DIRECT STATE-SELECTIVE MEASUREMENT OF THE COOLING RATES OF VIBRATIONALLY EXCITED $1^3\Sigma_g^+$ Na_2 ON THE SURFACE OF HELIUM CLUSTERS

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Optical excitation of the $(v' = 10)$ $1^3\Sigma_g^+ \leftarrow 1^3\Sigma_u^+$ transition of Na_2 formed on a He cluster surface yields dispersed emission which can only be modeled by including significant contributions from all $(v' = 0 - 10)$ lower vibrational levels of the excited electronic state. The process by which this vibrational energy is transferred from the excited dimer to the He nanodroplet has been characterized using state-selective time-correlated single photon counting. We have measured the onset of fluorescence arising from lower v' levels upon excitation of various higher vibrational states and find that in all cases fluorescence begins in less than 250 ps. Along with the non-exponential nature of the distribution of level popuolations, this leads us to conclude that the vibrational energy is not transferred in a cascading process but rather by means of multiquanta jumps. Following the vibrational deexcitation, the dimer emits in the gas phase, having desorbed from the cluster surface.