

## MILLIMETER–WAVE–MILLIMETER–WAVE DOUBLE RESONANCE SPECTROSCOPY

OLIVER ZINGSHEIM, LUIS BONAHA, HOLGER S. P. MÜLLER, FRANK LEWEN, SVEN THORWIRTH,  
STEPHAN SCHLEMMER, *I. Physikalisches Institut, Universität zu Köln, Köln, Germany.*

Complex organic molecules show regularly very rich spectra, sometimes at a line density close to the confusion limit. The plethora of lines often originates from the presence of different conformers and/or low-lying vibrational states. The analysis of spectra may considerably be further complicated by vibration-rotation or other interactions. However, accurate spectroscopic predictions in the millimeter-wave region are essential for identifying molecules in space. Double resonance techniques can help to solve these challenges.

We present first millimeter-wave–millimeter-wave double resonance (DR) spectra to unambiguously assign new pure rotational lines of propanal ( $\text{C}_2\text{H}_5\text{CHO}$ ). As already shown in chirped pulse Fourier transform microwave (CP-FTMW) experiments<sup>a</sup>, the Autler-Townes splitting<sup>b</sup> allows for distinguishing between regressive or progressive energy level schemes. The Autler-Townes splitting is clearly visible in our 2D spectra. Furthermore, implementation of a double modulation technique (pulse modulation of pump and frequency modulation of probe source) allows for confusion- and baseline-free spectra containing only the line(s) of interest. We discuss details of the observed Autler-Townes splitting and possible future applications, such as automatization and incarnation of baseline-free DR spectroscopy in chirped pulse experiments.

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<sup>a</sup>D. Schmitz et al., *J. Phys. Chem. Lett.* **6** (2015) 1493

<sup>b</sup>S. H. Autler and C. H. Townes, *Phys. Rev.* **100** (1955) 703