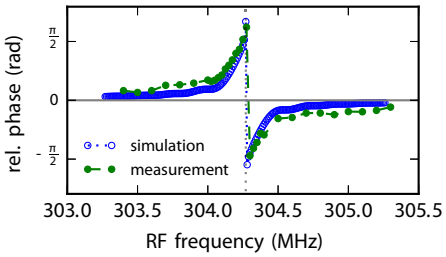
ON THE PHASE DEPENDENCE OF DOUBLE-RESONANCE EXPERIMENTS IN ROTATIONAL SPECTROSCOPY

DAVID SCHMITZ, V. ALVIN SHUBERT, ANNA KRIN, *CoCoMol, Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg, Germany*; DAVID PATTERSON, *Department of Physics, Harvard University, Cambridge, MA, USA*; MELANIE SCHNELL, *CoCoMol, Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg, Germany*.

We report double-resonance experiments using broadband chirped-pulse Fourier transform microwave spectroscopy that facilitate spectral assignment and yield information about weak transitions with high resolution and sensitivity. Using the diastereomers menthone and isomenthone as examples, we investigate both the amplitude and the phase dependence of the free-induction decay of the microwave signal transition from pumping a radio frequency transition sharing a common level.



We observe a strong phase change when scanning the radio frequency through molecular resonance. The direction of the phase change depends on the energy level arrangement, i.e., if it is progressive or regressive. The experimental results can be simulated using the density-matrix formalism using the three-level Bloch equations and are best described with the AC Stark effect within the dressed-state picture, resulting in an Autler-Townes splitting. The characteristic phase inversion allows for a) the precise frequency determination of the typically weak radio frequency transitions exploiting the high sensitivity of the connected strong microwave signal transition and b) definitive information about the connectivity of the energy levels involved, i.e., progressive vs. regressive arrangements.