Minicolloque n° OPS27 Laser-driven (ultra)fast dynamics: from molecules to materials (I & II) **X Oral** **□ Poster**

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

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Fused silica is an indispensable material in emergent photonic applications due to its unique optical, mechanical, and thermal properties, especially when it is nano-structured by an ultrashort laser pulse. The precision of the laser-induced modifications relies heavily on the control of the electron excitations and transient optical properties during the laser pulse. In this work we explore the fused silica band gap at high densities of excited electrons, using Density Functional Theory (DFT).

Fused silica is a glass consisting of silica (SiO_2) in amorphous form. It includes various structural variations since the topological arrangement of the SiO_4 tetrahedra is not unique. We model fused silica as a molecule consisting of six SiO_4 tetrahedra. Fused silica geometry parameters (bond lengths and angles) and electronic structure reported experimentally are well reproduced with a reasonable computational demand. Figure 1 shows the gap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) for molecular structures with different number of SiO_4 tetrahedra. The HOMO-LUMO gap in this case represents the optical gap of fused silica measured experimentally.

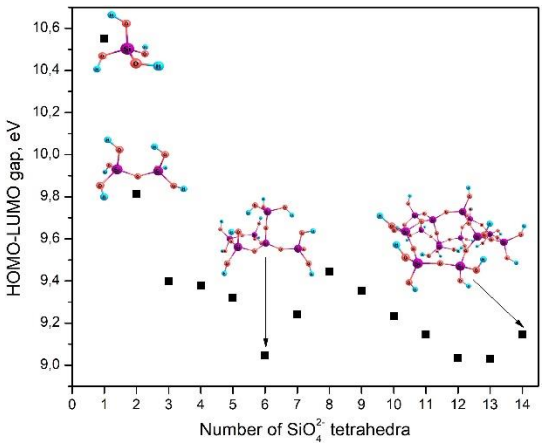

Figure 1: Energy differences between HOMO and LUMO versus the number of SiO_4^{2-} in the molecular system.

To imitate photo-electronic processes and electronic structure evolution under ultrafast laser irradiation the finite-temperature fractional occupation DFT was used. This technic introduces an electronic temperature (T_e) that applies a Fermi-Dirac-like occupation number smearing over all the orbitals of the system. An electronic temperature on the order of 1 eV leads to small atomic displacements after the geometry optimization but to strong HOMO-LUMO gap modification (increase or decrease depending on T_e). Such band-gap modification will affect nonlinearly the photoexcitation, strongly impacting the fused silica optical response under intense ultrashort laser pulse. At electronic temperature higher than 2.5 eV the system loses its thermodynamical stability and breaks down. Such temperature corresponds to the excited electron density on the order of 10^{22} cm^{-3}, comparable to the fused silica damage threshold reported experimentally.