Non-adiabatic dissociation of rovibrationally excited acetylene
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Abstract
Two- and three-color vibrationally mediated photodissociation/probing was applied to explore the dissociation dynamics of acetylene, \( \text{C}_2\text{H}_2 \), from the bound portion of the \( \tilde{A} \rightarrow \tilde{A} \) trans-bent excited electronic state.

Introduction
Photodissociation of vibrationally excited molecules is a well known method for manipulating and exploring bond cleavage. Deposition of energy into skeletal motions strongly coupled to the reaction coordinate leads in preferred cases to control of photodissociation pathways and photoproducts. Acetylene serves as an important prototype as it bridges the gap between diatomic and large molecules.

Method
Direct near infrared (IR) excitation efficiently prepared rovibrational levels in the region of three \( \text{C}-\text{H} \) stretch quanta ( \(-9640 \text{ cm}^{-1}\) ) and UV photons in the \(-243 \text{ nm} \) region promoted the pre-excited \( \text{C}_2\text{H}_2 \) molecules to the \( \tilde{A} \rightarrow \tilde{A} \) state and dissociated them. UV photons from the same laser or from an independent UV laser subsequently ionized the H photofragments via resonantly enhanced multiphoton ionization (REMPI). Tagging of the H photofragment yield vs the IR or UV laser wavelength resulted in IR or UV dependent action spectra, representing rovibrational excitation and rovibrionic absorption spectra, respectively. Photoacoustic absorption spectra were monitored simultaneously for calibration.

Results
- IR photoacoustic absorption spectra
- UV action spectra (0030°) and (1112°)


depiction of data and analysis

Conclusions
The main factor driving the dissociation is the initial state preparation and the subsequent excitation to the bound portion of the \( \tilde{A} \rightarrow \tilde{A} \) state. In the latter, numerous vibrionic states involving torsion and antisymmetric cis-in plane bend modes are accessed affecting the dissociation efficiency.

References