ROTATIONALLY-RESOLVED SCATTERING OF FORMALDEHYDE FROM THE Au(111) SURFACE: AN AXIS SPECIFIC ROTATIONAL RAINBOW AND ITS ROLE IN TRAPPING PROBABILITY

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The conversion of translational to rotational motion often plays a major role in the trapping of small molecules at surfaces, a crucial first step for a wide variety of chemical processes that occur at gas-surface interfaces. However, to date most quantum-state resolved surface scattering experiments have been performed on diatomic molecules, and very little detailed information is available about how the structure of non-linear polyatomic molecules influences the mechanisms for energy exchange with surfaces. In the current work, we employ a new rotationally-resolved 1 + 1′ resonance-enhanced multiphoton ionization (REMPI) scheme to measure rotational distribution in formaldehyde molecules directly scattered from the Au(111) surface at incident kinetic energies in the range 0.3–1.2 eV. The results indicate a pronounced propensity to excite $a$-axis rotation (twirling) rather than $b$- or $c$-axis rotation (tumbling or cartwheeling), and are consistent with a rotational rainbow scattering model. Classical trajectory calculations suggest that the effect arises—to zeroth order—from the three-dimensional shape of the molecule (steric effects). The results have broad implications for the enhanced trapping probability of prolate and near-prolate molecules at surfaces.