ACTINYL IONS IN $Cs_2UO_2Cl_4$ CRYSTAL

SPIRIDOULA MATSIKA, RUSSELL M. PITZER, *Department of Chemistry, The Ohio State University, 100 W.18th Avenue, Columbus, OH, 43210*; ,.

During the last years in our group we have been studying the actinyl ions, their electronic spectra, symmetric-stretch vibrational frequencies and actinide-oxygen (An-O) equilibrium distances. Most experimental results, though, exist in solid or liquid phase, so a direct comparison between our previous theoretical results and experimental results is not plausible. For this reason, we have modeled the $Cs_2UO_2Cl_4$ crystal in which both the uranyl and neptunyl spectra have been observed. The central atom, uranium or neptunium and the closest oxygens and chlorines are treated with Relativistic Effective Core Potentials(RECPs) and basis sets, the closest cesiums are treated with all electron potentials and the rest of the crystal with point charges. We performed spin-orbit Graphical Unitary Group Approach (GUGA) Configuration Interaction calculations, similar to the ones for the free ions. The An-O bond distances in the crystal increase by approximately $0.07\mathring{A}$ compared to the free ions and the symmetric stretch vibrational frequencies decrease by about 200 cm^{-1}. It seems that most of the previous differencies between theory and experiment are due to the crystalline environment rather than the insufficiency of theory.