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Preparation of a single highly vibrationally excited quantum state using Stark induced adiabatic Raman passage[1] WILLIAM PERREAULT, NANDINI MUKHERJEE, RICHARD ZARE, Stanford Univ — Stark induced adiabatic Raman passage (SARP) allows us to prepare an appreciable concentration of isolated molecules in a specific highly excited vibrational level. As a demonstration, we transfer nearly 100% of the HD (v=0, J=0) in a supersonically expanded molecular beam of HD molecules to HD (v=4, J=0). This is achieved with a sequence of partially overlapping nanosecond pump (355 nm) and Stokes (680 nm) single-mode laser pulses of unequal intensities. The experimental spectral broadening with pump to Stokes delay and saturation against Stokes power suggest that complete population transfer has been achieved from the initial HD (v=0, J=0) to the target (v=4, J=0). By comparing our experimental data with our theoretical calculations we are able to draw two important conclusions: (1) using SARP a large population ($>10^{10}$ molecules per laser pulse) is prepared in the (v=4, J=0) level of HD, and (2) the polarizability $\alpha_{00,40}$ (0.6 x 10^{-41} Cm^2V^{-1}) for the (v=0, J=0) to (v=4, J=0) Raman overtone transition is only about five times smaller than $\alpha_{00,10}$ for the (v=0, J=0) to (v=1, J=0) fundamental Raman transition. This capability of preparing selected, highly excited vibrational quantum states of molecules opens new opportunities for fundamental scattering experiments.

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William Perreault
Stanford Univ

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