

CLUSTER-SIZE EFFECT IN THE ROTATIONAL RELAXATION OF HCCCN EMBEDDED IN ^4He CLUSTERS. A DOUBLE RESONANCE EXPERIMENT

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We have measured MW-IR double-resonance spectra for HCCCN molecules embedded in ^4He clusters. Microwave radiation at ≈ 9 GHz is used to pump the $(v = 0, J = 3) \leftarrow (v = 0, J = 2)$ rotational transition while a color center laser at $\approx 3300 \text{ cm}^{-1}$ is scanned through the ν_1 band. The observed patterns provide information on the relaxation of rotational energy inside the cluster: at large ($N > 5000$) cluster sizes, rotational relaxation appears to follow a “strong collision” model in which memory of the initial J state is lost after a relaxation event; for smaller cluster sizes, the rotational relaxation is shown to be affected by the presence of resonance phenomena, which are likely due to the energy level structure of the host cluster. In particular the possible role of surface excitations (riplons) will be discussed.