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Strongly localized electronic screening of Mg intercalants in the Chevrel phases Mo_6S_8[1] FLORIAN THÖLE, Materials Theory, ETH Zurich, DAVID PRENDERGAST, The Molecular Foundry, Lawrence Berkeley National Laboratory — The problem of multivalent ion insertion into cathode materials is still poorly understood. Using the Chevrel phases (CPs) as a model material, we study the effect of Mg ion insertion using density functional theory (DFT). By inspection of the electron charge density difference associated with the insertion of Mg into a supercell of the material, which is metallic below the full intercalation limit, we arrive at the conclusion that the response to Mg insertion is best described in terms of a local screening cloud, effectively shielding the Mg charge with a length scale on the order of one unit cell. The density differences are localized mostly on the nearest neighbor sulfur anions with only minor differences on the nearest Mo cations. This behaviour is surprising, because in an ideal metal one might expect that insertion of an isolated ionic dopant might lead to localized screening with the Thomas-Fermi length scale $r_{TF} = 0.29$ Å, while in an insulator or semiconductor comprising transition metal cations, traditional redox chemistry might be expected. To provide a link to experiments, we simulate X-ray absorption spectra for the Mg K-edge. In full agreement with our model, the resulting spectra show an edge position which lies between metallic Mg and ionic MgO.

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