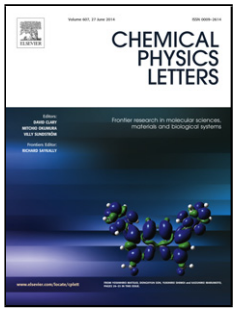

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Multi-center semi-empirical quantum models for carbon under extreme thermodynamic conditions

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Abstract

We report on the development of many-body density functional tight binding (DFTB) models for carbon, which include either explicit or implicit calculation of multi-center terms in the Hamiltonian. We show that all of our methods yield accurate eigenstates and eigenfunctions for both ambient diamond and transitions to molten, metallic states. We then determine a three-body repulsive energy to compute accurate equation of state and structural properties for carbon under these conditions. Our results indicate a straightforward approach by which many-body effects can be included in DFTB, thus extending the method to a wide variety of systems and thermodynamic conditions.

1. Introduction

The equation of state of materials under extreme pressures and temperatures (P $>$ 10 GPa, T $>$ 1000 K; 1 GPa = 10 kbar) is of great importance for understanding planetary interiors and the chemical reactivity that occurs under strong dynamic compression. Knowledge of the high pressure-temperature properties of simple compounds such as solid phases of carbon is needed to devise models of Neptune and Uranus[1], brown dwarfs[2], and extra-solar carbon planets[3]. In addition, carbon is a major component of candidate materials for the design of fusion capsules for the National Ignition Facility (NIF)[4]. Late-time soot condensation is known to account for significant energy release in detonating carbon-rich energetic materials[5]. Similar questions arise in origin of life studies, where the creation of organic

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