Coherent superposition of two rotational quantum states in the time domain: Microwave spectroscopy without microwaves

T. Laarmann
Photon Science, Deutsches Elektronen Synchrotron (DESY), Germany

Femtosecond laser pulses in the near-infrared spectral range were used to excite a coherent superposition of two rotational quantum states of cold carbon monoxide in a non-resonant Raman process. The associated nuclear motion is followed in time by subsequent Coulomb explosion with soft X-ray FLASH pulses at high intensity in a pump-probe scheme. The coupling of J = 0 and J = 2 states results in an asymmetry of spatial fragmentation patterns detected parallel to the laser polarization axis. The observed wave packed oscillation prevails for at least 1 nanosecond covering more than 150 recurrences without dephasing. This observation can serve as a new route to disentangle complex rotational couplings and ultrafast decoherence phenomena that occur in complex systems and environments such as doped helium droplets in real time with a spectral and temporal resolution limited by the length of the delay scan and the femtosecond pulse duration, respectively.