First principles calculations of the Urbach tail in the optical absorption of silica glass

BABAK SADIGH, Lawrence Livermore National Laboratory, PAUL ERHART, Chalmers University of Technology, DANIEL ABERG, ERIC SCHWEGLER, JEFF BUDE, Lawrence Livermore National Laboratory, OPTIC DAMAGE MODELING TEAM — We present density-functional theory calculations of the optical absorption spectra of silica glass for temperatures up to 2400 K. The calculated spectra exhibit exponential tails near the fundamental absorption edge that follow the Urbach rule, in good agreement with experiments. We also discuss the accuracy of our results by comparing to hybrid exchange correlation functionals. By deriving a simple relationship between the exponential tails of the absorption coefficient and the electronic density-of-states, we establish a direct link between the photoemission and the absorption spectra near the absorption edge. This relationship is subsequently employed to determine the lower bound to the Urbach frequency regime. Most interestingly, in this frequency interval, the optical absorption is Poisson distributed with very large statistical fluctuations. Finally, we determine the upper bound to the Urbach frequency regime by identifying the frequency at which transition to Poisson distribution takes place.

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