



Fig. S1: Overall view of the ν_9 band of isoquinoline: in black, the SPIRALES spectrum; in red, the ν_9 band simulated at $T_{\text{rot}} = 35(5)$ K using a Gaussian line shape of 0.0017 cm^{-1} FWHM. Two insets display in the R-branch a good agreement between (obs-calc) ν_9 spectra, while the observed structured Q-branch split is not reproduced by the simulation.