Quantum Control Spectroscopy: Nonlinear (micro-) spectroscopy with tailored pulses

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Abstract—We demonstrate the use of shaped 10 fs pulses for multimodal microscopy. The combination of a broadband oscillator and a pulse shaper provides a flexible light source that can be optimized for various nonlinear effects produced in the sample either for signal intensity or selectivity. Switching between narrowband and broadband Multiplex CARS, elimination of unwanted two-photon fluorescence or direct determination of the linear Raman spectrum can be easily achieved.

Keywords—Nonlinear optical microscopy; coherent control; coherent anti-Stokes Raman; ultrafast optics; pulse shaping.

I. SUMMARY

Optical non-linear effects are of great interest for microscopy of soft matter in biology and medical research as well as of new materials in applied sciences. Second harmonic generation (SHG), two-photon fluorescence (2PEF) and coherent anti-Stokes Raman scattering (CARS) are examples that show well known advantages like intrinsic sectioning capability and information about structure as well as chemical composition at the micrometer scale without any labelling. These signals may even be detected simultaneously, combining their specificities in multimodal microscopic imaging. However, often different contributions, especially 2PEF and CARS, spectrally overlap which makes the interpretation of each of them difficult. Furthermore, high photodamage with ultrashort pulses and the complexity of spectra in biological tissue might limit the outcome in useful information.

In this contribution, we present the application of quantum control spectroscopy to circumvent these problems by using only one ultra broadband laser pulse of sub 10fs which is shaped by means of a dispersion-free 4f-line and a spatial light modulator in its Fourier plane. The shaped pulse can be designed such that a selected nonlinear process is specifically excited and probed, while minimizing unwanted contributions [1,2]. Not only coherent Raman spectra can be measured which were covered by two-photon fluorescence [3] but also the original linear Raman response can be directly obtained [4,5]. Key in this process is the application of double quadrature spectral interferometry (DQSI) [6], which requires measurements of four different phases of the gate and is based on the sinusoidal dependence of the measured signal on the phase of the gate. Once the different schemes have been implemented and explored multimodal microscopy can be easily performed just by changing the phase on the shaper and probing the different nonlinear contrast mechanism, like 2PEF, SHG, broadband Multiplex CARS [4], narrowband single frequency CARS or even linear Raman [5].

Fig. 1. (top) Single-beam-CARS setup combining an ultrabroadband sub-10 femtosecond laser, a pulse shaper and a microscope; (bottom) two different phase functions for multiplex CARS (left) and narrowband CARS (right).

REFERENCES