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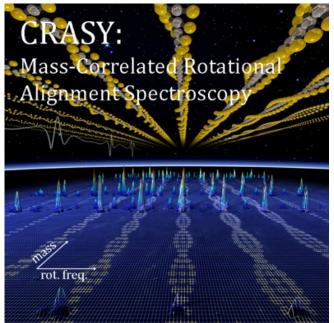
## Structure and photochemistry of biomolecules

We use femtosecond time-resolved spectroscopy to observe photochemical processes in isolated biomolecules, predominantly building blocks of DNA. Molecular clusters reproduce relevant structural features, such as base-pairing and base-stacking and reveal how a local environment affects molecular properties. Biologically relevant systems often contain multiple structural isomers or even

unknown molecular structures. Femtosecond pump-probe experiments rarely offer sufficient spectroscopic information to assign structures and are therefore insufficient to characterize such systems. With correlated rotational alignment spectroscopy, we overcome this limit by high-resolution correlating rotational spectra with femtosecond pump-probe data. This technique will open the door for the isomer-selective investigation of electronic structure and dynamics, for the characterization of impure samples, and for the unambiguous characterization of small molecular clusters.

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The coherent molecular motion of a rotational wavepacket (top) is observed by mass-spectrometry and allows the correlation of molecular mass and rotational frequency (bottom).