Single laser source diagnostics by space-time femtosecond coherent Raman imaging

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Conventional femtosecond non-linear spectroscopy primarily focuses on temporal phase coherence through time- or frequency-resolved methods. In this work, we introduce an alternative experimental framework based on spatial phase coherence and report the first direct observation of its effects in femtosecond rotational coherent Raman scattering (fs-RCRS) in the gas phase. We demonstrate that the third-order susceptibility exhibits periodic spatial modulation across the transverse plane, leading to non-uniformities at each rotational rephasing time. This discovery complements the standard approach to femtosecond spectroscopy and opens new avenues for single-shot diagnostics, spatio-temporal imaging, and improved accuracy in coherent Raman spectroscopic methods.

The concept of spatial phase coherence is illustrated in Figure 1. A time trace, measured using a folded BOXCARS arrangement and recorded from a single pixel on the CCD, along with its theoretical reconstruction, is shown in Figure 1(a). Figure 1(b) presents a two-dimensional image of the signal at a probe delay of 4.19 ps. Given that the transverse overlap of the laser fields at the focal point is approximately 150 µm, the probe inherently covers a temporal interval of about 500 fs, capturing the rotational dynamics encoded in the excess spatial phase $[\omega t - (k_x x + k_y y)]$. This phase correlation maps the temporal evolution as a function of x and y, producing the single-shot image in Figure 1(b), which effectively reflects the temporal dynamics highlighted in Figure 1(a). Figure 1(c) shows the theoretical prediction of Figure 1(b). The interplay between spatial and temporal phase coherence causes the signal to exhibit an apparent diagonal movement from the top left to the bottom right as the probe delay increases. The time trace extracted from the black dot in Figure 1(b) is plotted in Figure 1(a).
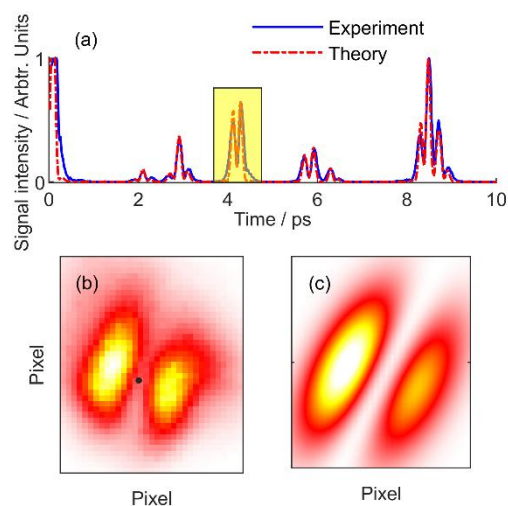

FIG 1. (a) Experimental time trace of fs-RCRS signal recorded in air at room temperature and atmospheric pressure (blue) and simulation (red dashed) from the pixel marked in (b). (b) Spatial signal at a 4.19 ps delay. The horizontal axis (33 pixels) corresponds to the time window highlighted in (a), with pixels before/after the center representing earlier/later times. (c) Simulated image of (b).