

Comment on "Formation of a Dodecagonal Quasicrystalline Phase in a Simple Monatomic Liquid"

In a recent paper Dzugutov [1] describes a molecular dynamics cooling simulation where he obtained a large monatomic dodecagonal quasicrystal from a melt. The structure was stabilized by a special potential [2] designed to prevent the nucleation of simple dense crystal structures. In this Comment we will give evidence that the ground state structure for Dzugutov's potential at $T = 0$ and $P \approx 0$ is an ordinary bcc crystal. More detailed descriptions of the phase diagram of the Dzugutov potential are in preparation [3,4].

The results have been obtained by molecular dynamics (MD) simulation. The constraint method [5] was used to simulate N - V - T or N - P - T ensembles, together with an extension that permits constant temperature or pressure gradients also. The equations of motion are integrated by a fourth-order Gear algorithm. The time increment was $\delta t = 0.0005$ (all quantities are given in Lennard-Jones units).

The initial structures are generated by melting and equilibrating crystals with 250, 500, or 1024 atoms at $T = 4.0$ and densities of $\rho = 0.863, 0.875, \text{ and } 0.867$, respectively for 10^5 time steps. The 250 atom sample was subsequently cooled at rates of $-0.002/\delta t$ and $-0.001/\delta t$. With the faster rate one obtains a tetrahedrally close packed (tcp) structure [6] where all atoms are 12-, 14-, or 15-fold coordinated and surrounded by Frank-Kasper polyhedra. The quasicrystal also belongs to this class, but the bcc does not since its coordination polyhedron is not of the Frank-Kasper type. The radial density functions of all the structures are quite similar, although the first maximum differs in shape between bcc and tcp in a good quality sample.

Cooling the 500 atom system at $-0.001/\delta t$ and the 1024-atom system at $-0.00025/\delta t$ also leads to bcc crystals. At faster rates the large system shows partial ordering, but the structure as a whole did not form an ordered crystal.

Cooling the fluid at high *constant pressure* always yields a bcc structure which sometimes evolves into a fcc structure at low enough temperature. At $P = 5$ a tcp structure is obtained at $-0.002/\delta t$ and a bcc crystal at $-0.001/\delta t$. If the fluid is compressed at constant temperature the results are similar. In the range from $T = 2.5$ to 1.0 the bcc structure forms at a compressing rate of $0.1/\delta t$. At $T = 0.6$ a twinned crystal was found with a few tcp atoms. It has been shown by density functional calculations [3] and energy minimization calculations [4] that the bcc structure has indeed the lowest enthalpy at low pressure and $T = 0$. At increasing density and pressure the tcp σ phase [6] (a low order approximant

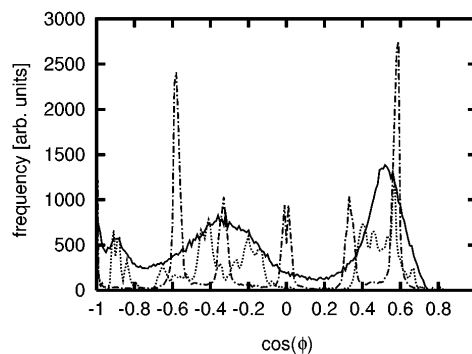


FIG. 1. Bond angle correlation function for nearest neighbor atoms. The histogram gives the frequency of angles as a function of the cosine. The liquid (full line) is similar to a tcp structure (dotted line), whereas the perfect bcc structure is completely different, but close to the nucleated structure (dash-dotted line).

of the quasicrystal) becomes more stable, but at high density the fcc structure is the most stable. The difference between bcc and σ phase is rather small so that it is currently not clear which one is more stable at low and moderate pressure and $T > 0$.

At high cooling rates one obtains a glass. In an intermediate range a tetrahedrally close packed structure or a quasicrystal is favored since its structure is close to the fluid structure (Fig. 1). At slow cooling rates and higher pressures the system has enough time to nucleate into the stable bcc structure. With increasing system size it takes more and more time for the fluid to reorder into the bcc structure, therefore the tcp structure will be generated.

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