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Bonding in alpha-quartz (SiO_2): A view of the unoccupied states

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ABSTRACT

High-resolution core-loss and low-loss spectra of α-quartz were acquired by electron energy-loss spectroscopy (EELS) with a transmission electron microscope (TEM). Spectra contain the Si L_1, $L_{2,3}$, K, and O K core-loss edges, and the surface and bulk low-loss spectra. The core-loss edges represent the atom-projected partial densities of states of the excited atoms and provide information on the unoccupied s, p, and d states as a function of energy above the edge onset. The band structure and total density of states were calculated for α-quartz using a self-consistent pseudopotential method. Projected local densities of Si and O s, p, and d states (LDOS) were calculated and compared with the EELS core-loss edges. These LDOS successfully reproduce the dominant Si and O core-loss edge shapes up to ca. 15 eV above the conduction-band onset. In addition, the calculations provide evidence for considerable charge transfer from Si to O and suggest a marked ionicity of the Si-O bond. The experimental and calculated data indicate that O 2p-Si d π-type bonding is minimal. The low-loss spectra exhibit four peaks that are assigned to transitions from maxima in the valence-band density of states to the conduction band. A band gap of 9.65 eV is measured from the low-loss spectrum. The structures of the surface low-loss spectrum are reproduced by the joint density of states derived from the band-structure calculation. This study provides a detailed description of the unoccupied DOS of α-quartz by comparing the core-loss edges and low-loss spectrum, on a relative energy scale and relating the spectral features to the atom- and angular-momentum-resolved components of a pseudopotential band-structure calculation.