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Electronic structure and excitations in oxygen deficient $CeO_{2-\delta}$ from DFT calculations C. LANE, Northeastern U., T. JARLBORG, U. of Geneva, B. BARBIELLINI, Northeastern U., YUNG JUI WANG, ALS/LBNL, R.S. MARKIEWICZ, Northeastern U., ZHI LIU, ZAHID HUSSAIN, ALS/LBNL, A. BANSIL, Northeastern U. — Mixed valent cerium oxides (ceria) are technologically important materials with remarkable properties useful for applications in heterogeneous chemical and electrochemical catalysis. We investigated the equilibrium electronic structures of supercells of $CeO_{2-\delta}$ within the Density Functional Theory (DFT), wherein properties such as lattice constants, bulk moduli and magnetic moments were well reproduced by the generalized gradient approximation (GGA) without the need to introduce the Hubbard U parameter. The chemical expansion and magnetic moment were calculated for Ce_4O_{8-N} as a function of N, which for $N = 8$, removing all the oxygen atoms, the fcc non-magnetic α-phase of Ce was recovered. In the ground state of defective ceria, the Ce-f majority band resides near the Fermi level, but appears at 2 eV below the Fermi level in photoemission spectroscopy experiments. We have demonstrated that x-ray photoelectron spectroscopy (XPS) relaxation effects yield a renormalization of f-levels away from the Fermi level for electron excitation spectroscopies, which is also consistent with Ce-M and O-K x-ray absorption spectroscopy. Work supported in part by the US Department of Energy.

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