Dzugutov Replies: The dodecagonal quasicrystal [1] is not the ground state for the system [2] investigated by Roth [3]. It was concluded [4] to be stabilized by the entropy produced by thermally activated phason dynamics. This implies its low-temperature instability with respect to a lower-energy phase. Two conceivable candidates for the latter are σ phase, a closest approximant of the quasicrystal, and bcc. At the density $\rho = 0.84$, at which the quasicrystallization was observed, bcc has lower energy than the σ phase for $T < 0.3$ (Fig. 1). One possible assumption then may be that the high rate of cooling reported by Roth, which was about 2 orders of magnitude faster than the rate of quasicrystal formation [1], brought the system to bcc-dominated temperature domain before a competing pattern of nucleation could develop. However, because of the exceedingly slow rate of the liquid dynamics at that temperature range, such a transformation would be inconceivable within the relevant time scale. This infers that the bcc transition occurred at a higher temperature.

In order to locate the crystallization temperature, we cooled the liquid at $\rho = 0.84$ in a stepwise manner. At each step, after reducing the temperature by $\Delta T = 0.01$, the system was thoroughly equilibrated. For the systems of 250, 432, and 686 atoms, rapid bcc crystallization was observed at $T = 0.7$, 0.64, and 0.56, respectively. Thus, the crystallization occurs in the temperature domain where bcc is energetically inferior to the σ phase, which indicates its kinetic origin. Another important aspect of this transition is its strong size dependence. Note that the system of 16 384 atoms remained in the equilibrium liquid state at $T = 0.5$ [5].

This observation may be explained as follows. Under cooling, quasicrystal-forming liquids show a tendency to form a network of local icosahedral units [6]. The increase of the correlation length leads to undercoolability [7], high viscosity [8], non-Arrhenius behavior and anomalous diffusion [5]. When the correlation length, growing with cooling, exceeds the system size, periodic boundary conditions interfere with the relaxation process, reducing the accessible volume of the phase space. The liquid, losing its entropy-induced stability, is then forced to transform into the bcc-related liquid phase which is entropically favorable due to its less pronounced structural correlations. Eventually it crystallizes into bcc solid. At a faster cooling rate, this kinetic scenario is blocked by glass formation.

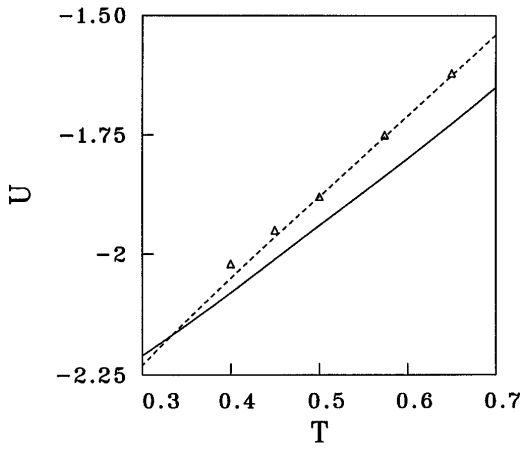

FIG. 1. Potential energy, per atom, of different phases with the pair potential [2], at the density $\rho = 0.84$. Solid line, σ phase; dashed line, bcc; triangles, quasicrystal [4].

Thus, the size-related effect observed here allows one to explore the increase of the correlation length in an icosahedrally ordered liquid under cooling.

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