

Very Efficient Methods for Evaluation of Properties of Crystalline Phases by Molecular Simulation

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The crystalline structure of a solid-state material has a profound effect on its mechanical, electronic, chemical, magnetic, and optical properties. Indeed almost every practical use of a solid-state material hinges on its crystalline structure as much as its atomic makeup. Consequently, a key capability in any theory- or computation-based effort to develop new materials or to compute their properties is the ability to predict the crystalline form adopted by a given set of atomic species at a given thermodynamic state. This in turns requires the ability to compute solid-state properties with high efficiency and accuracy. Arguably our inability to do this reliably is one of the biggest obstacles to the design of new materials exhibiting desired properties. In this work we present a set of new methods to compute properties of crystalline phases. The methods are highly efficient to the extent that the modeled system is harmonic, but at the same time they do not rely on the harmonic character to produce correct results. Rather they yield data that focus strongly on characterizing the anharmonic contributions directly, so that precise results can be obtained without noise introduced from sampling harmonic effects (which can instead be characterized exactly via lattice dynamics methods). The methods are applied to several model systems, including the Lennard-Jones model, an extended Finnis-Sinclair model of bcc tungsten, clathrate hydrates modeled with the TIP4P potential, and fcc aluminum modeled using density functional theory. The approaches are shown to provide data of precision equal to that given by conventional methods, while requiring orders of magnitude less computation time.