A Continuous-Path Approach to Crystal Structure Prediction

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Abstract

The prediction of stable and metastable crystal structures at high pressures and finite temperatures remains among the central challenges in condensed matter physics. While existing heuristic algorithms, such as evolutionary structure searching or random sampling, have been widely and successfully used and have driven significant discoveries, they intrinsically lack the physical continuity required to accurately map complex free-energy landscapes. In this talk, we introduce a novel approach to phase space exploration: the cyclic strain-kinetic method. Rather than relying on discrete structural recombination or purely stochastic sampling, this method employs continuous pathways to efficiently traverse multidimensional potential energy surfaces at arbitrary pressure and temperature conditions. The algorithm deterministically escapes local topological traps and maps the true thermodynamic funnels of the system. We will demonstrate the applications of this method through a series of cases under extreme pressure, ranging from the phase transitions of simple elements to the high-pressure behavior of complex compounds. Finally, we will discuss the implications of this continuous-path approach for accelerating the discovery of novel high-pressure phases.