Molecular Structure Determination of Solid Carbon Dioxide Phase IV at High Pressures and Temperatures Based on MP2 Theory

Yanqiang Han[1], Jinyun Liu[2], and Jinjin Li[1]

[1]Shanghai Jiao Tong University
[2]Anhui Normal University

May 5, 2020

Abstract

Carbon dioxide has attracted considerable attention owing to its physics and abundant polymorphs. Despite decades of extensive experiments and theoretical simulations, the structure and properties of carbon dioxide under extreme pressures and temperatures are yet to be properly understood. Particularly, the intermediate phase IV of solid carbon dioxide, which separates the molecular phases at low pressures from the non-molecular phases at high pressures, has not been fully investigated, and its structure remains controversial. Here, based on the second-order Møller-Plesset perturbation (MP2) theory and the embedded fragment method, we study the crystal structure, equation of state, and Raman spectra of solid carbon dioxide phase IV at high pressures and temperatures. We demonstrate that the solid carbon dioxide phase IV is a molecular structure that remains in a molecular state rather than the bent state shown in other literatures, which is consistent with the experimental work by Datchi et al. and denies the observed results by Park et al. The proposed work is of great significance in determining the structure of the high-pressure phases of carbon dioxide and further exploring the new phase of molecular crystals.

Hosted file

`CO2phaseIV-Li.pdf` available at https://authorea.com/users/296866/articles/425886-molecular-structure-determination-of-solid-carbon-dioxide-phase-iv-at-high-pressures-and-temperatures-based-on-mp2-theory