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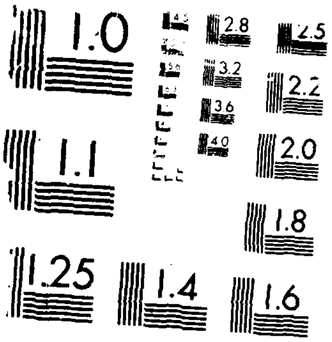

PY RESOLUTION TEST CHART

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Two dye laser systems were purchased for use in energy transfer studies and in unimolecular decomposition experiments. Individual angular momentum states of acetylene in the fourth vibrational state of the CH stretching mode were prepared. It was found that these molecules which contain 10,000 wave numbers of vibrational energy relax at several times the gas kinetic collision rate. There is a striking dependence on the initial rotational state excited implying relaxation of only relatively small amounts of rotational energy.

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Final Report

Grant No. AFOSR-86-0244

(15 July 1986 to 14 July 1987)

This grant is part of the University Research Instrumentation Program (URIP) that allowed us to advance our DoD sponsored research very significantly by purchasing lasers for use in our collisional energy transfer studies and in our unimolecular decomposition experiments. These benefits have come not only from the use of the hardware on our AFOSR sponsored projects but also by reducing competition for equipment by other projects. The net result has been some dramatic new collisional energy transfer results and spectroscopic measurements in our AFOSR sponsored project. In other DoD sponsored research, we have obtained new data in two photodissociation experiments and a new vibrational overtone spectroscopic measurement.

The primary goal of our URIP proposal was to advance our studies of collisional energy transfer in highly vibrationally excited polyatomic molecules. We have now succeeded in preparing individual angular momentum states of acetylene (C_2H_2) in the region of the third overtone of the CH stretching transition $3\nu_{CH}$ and monitoring the resulting highly vibrationally excited molecule using ultraviolet laser induced fluoresence. The experiments have three primary aspects: they explore the nature of the highly vibrationally excited state produced by vibrational overtone excitation, they exploit the two-photon combination of vibrational excitation and laser induced fluoresence to excite directly

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previously unobservable vibrations in the electronically excited molecule, and they follow the collisional relaxation of individual quantum states in highly vibrationally excited molecules. The collisional energy transfer studies have shown that these molecules that contain 10 000 cm^{-1} of vibrational energy relax at several times the gas kinetic collision rate. We note a striking dependence in the probe excitation spectra on the initial rotational state excited and have tentatitively concluded that the relaxation proceeds by extremely rapid vibrational energy transfer with only relatively small amounts of rotational relaxation. We have carefully determined the rate of loss of the excited molecules and are now identifying the final states of the relaxation. We expect to sort out the mechanism of the collisional energy transfer and extend the work to other vibrational levels and molecules.

The other DoD sponsored research that has benefitted from the increased experimental time available because of the URIP hardware is a unimolecular decomposition experiment funded by the Army Research Office. We have now studied the vibrationally mediated photodissociation of $HONO_2$ and made the first state resolved measurements on its vibrational overtone induced decomposition. In the vibrationally mediated decomposition experiments, one photon excites an overtone vibration and another fragments the resulting vibrationally excited molecule by exciting it to a repulsive electronic surface. This approach has revealed unique photodissociation dynamics, as seen in the quantum state distribution of the products. We have also made the first state resolved unimolecular decomposition measurements on this molecule by exciting the fourth ($5\nu_{OH}$) or fifth ($6\nu_{OH}$) OH stretching overtone vibration and probing the individual quantum states of the OH fragment.

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