementation and the parallelization of the different many-body interactions can be found in [7]. Besides the implementation and testing of new types of interaction, IMD has mainly been used in production runs for the study of the physical properties of quasicrystals.

2 Simulation of dislocations in icosahedral quasicrystals

The structure of a periodic crystal can be described as a periodic arrangement of copies of a single structural unit, the unit cell of the crystal. For quasicrystals, which are quasiperiodic rather than periodic, a similar description requires at least two such structural units. Geometrically, these units can be described as *tiles* decorated with atoms, which are forming a quasiperiodic tiling.

Like in ordinary crystals the plasticity of quasicrystals is mainly due to the generation and motion of dislocations. This has been proved in several experimental investigations [8]. While quasicrystals are brittle at room temperature there is a ductile high-temperature regime starting at about 70-80% of the melting temperature.

Theoretical work considering the deformation process qualitatively has not helped much in understanding the microscopic properties. Therefore MD simulations of shear deformations at various temperatures have been performed on a three-dimensional icosahedral quasicrystalline model system: the 3D-Penrose-Tiling [9] decorated with two types of atoms according to a structure model by Henley and Elser [10]. This atomic structure is stable if the atoms interact via simple pair potentials like the *Lennard-Jones potential*

$$V(r_{ij}) = \varepsilon_{ij}((\frac{\sigma_{ij}}{r_{ij}})^{12} - 2(\frac{\sigma_{ij}}{r_{ij}})^{6}) \tag{1}$$

where i and j denote the two atom types [11]. The structure was relaxed and a shear deformation was applied at various temperatures.

2.1 Method

In former simulations [12] a dislocation had been put into a quasicrystalline sample of 1,504,080 atoms by applying a displacement field of Peierls-Nabarro type

$$u_x(x) = -\frac{b}{2\pi} \arctan \frac{x}{\zeta} \tag{2}$$

where b is the length of the Burgers vector $\mathbf{b}$. This Burgers vector was obtained by an energy consideration according to [13]: the sample was cut along an easy glide plane (a plane with a large separation between the atoms) and the two halves were shifted rigidly. The mismatch energy was calculated, and shift directions corresponding to its minima were chosen as candidates for Burgers vectors.

The sample was relaxed by a microconvergence method [14] corresponding to an undercooling of the sample. The dislocation could be stabilized in this way. Then a shear deformation of 2.1% along the direction of the Burgers vector was applied. The dislocation moved through the sample creating a stacking fault in its wake. Both simulations required partly fixed and partly free boundaries, which causes large inhomogeneities in the distribution of system properties like temperature and pressure.

Therefore, in a second series of simulations we decided to apply the shear deformation in a continuous way, using a non-equilibrium MD (NEMD) algorithm. NEMD had previously been used for the simulation of transport processes, especially for the computation of the shear viscosity of fluids. In that case a Couette flow is modeled by the application of a linear velocity field. The shear viscosity is the ratio of the pressure and the applied velocity gradient. This has several advantages over the linear response method where the time correlation functions are integrated: it is less expensive in computer time and avoids the poor signal-to-noise ratio of the latter. As NEMD algorithms require a modification of the equations of motion they are often called *synthetic algorithms*.

The formal analogy between a Couette flow and a shear deformation can be used to formulate a Hamiltonian of a dynamics explicitly containing the shear deformation. Such a deformation adds energy to the system that can be expressed in terms of the infinitesimal deformation $\nabla_\alpha u_\beta$ where u_β is the displacement

$$\delta E = \sum_{\alpha,\beta} \sigma_{\alpha\beta} \delta(\nabla_\alpha u_\beta). \tag{3}$$

The rate of dissipated energy is then

$$\dot{E} = \sigma_{\alpha\beta} \delta(\dot{\nabla}_\alpha u_\beta) = \frac{d}{dt} \left[\sum_i (q_\alpha^{(i)} p_\beta^{(i)}) \nabla_\alpha u_\beta \right] \tag{4}$$

where $q_\alpha^{(i)}$ and $p_\alpha^{(i)}$ are the position and momentum coordinates of the i-th particle. With H_0 being the Hamiltonian of the unperturbed system the following ansatz is chosen

$$H_{Doll} = H_0 + \sum_i (q_\alpha^{(i)} p_\beta^{(i)}) \nabla_\alpha u_\beta \tag{5}$$

yielding the equations of motion

$$\dot{q}_\alpha^{(i)} = \frac{p_\alpha^{(i)}}{m_i} + q_\beta^{(i)} \nabla_\alpha u_\beta, \quad \dot{p}_\alpha^{(i)} = f_\alpha^{(i)} - q_\beta^{(i)} \nabla_\alpha u_\beta \tag{6}$$

The fantasy name *Doll's tensor* shall stress that we deal with a synthetic algorithm here. The dissipation is the same as the one from linear response theory. It even does not change if the velocity gradient is transposed in the momentum equation:

$$\dot{p}_\alpha^{(i)} = f_\alpha^{(i)} - q_\beta^{(i)} \nabla_\beta u_\alpha. \tag{7}$$

Contrary to the Doll's tensor equations, these equations of motion reduce to the Hamiltonian equations if the velocity field is constant. Their name *SLLOD equations* shall remind of the transposition.

The SLLOD-ensemble must be applied together with periodic boundary conditions in all directions. As energy is permanently added to the system, a thermostat has to be used. In our case a Nosé-Hoover-Thermostat [15] was applied, where a heat bath is coupled to the system, which effectively rescales the momenta of the particles. A critical parameter is the mass of the thermostat, which must be chosen appropriately.

2.2 Visualization

A dislocation is a line defect that can be created by an incomplete slip of two crystal halves along a *glide plane*. The dislocation line is the boundary between the displaced and undisplaced parts of the crystal. If the dislocation *glides* — moves within its glide plane — the slipped region is enlarged. In periodic crystals the atoms within a narrow region around the dislocation line — the *dislocation core* — are displaced from their ideal positions, but far away from the dislocation line the atoms are again in perfect order, provided the Burgers vector of the dislocation is a primitive translation of the crystal lattice. The same is true for the atoms in the wake of a gliding dislocation. This is a direct consequence of the translational periodicity of the crystal lattice. Quasicrystals lack such a translational periodicity, so that every dislocation leaves behind a *stacking fault*, a plane of atomic misalignment.

In ordinary crystals every atom of a given type has the same local environment, so that all these atoms have the same potential energy. Therefore, dislocations in ordinary crystals can easily be identified by plotting only the atoms having an excessive potential energy. They form narrow "tubes" indicating the position of the dislocation line. The situation is more complicated in a quasicrystal. In our model, for example, the atoms of one type are subject to potential energies varying over a range from -24 to -15 LJ units, while the excess energy of an atom in the dislocation core is only a few LJ units. We have implemented the following visualization methods in IMD:

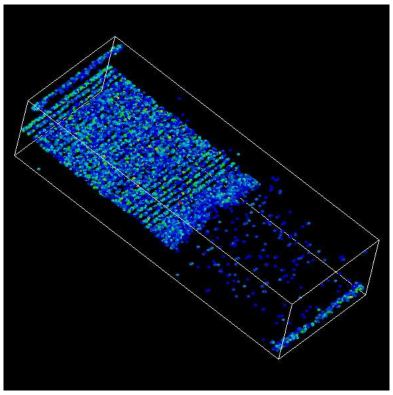

Fig. 1. Visualization of the dislocation line and the stacking fault. Only atoms with a large potential energy compared to the initial value are plotted.

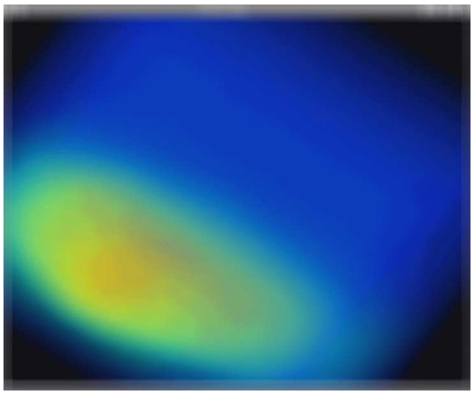

Fig. 2. Volume visualization of the distribution of the kinetic energy. Like in Fig. 1 the energy increases from blue to red colours.

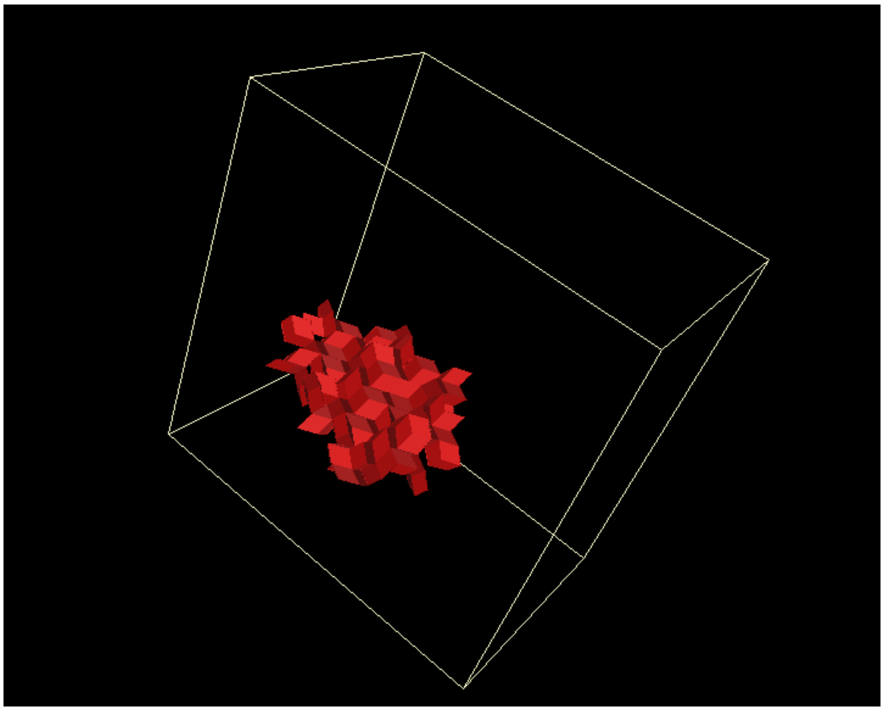

Fig. 3. Visualization of the boundary surface between defective and undefective regions. Rhombohedra with distorted bonds form the defective region.

- *Potential energy difference.* The potential energy of an atom is compared to its energy in the initial state not containing the defect. If the difference exceeds a threshold the atom is plotted (Fig. 1). The dislocation core and the stacking fault cannot be separated by this method.
- *Kinetic energy.* The kinetic energy is sampled into an array of boxes. We obtain pictures like Fig. 2 which do not allow for a precise determination of the current position of the dislocation.
- *Retiling of the sample.* The initial rhombohedral tiles and their edges — in the following referred to as *bonds* — are stored in a list. For a given snapshot of the simulation the rhombohedra with strongly distorted bonds are deleted from the list. In general, each rhombohedron face belongs to two rhombohedra except those whose rhombohedra have been deleted. By plotting the faces where one rhombohedron is deleted, but the other is not, we obtain a surface separating the defective and the undefective region (Fig. 3). This allows for a precise determination of the current position of the dislocation.

2.3 Results

Simulations at three different temperatures corresponding to 20, 50, 66% of the melting temperature have been performed. The stress-strain curves (Fig. 4) show a behaviour corresponding to Hooke's law. At deformations between 14 and 17% there is a yield drop indicating the onset of plastic deformation. The critical strain decreases with increasing temperature while both the critical stress and the shear modulus increase. This unphysical behaviour is a consequence of the NVT ensemble, where the volume is kept constant. The hydrostatic pressure of the sample is larger for the high temperature simulations. Simulations with an NPT ensemble provide a solution to this problem, but the resulting volume fluctuations are very difficult to control.

Due to the periodic boundaries in all directions, a dislocation dipole has nucleated enclosing a stacking fault (Fig. 3). The two dislocations move away from each other, enlarging the stacking fault. The dislocation line bulges out during its motion. This is due to pinning at *clusters*, atomic sites of high symmetry [16]. A detailed analysis of the dislocation motion is in progress.

3 Further applications: fracture and shock wave simulations

IMD has also been used for the study of other mechanical properties of quasicrystals. Propagation of mode-I cracks have been studied in two-dimensional decagonal ordered and randomized model quasicrystals in dependence of temperature. For low temperatures and perfect ordered systems the crack is propagating by dislocation emission. Due to the quasiperiodicity, dislocations,

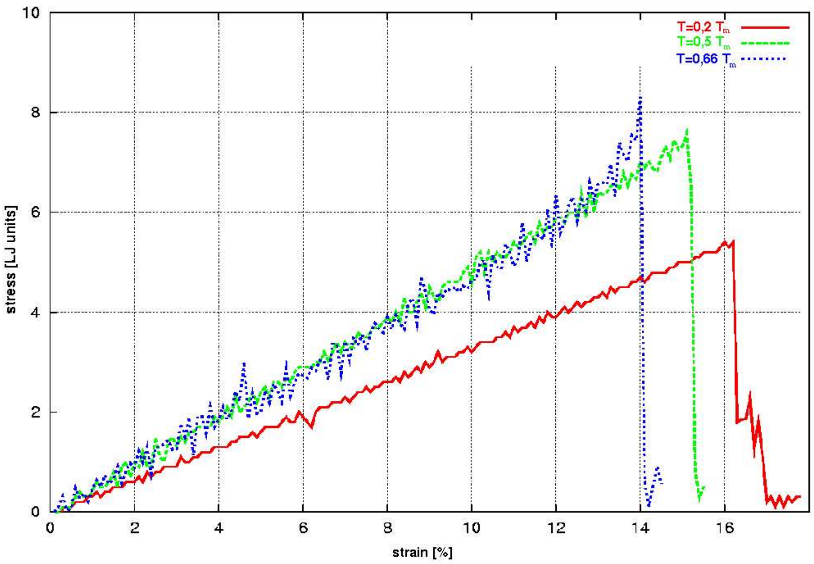

Fig. 4. Stress-strain curve for various temperatures.

which are emitted in a zig-zag fashion, are followed by phason walls. Along these walls the interfacial energy is lowered, and the material is opening, causing a rough cleavage surface. At intermediate temperatures this dislocation emission process still works, but with much longer dislocation paths. At more elevated temperatures the formation of voids in front of the crack was observed.

The phason-phonon coupling constant of a two-dimensional quasicrystalline model system was determined using a decomposition of its elastic energy into symmetry invariant terms. By applying a phononic strain to the samples at fixed phasonic strain, parabolas were obtained whose vertices are displaced from zero. From this displacement the coupling constant can be calculated. By varying the choice of the phasonic strain the complete set of quasicrystalline elastic constants could be determined.

Finally, shock waves have been simulated in icosahedral quasicrystals and structurally similar crystals [17]. The behaviour 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 increasing strength of the shock waves, until the structure finally turns completely amorphous at very high shock wave intensity.

4 Software for Visualization

For the visualization of volume data, the package `volimd` [18] is available. It can be used to render energy distributions, for example. A new version of the volume visualization module is currently under development in cooperation with J. Schulze-Döbold of SFB 382 [19,20].

Even though IMD provides socket connections for online visualization, to which `volimd` can connect, this approach has turned out to be not very useful, at least in three dimensions. The reason is that elaborate post processing is necessary to filter the *interesting* information from the enormous amount of *available* information.

Our main interest lies in the determination of strongly localized defects of the size of a few atoms in a bulk of several million atoms. If the volume visualization is refined to the atomic level the data files soon become intractably large. If the defect atoms are singled out by one of the methods described in section 2.2, the volume picture is still too diffuse to be useful.

Currently the most practicable way to figure out what happens in the simulation box is to postprocess the data after the simulation by methods like those described in section 2.2, and then to display the results by programs which have been developed for molecular modelling and for the representation of polyhedra.

Some of the programs found useful are be listed below. Most of them are freely available, and all of them provide output routines which permit to create picture files.

- Atom positions can efficiently be visualized using the program `rasmol` available at www.OpenRasmol.org.
- A very useful program to visualize polyhedra and the tiles of quasicrystals is `geomview`, available at www.geomview.org. Input for `geomview` can easily be created by `qhull`, which has originally been developed to compute convex hulls and Voronoi decompositions. It is available at www.geom.umn.edu/software/download/qhull.html.
- VMRL scenes have been created to display vector fields, which can be rendered by any VRML viewer, like the OpenInventor based `ivview`, or the older `vrweb` (available at www.iicm.edu/vrweb). More about VRML can be found at www.vrml.org.

More insight into physical processes happening during simulation can be achieved if the dimension "time" is taken into account. Often complicated phenomena are understood only if the simulation run is presented as a computer movie.

If embedded into shell-scripts, `rasmol`, `geomview` or even `gnuplot` can be used to generate sequences of pictures. With `montage` from the ImageMagick package pictures may be combined to display several aspects of the simulations at the same time. The sequence of pictures can then be transformed into computer movies with the SGI `mediaconvert` or `dmconvert` programs. For high-quality movies the Stuttgart computing center RUS provides support through its Videoservice.

A number of computer movies generated from IMD simulations can be found on the IMD home page [3].

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