Quantum Multidimensional Spectroscopies with X-Ray Light, Entangled Photons, and in Optical Cavities

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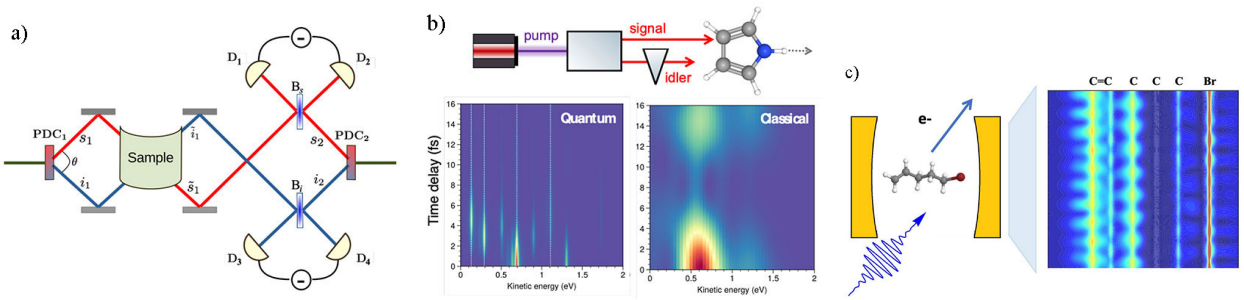

Figure 1: (a) Setup of the nonlinear interferometer with the two nonlinear crystals PDC1/2, beamsplitters Bs/i and detectors D1-4. (b) Entangled photoelectron signal. (upper) Schematic of the setup and a comparison of the photoelectron signal with entangled and classical light fort the photodissociation of pyrrole. (c) Schematic of the setup of manipulating molecular charge migration in an optical cavity

We present several novel femtosecond spectroscopic techniques that make use of the quantum nature of light and femtosecond X-ray pulses.

The nonlinear interferometer setup of Fig. 1(a) can isolate exciton-scattering processes in the photosyetem II reaction center.

The spectro-temporal correlation of time-energy entangled photon pairs was employed to overcome the Fourier limit of the temporal and spectral resolutions in conventional photoelectron signals of the photodissociation dynamics of pyrrole using the nonadiabatic wave packet simulations shown in Fig 1(b). The spectral resolution is achieved by electron detection and temporal resolution by a variable phase delay.

The ultrafast electronic charge dynamics in molecules upon photoionization while the nuclear motions are frozen is known as charge migration. Quantum dynamics simulations of photoionized 5- Bromo-1-pentene shown in Fig. 1(c) and Fig. 2 reveal that the charge migration process can be enhanced by placing the molecule in an optical cavity and monitored by time-resolved photoelectron spectroscopy. Figure 2 shows the time-dependent polariton charge density differences (from the bare molecule) and the corresponding time-resolved photoelectron signals. The collective nature of the polaritonic charge migration process is examined. We find that molecular charge dynamics in a cavity is local and does not show many- molecule cooperativity. The same conclusion should apply to cavity polaritonic chemistry.

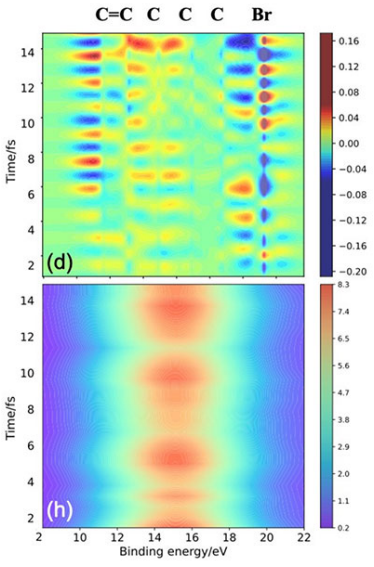

Figure 2:

References

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