Supporting Information

Kohara et al. 10.1073/pnas.1104692108

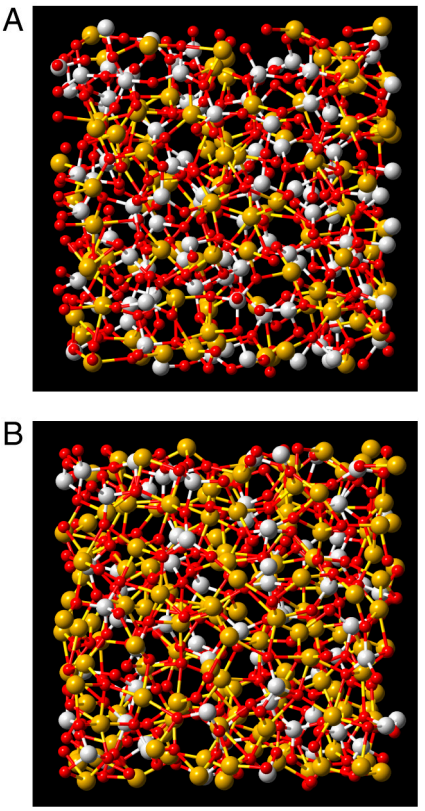

Fig. S1. Visualization of the DFT optimized structure of (*A*) $MgSiO_3$ glass (510 atoms) and (*B*) Mg_2SiO_4 glass (511 atoms). Light gray, silicon; Red, oxygen; Gold: magnesium.

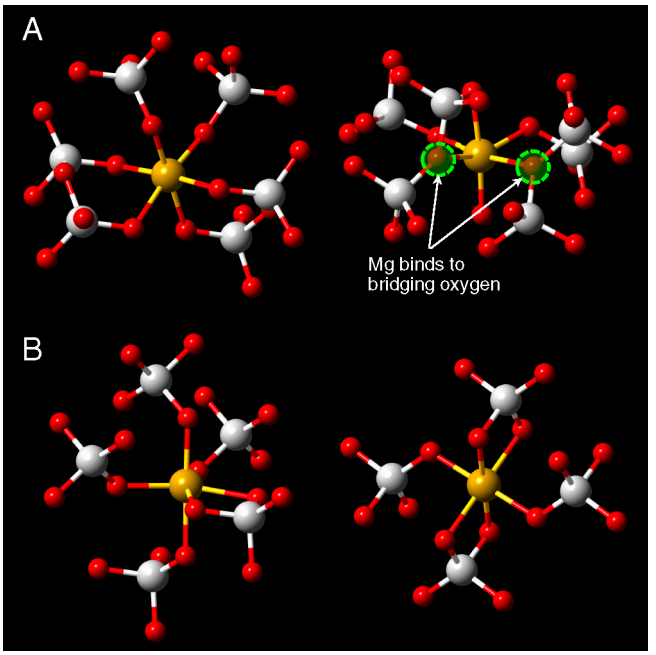

Fig. S2. Local environment around Mg cations in the crystalline phase of (*A*) $MgSiO_3$ and (*B*) Mg_2SiO_4. Both structures comprise two classes of Mg-sites. Light gray, silicon; Red, oxygen; Gold: magnesium.

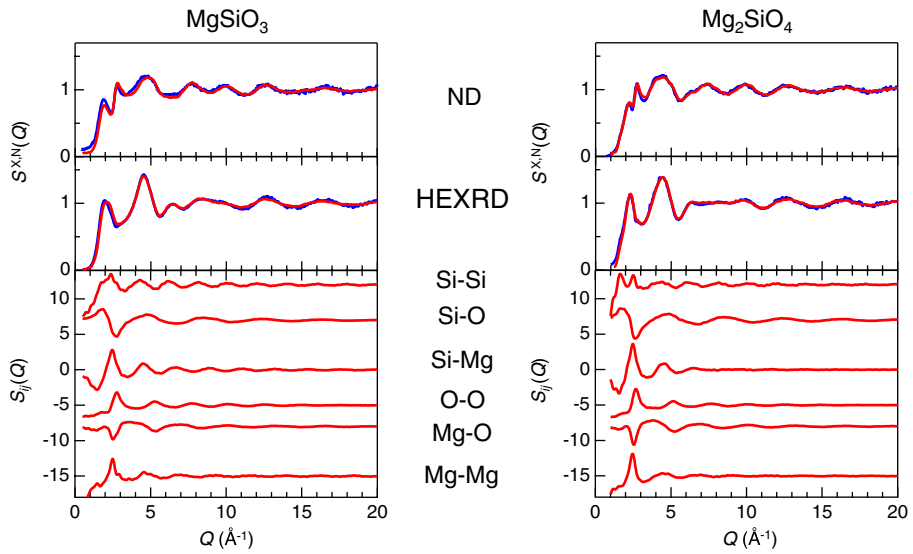

Fig. S3. Experimental X-ray and neutron total structure factors, $S(Q)$, of $MgSiO_3$ and Mg_2SiO_4 glasses together with total $S(Q)$ and partial structure factors, $S_{ij}(Q)$, obtained from RMC modeling. Blue line, Experimental data; Red line, RMC model. Experimental X-ray diffraction data was measured at high-energy X-ray diffraction beamline BL04B2 of SPring-8 using 61.6 keV X-rays.

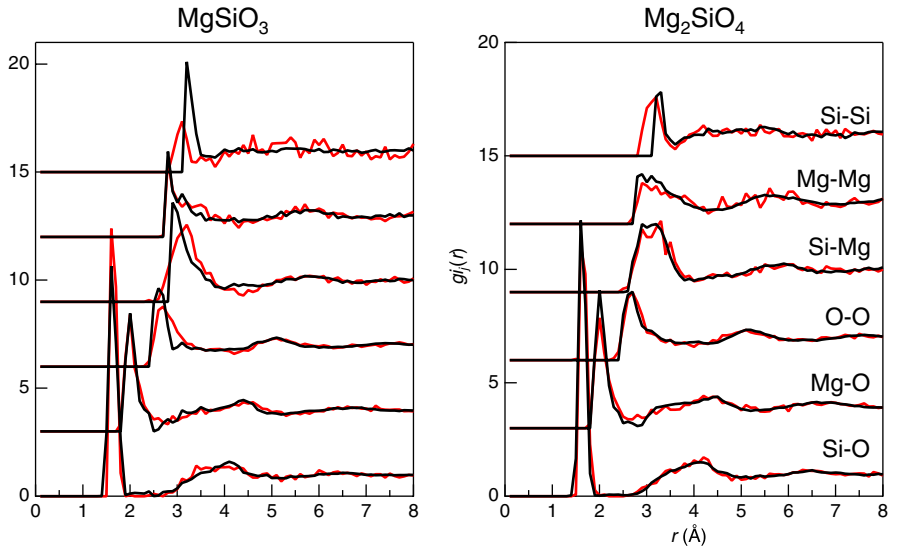

Fig. S4. Partial-pair distribution functions, $g_{ij}(r)$, of $MgSiO_3$ and Mg_2SiO_4 glasses obtained from the RMC model (back line) and the DFT model (red line).

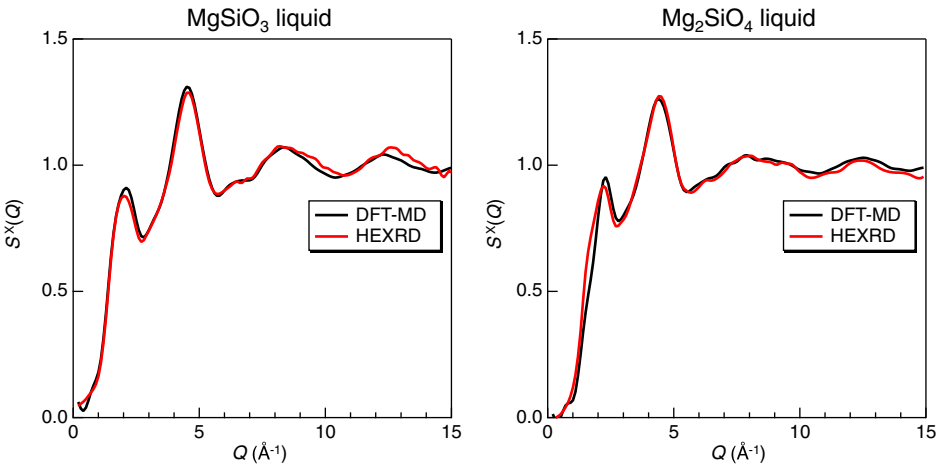

Fig. S5. X-ray structure factors, $S^x(Q)$, of $MgSiO_3$ (2,153 K) and Mg_2SiO_4 (2,223 K) liquids obtained from the high-energy X-ray diffraction measurement (red line) and DFT-MD data (black line). The DFT-MD simulations of 510/511 atoms were performed in the canonical *NVT* ensemble (thermostat, average temperatures 2,180 K and 2,230 K for $MgSiO_3$ and Mg_2SiO_4, respectively) by starting from an RMC configuration, and they lasted 30 ps in total (1 fs time step). In order to accommodate structural relaxation, only snapshots from the last 15 ps were used for the structure factor plot.

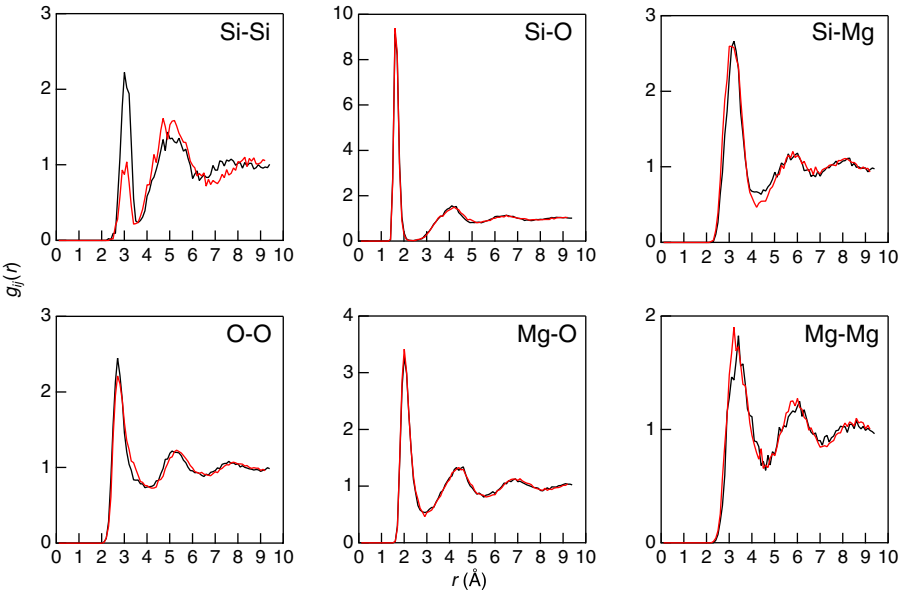

Fig. S6. Partial-pair distribution functions, $g_{ij}(r)$, of $MgSiO_3$ (black, 2,153 K) and Mg_2SiO_4 (red, 2,223 K) liquids obtained from the DFT-MD simulations. The DFT-MD data is averaged over the last 15 ps of the 30 ps MD trajectory. Differences in the Si-Si partials are due to the smaller concentration of Si and vanishing SiO_4 network in Mg_2SiO_4. The first peak of the Mg-O partial is slightly higher for Mg_2SiO_4 which explains the small difference in coordination number: $N_{Mg\text{-}O} = 4.03$ in $MgSiO_3$ liquid (up to 2.5 Å) and 4.79 (up to 2.8 Å) while $N_{Mg\text{-}O} = 4.19$ in Mg_2SiO_4 liquid (up to 2.5 Å) and 5.02 (up to 2.8 Å).