Molecular dynamics simulations of seismic discontinuities and phase transitions of MgSiO$_3$ from 4 to 6-coordinated silicate via a novel 5-coordinated phase

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ABSTRACT

This paper reports detailed molecular dynamics simulations over a large pressure-temperature range (0–140 GPa and 300–2000 K) in the Earth’s mantle starting from the upper mantle phase enstatite MgSiO$_3$. The simulated seismic velocities show several discontinuities corresponding to the phase transitions of enstatite. With increasing pressure, enstatite MgSiO$_3$ transforms first to a new novel five-coordinated silicon phase, and then to the lower-mantle perovskite phase involving six-coordinated silicon atoms. The new intermediate phase is crystalline but orientationally disordered. The calculated seismic velocities and densities across the phase transitions for a pure MgSiO$_3$ mantle are consistent with previous estimates. These studies suggest that the major discontinuities between the upper mantle, transition zone, and the lower mantle could arise partially due to the changes in the silicon coordination.

INTRODUCTION

Magnesium silicate in various polymorphic forms constitutes a major component of the Earth’s mantle. The upper mantle, which extends to a depth of 440 km, contains olivine, pyroxene and garnet phases, whereas the lower mantle below 660 km depth is largely made of the perovskite phase (Ringwood 1975). Apparently, the most important difference between the upper and lower mantle silicates is that the silicon coordination changes from 4 to 6 respectively, whereas the Mg coordination also increases from 6 to 8. This kind of information is indirectly inferred from seismic observations and compositional modeling of the Earth’s interior based on accurate information about the structure and thermodynamic properties of the constituent phases (Dzeiwnskis and Anderson 1981; Duffy and Anderson 1989; Poirier 1991; Hemley 1998; Jeanloz 1986; Jeanloz and Thompson 1983). Accurate modeling of mantle minerals is therefore of utmost importance, and simultaneously is also a major challenge in condensed matter physics.

Several important first principles calculations (Wentzcovich et al. 1998; Teter et al. 1998; Allan and Teter 1987; Gonze et al. 1992; Keskar and Chelikowsky 1992) as well as classical molecular dynamics simulations (Chaplot and Sikka 1993; Demiralp et al. 1999; Tse and Klug 1991; Badro et al. 1996) have been performed on the various polymorphs of SiO$_2$, which may be regarded as a prototype system. First principles calculations of the elastic anisotropy and wave velocities of MgO at lower mantle conditions have been also reported (Karki et al. 1999). However, the studies on the magnesium silicate phases have been limited due to their much higher structural complexity. Some interesting first principles calculations of the structural and elastic properties have been reported on enstatite (Wentzcovitch et al. 1995) and perovskite (Wentzcovitch et al. 1993; Stixrude and Cohen 1993; Warren and Ackland 1996; Wolf and Bukowinski 1985; Karki et al. 1997). Empirical potentials have also been successfully used in simulating the thermodynamic properties of the various enstatite polymorphs (Matsui and Price 1992; Mendelssohn and Price 1997; Ghose et al. 1994; Choudhury et al. 1998) and MgSiO$_3$ perovskite (Chaplot et al. 1998; Matsui and Price 1991).

We report here MD simulations as a function of pressure at high temperature starting from an important upper mantle phase, orthoenstatite. The structure of orthoenstatite is extremely complex, having 80 atoms per unit cell and being comprised of double MgO$_6$ octahedral bands and single silicate SiO$_4$ tetrahedral chains. In the simulations at a fixed temperature of 900 K, enstatite (involving 4-coordinated silicons) transformed to a new phase involving 5-coordinated silicon at 13 GPa, which further transformed at 16 GPa to the lower mantle phase perovskite (involving 6-coordinated silicon). These simulations have several important implications. These are probably the first simulations of seismic discontinuities involving the phase transitions of enstatite over the entire pressure range in the Earth’s mantle and provide an opportunity to examine and visualize microscopically at the atomic level the key mechanisms of the transitions. Perhaps not surprisingly, even though we have chosen only one representative starting phase, the calculated discontinuities in the sound velocities and density across the simulated transitions are in very good qualitative, or even fairly quantitative agreement with the best available estimates in the mantle. This strongly reinforces the confidence in our results. Further, we predict the existence of a new five-coordinated silicate phase which may exist at pressures and temperatures characteristic of the Earth’s transition zone between the upper and the lower mantle and whose seismic properties appear to be close to those estimated for the transition zone.