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Structure and dynamics of protonated Mg_2SiO_4: An ab-initio molecular dynamics study

Michael Haiber, Pietro Ballone, and Michele Parrinello

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Germany

Abstract

We studied structural and dynamical properties of H^+ absorbed in Mg_2SiO_4 by ab-initio molecular dynamics. We first calculated the $T = 0$ equation of state of pure forsterite as a function of pressure, and we determined the relative stabilities of the olivine, β-spinel, and spinel polymorphs. The results show that the ab-initio model successfully reproduces the known structural properties of the system. In the protonated phases, in agreement with experimental evidence, our computations show that H^+ is absorbed preferentially in the β-spinel phase. The most stable absorption site is located close to the O1 atom, which is coordinated by five Mg^{2+} cations and not directly bound to Si. In addition to this stable absorption site, the computation reveals other low-energy positions, forming an extended network of hydrogen bonds, that could play an important role in the diffusion of H^+ in β-spinel. We analyze the dependence of structure and dynamics of the pure and protonated phases as a function of temperature and pressure.