SPECTROSCOPIC INVESTIGATION OF URANYL CHLORIDE ISOLATED IN SOLID Ar

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Laser-induced fluorescence and infra-red absorption spectra have been recorded for matrix-isolated uranyl chloride (UO_2Cl_2). Pulsed laser excitation was examined using a XeCl Excimer laser (308 nm) and a dye laser operating in the 365-510 nm range. Several absorption and emission band systems were observed. The emission spectra were dominated by a nearly harmonic vibrational progression with a frequency of 843 cm^{-1} starting at 20310 cm^{-1}. The electronic absorption spectra also were dominated by a nearly harmonic vibrational progression with a frequency of 710 cm^{-1} starting at 20358 cm^{-1}. The absorption spectra consist of two series of bands. One is weak and sharp which is attributed to the charge transfer transitions from the lowest lying orbital to U 5f. The other is more intense and broad, and has been assigned to transitions to U 6d orbitals. The fluorescence decay of the emitting state was bi-exponential, with characteristic decay lifetimes of 50 and 260 μs.

Past studies have yielded optical spectra of crystalline uranyl chloride and uranyl chloride complexes in acetone and other organic solvents. Data for the matrix isolated molecule indicate that the spectrum is minimally perturbed by the local environment for a diverse range of condensed phase compositions. Assignment of the electronic transitions of UO_2Cl_2 is discussed in terms of the electronic structure of the ${UO_2}^{2+}$ sub-unit.