Locally Preferred Structure in Liquids

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A large body of experimental and numerical work on supercooled liquids has confirmed the tendency, increasing on lowering temperature, to form local icosahedral order. The competition between extension of local liquid order, different than that of the underlying crystal, and global constraints associated with periodic tiling of the entire space, gives rise to geometric frustration[1], which has been connected to the dynamic slowing down in the supercooled liquid state[2].

The nature of the local icosahedral order in bulk liquids is still unclear. Indeed, it is a well known result that for an isolated system of 13 atoms interacting via spherically symmetric potentials, like the Lennard Jones potential, the lowest energy configuration is not a close packed structure but the icosahedron. This is due to the fact that in an isolated cluster most of the energetics is related to the surface. This situation does not, of course, occur in bulk liquids.

We propose a method to determine the locally preferred structure in bulk liquids[3]. The latter is determined numerically as the ground state of the “effective” energy surface of small clusters embedded in a liquid-like environment. The effective energy is the sum of the intracluster interaction potential and of an external field that describes the influence of the surrounding bulk liquid at a mean-field level. In this way we minimize the surface effects present in isolated clusters without introducing full blown geometric frustration (which hinders the manifestation of icosahedral order).

We find that the locally preferred structure of the Lennard Jones liquid is the icosahedron and propose a general method to predict the local preferred configuration in molecular systems.

References

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