STRUCTURE AND SPECTRA OF UO_2F_2

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Uranyl fluoride (UO_2F_2) is a product of the reaction of UF_6 with moisture. There are ca. 200,000 tons of (depleted) UF_6 stored in Ohio and ca. 500,000 tons stored in Kentucky. Thus the detection of a luminescent product of leaking UF_6 with (humid) air may be useful. Studies of solutions containing $UO_2F_n^{2-n}$ species have shown that UO_2F_2 luminescence is the most intense. Thus we have begun the study of the structure and spectra of UO_2F_2. Initially we were studying the isolated molecule, but we added solvating water molecule (probably 3), and optimized the structure to get the spectra for species in solution. Solvation effects for this neutral molecule are presumably smaller than those for ions.

We use relastivistic core potentials to replace the core electrons on all atoms, leaving 50 valence electrons to be treated explicitly. We use basis sets of polarized double-zeta size, as developed in our group. Initial structural studies used DFT (courtesy of D.A. Dixon) to give a (non-planar) C_{2v} structure. MOs are obtained from SCF and MCSCF calculations. The spin-orbit interaction and electron correlation are included using spin-orbit configuration interaction (SO-GUGA in the Columbus programs). Spin-orbit and equatorial-ligand (F^-, H_2O) interactions compete in determining the splittings of the known ($^3\Delta_g$) luminescent state of the uranyl ion.