This talk will discuss the jet-cooled vibronic spectroscopy of 4-isocyanobenzonitrile (4IBN, CN-C₆H₄-CN) via resonant two-photon ionization, laser induced fluorescence, dispersed fluorescence, and UV-UV hole burning methods. Much of the vibronic spectroscopy of the molecule shows strong similarities with the symmetric structural isomers dicyano and di-isocyanobenzene. However, in 4IBN, we have identified a series of weak transitions extending well red of the ππ* origin. UV holeburning spectroscopy has been used to confirm that these transitions arise from the same ground state as the others, pointing to the presence of another electronic state. Based on comparison with other nitrilebenzenes, we postulate that the large dipole moment of 4IBN opens the possibility of a charge transfer (CT) state, much as occurs in para-amino substituted benzonitriles. The rich vibronic spectroscopy of this molecule will be explored using dispersed fluorescence.