An ultrafast algorithm for ultrafast time-resolved coherent Raman spectroscopy

Dirk van den Bekerom[1], *Francesco Mazza*[2]

1. *Unit Energy & Materials Transition, Netherlands Organisation for Applied Scientific Research TNO, Urmonderbaan 22, Geleen, Netherlands*
2. *Chair of Optical Diagnostics in Energy, Process and Chemical Engineering, RWTH Aachen University, Schinkelstrasse 8, 52062 Aachen, Germany*

Coherent Raman Spectroscopy (CRS) is a state-of-the-art non-linear optical tool for chemically reacting flows that allows for state-resolved measurements of coherence- and energy-transfer in molecular collisions. Nonetheless, quantitative CRS has thus far been limited to a surprisingly small number of simple (mostly diatomic) molecules, because synthesizing the spectra of complex polyatomic molecules, possibly including tens of billions of Raman transitions, has remained an unsolved bottleneck. In this contribution we introduce a novel algorithm for quantitative ultrafast coherent Raman spectroscopy (CRS) in gas-phase media, based on the mathematical framework of the integral transform formulation [1]. The algorithm can be applied both in the frequency- and time-domain, and is up to a million times faster than conventional methods (figure 1 – left), with the approximation error in the frequency-domain being less than 0.1%. This spectral synthesis algorithm is fast enough to allow for on-the-fly evaluation of the CRS signal, with a clear potential for the commercialization of turn-key CRS spectrometers, capable of live monitoring gas-phase chemical reactions (figure 1 – right). Moreover, the rapid speed-up for large numbers of spectral lines (>1M) opens the door for quantitative time-resolved CRS on complex polyatomic molecules, enabling unexplored research avenues in the field of gas-phase physical chemistry.

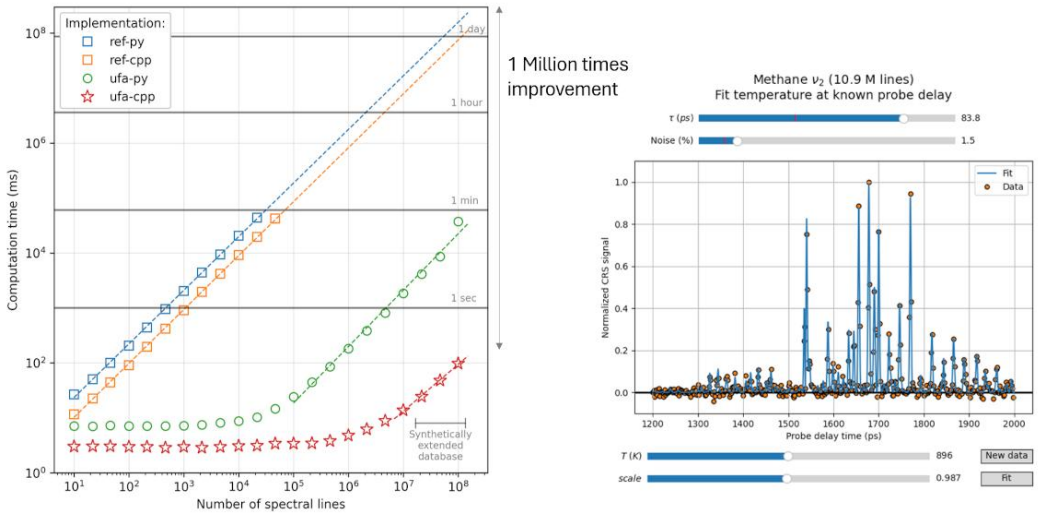

Figure 1. (left) benchmark of the new algorithm demonstrates a million times improvement in performance compared to state of the art. (right) the ultrafast algorithm allows for

1. van den Bekerom and Pannier, J. Quant. Spectrosc. Radiat. Transf. 261, 107476 (2021)