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DFT modeling of fused silica electronic structure under strong laser-induced excitation

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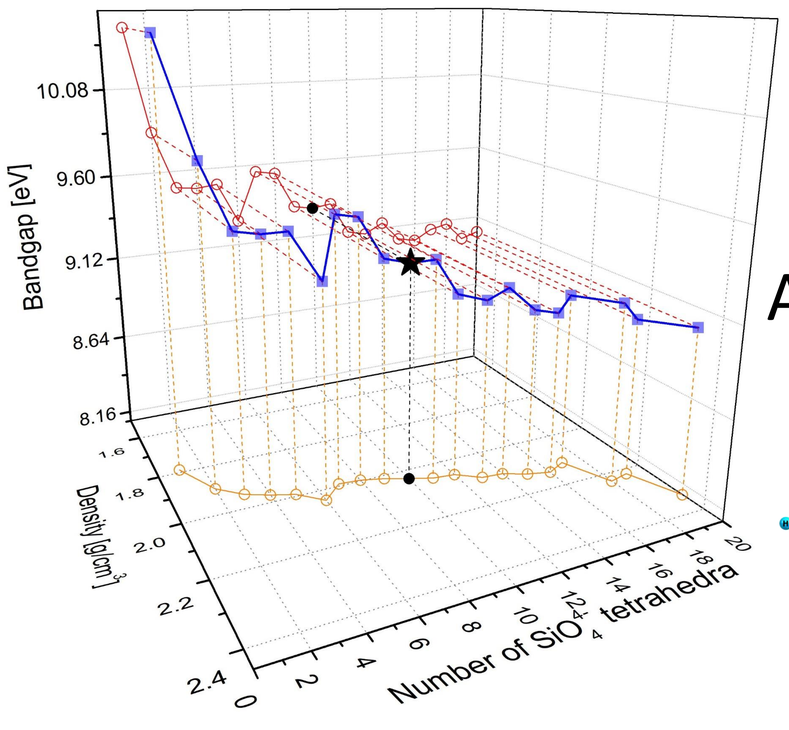
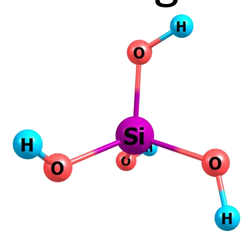
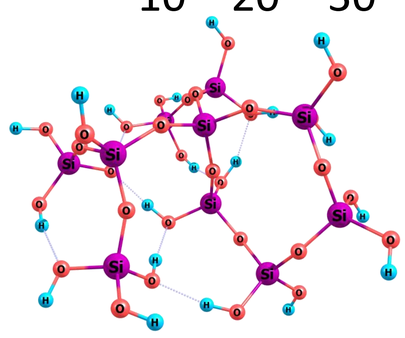
Molecular model approach



Building block

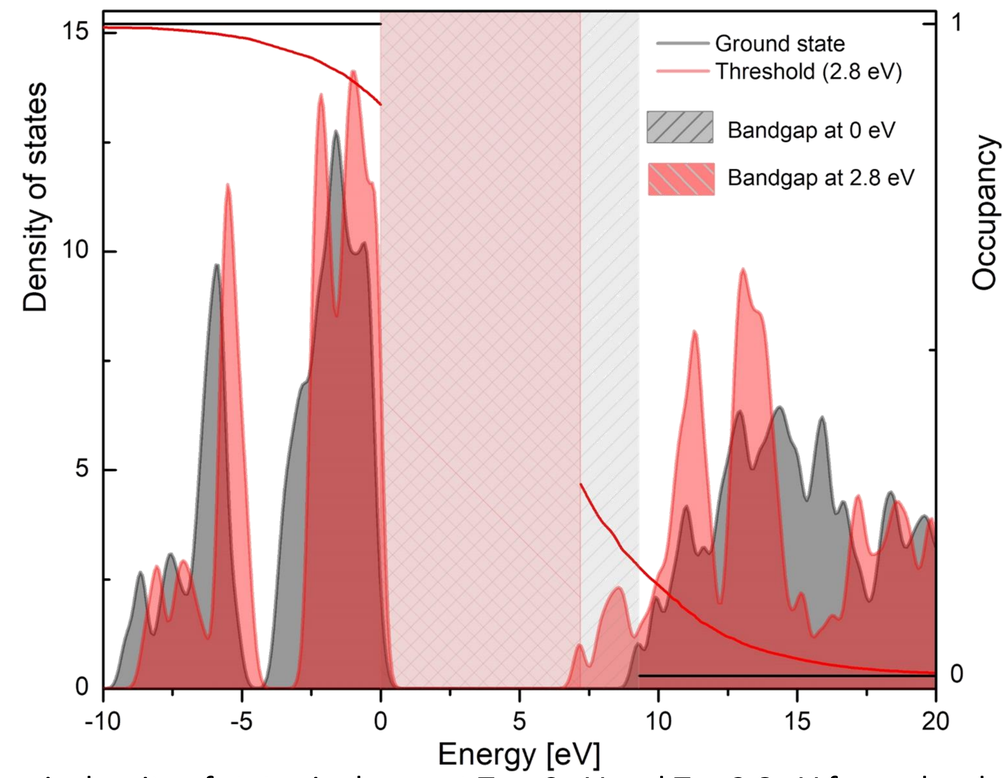


Adequate choice $Si_{10}H_{20}O_{30}$



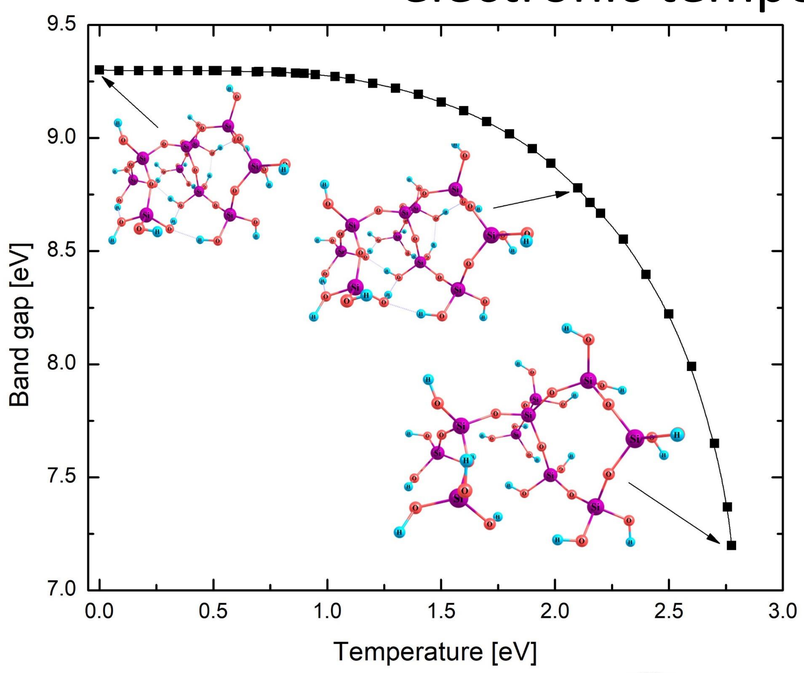
Experimental values of Bandgap is around 9 eV and Density is 2.21 g/cm^3

Electronic Density of States

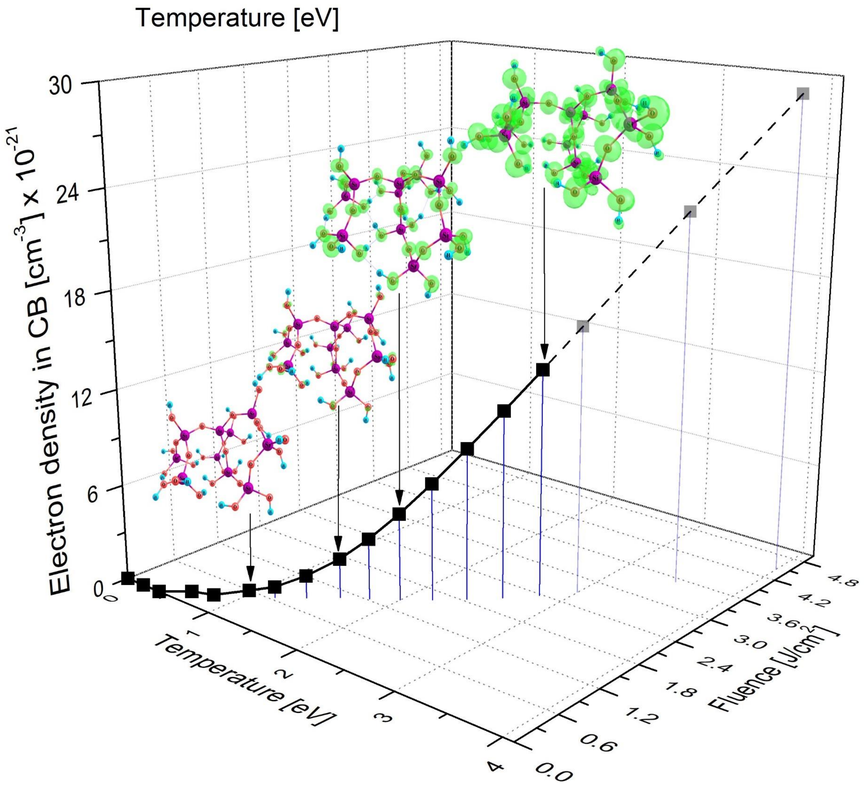


Electronic density of states is shown at T_{el} = 0 eV and T_{el} =2.8 eV for molecular system. The Fermi-Dirac distribution at different electronic temperatures (black – 0 eV, red line – 2.8 eV) The bandgap is highlighted in gray with left sparse pattern (0 eV) and red with right sparse pattern (2.8 eV). The threshold stability of the molecular system is around 2.8 eV. Above this energy, the Si-O bond-breaking starts.

Evolution of electronic structure at different electronic temperature

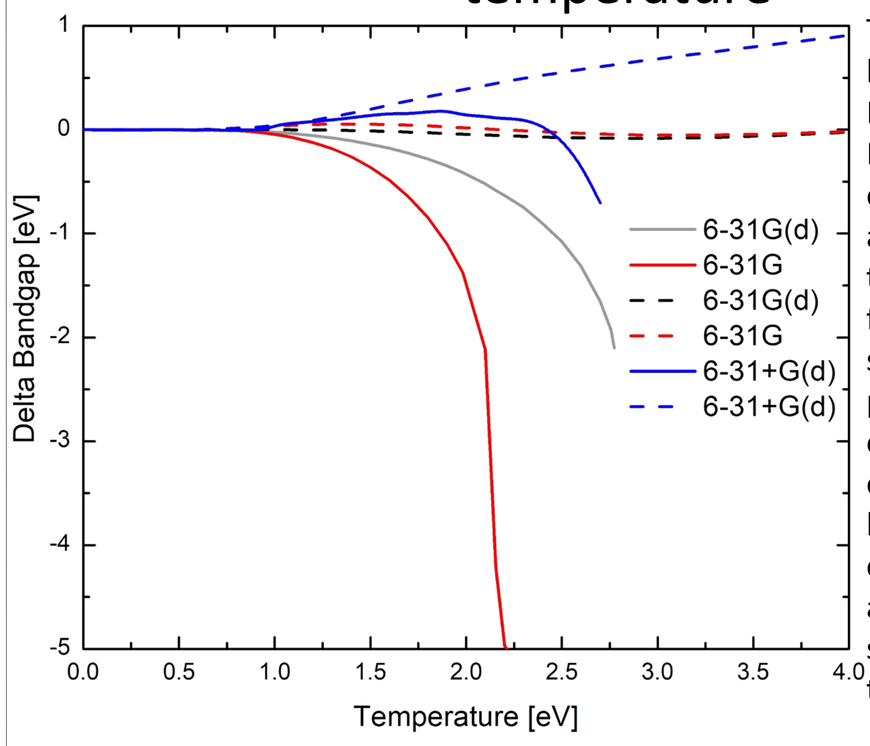


During the geometry relaxation of the fused silica at different electronic temperature which imitate the laser induced the bandgap is decreased. Below the curve, the snapshot optimized structures were shown. Root mean square displacement of the Si and O atoms from ground state reach to 0.7 Å and the volume of the system increased on 20% at threshold (2.8 eV)



Calculated electron density in the conduction band at different electronic temperatures with fluence created by 5 fs laser pulse. The pictures above the graph are the fraction occupation number weighted density (FOD) plots at different electronic temperatures (1, 1,6, 2, 2.77 eV) with contour value (σ) =0.01 eBohr^{-3} At low T_{el} the electronic density is localized over oxygen atoms. As the T_{el} rises to the threshold and higher, the electron density is localized as well as over silicon atoms.

Bandgap changes under high electronic temperature



The change of the bandgap (Delta Bandgap = Bandgap at ground state – Bandgap at defining electronic temperature) as a function of electronic temperature. The results for three different basis set were shown. Solid lines presented the result obtained after geometry optimization and dash lines - without structure optimization. The data after optimization was shown below damage thresholds.

At the first moment, the Bandgap increases by 0.5 eV. This effect changes to the opposite trend during the geometry relaxation. The general trend of Bandgap decreasing was found regardless of the choice of calculation method. The region between blue and red solid curves shows the borders of calculated decreasing of bandgap upon electron excitation. Since the displacement of the silicon and oxygen atoms is very small (less than 0.7 Å) the time-lapse of atomic movement can occur in the femtosecond region.

Conclusions

Under strong laser-induced excitation the spatial and electronic structure is changed. In the beginning, the Bandgap is increased. During the atomic relaxation, the Bandgap is decreased. The main reason for such behavior is redistribution of electron density over the atoms resulting in atomic displacement and reducing of Bandgap. The atomic displacement is very small which can occur in the tens of femtosecond range. Significant displacement of the atoms from the stationary point leads to the destruction of the system. The threshold of the stability of the fused silica is around 2.7-2.9 eV.