

THE ULTRAFAST SPECTROSCOPY LABORATORY PICOSECOND OPA SYSTEM

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INTRODUCTION

Many ultrafast time-resolved techniques require two broadly and independently tunable, synchronised light pulses. Recent advances in short pulse laser technology and non-linear crystal materials such as BBO have encouraged the rapid development of optical parametric oscillators (OPOs) and traveling wave optical parametric amplifiers (OPAs) capable of meeting the needs of ultrafast spectroscopists. We have developed kilohertz repetition rate OPA systems providing a stable, high repetition rate, high spectral brightness laser source tunable through the UV and visible wavelengths¹⁾. The system is optimised for femtosecond/picosecond, pump and probe time-resolved absorption, reflectance and resonance Raman spectroscopies.

DESCRIPTION OF THE SYSTEM

The OPA system is based on two type I BBO (5mm, 27°) crystals which are pumped by a single Spectra-Physics titanium sapphire regenerative amplifier system operating at 1 kHz repetition rate around 800 nm, 600-800µJ pulse energy and ~1 ps pulse width (figure 1)²⁾. Figure 2 is a

more detailed diagram of the narrow linewidth OPA. A three stage pump arrangement is used in each OPA. The first stage generates seed light by optical parametric generation, OPG, ('superfluorescence' generation). A 60 cm lens, f1, focuses a ~20 µJ split of 400 nm pump light into the BBO crystal. Tilting this lens creates an aberrated focus elongated in the direction of angle walk-off between pump signal and idler within the crystal. Since the total gain is a stronger function of effective crystal length than of pump intensity this enhances the OPG process. For the second stage the same pump beam and the OPG light are retro-reflected back to the crystal. The pump beam is re-focused into the crystal using lens f2. Group velocity mismatch accrued in the OPG stage is compensated by a 'magic' mirror. The output from this stage is a beam with several 10's of nJ of energy and provides good quality seed light for the final amplifier stage. Both OPAs use this configuration but OPA 1 uses a grating dispersion element, G, to spectrally filter the pulse prior to injection into the final

amplification stage. This filter also reconstitutes the beam with

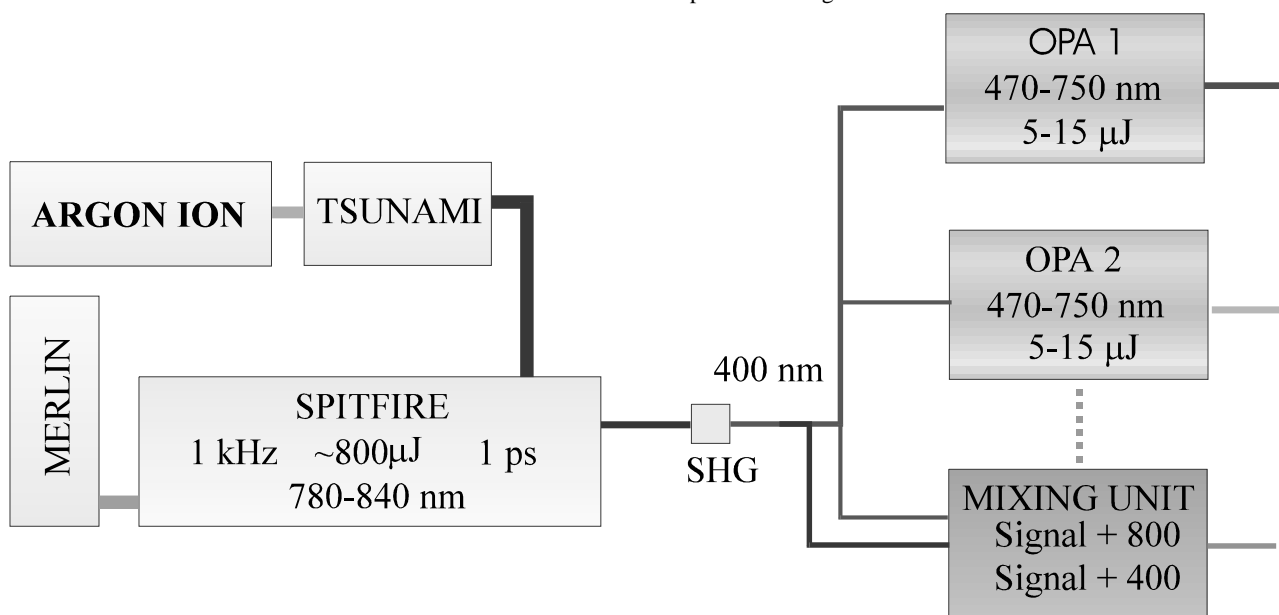


Figure 1: The Ultrafast Spectroscopy Laboratory picosecond OPA system

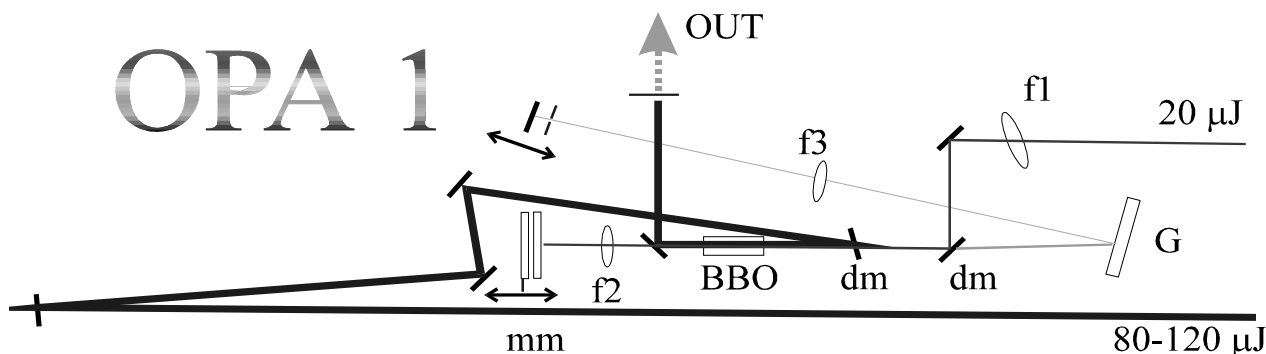


Figure 2: Expanded view of OPA 1. BBO = type I beta barium borate crystal, f1,2 and 3 are focusing lenses. DM are dichroic mirrors. G is a 600 l/mm grating. mm is a 400 nm dichroic mirror and broadband mirror combination with variable separation.

FACILITY DEVELOPMENTS

no spatial chirp before final amplification and lifts the seed beam in the vertical plane so that the amplified light can be picked off.

For the last stage the pump beam energy in OPA1 is typically 80-120 μJ and in OPA2 60-80 μJ . Care is taken to maintain good spatial beam quality in the pump and to minimise beam divergence because these have adverse effects on efficiency and linewidth. The overall pump to signal conversion efficiency of OPA1 and OPA2 is 10 % and 15 % respectively.

Typical performance curves for the OPAs are shown in Figure 3. Using both OPAs and a combination of non-linear conversion processes continuous tuning from 220 - 2200 nm at $> 0.1\text{-}1 \mu\text{J}$ at the sample is possible. We estimate the spatial beam quality to be < 2 times diffraction limited and the time bandwidth product 1.8 times that of a Gaussian pulse. Shot to shot stability is around $\pm 10\%$ but constant output is routinely maintained over 10 hours or more greatly facilitating the acquisition of weak Raman signals.

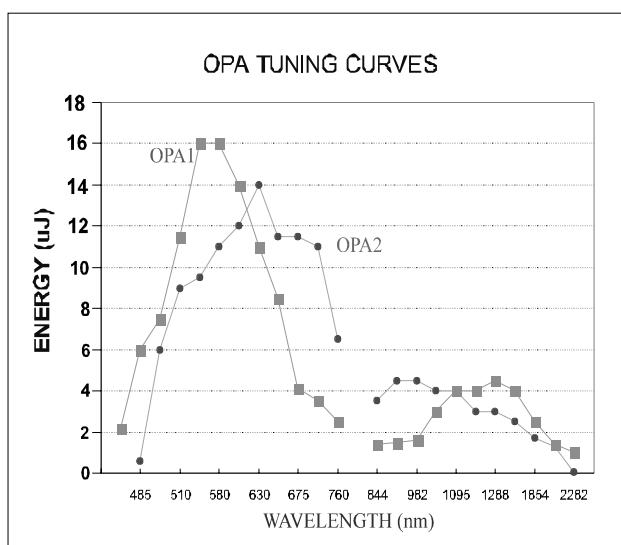


Figure 3: Signal and idler tuning curves.

For some applications where the ps TR^3 spectroscopy may require higher spectral resolution the OPA beams can be further spectral filtered using another external grating pulse shaper like the one used before the third stage of OPA1.

SPECTROSCOPIC APPARATUS

The Ultrafast Spectroscopy Laboratory houses apparatus designed for time resolved resonance Raman (TR^3) spectroscopy and complimentary transient absorption and gain spectroscopy. An optical delay up to 6 ns controls timing between pump and probe to a level of better than 50 fs. For the TR^3 experiments, beams are overlapped at the sample in a crossed beam arrangement of a few degrees using broadband aluminium mirrors and individually adjustable focusing lenses. Overlap at the sample is made using an achromatic equivalent plane imaging system consisting of a thin beam splitter, a 50 μm dye cell containing a near IR dye and a CCD with microscope objective to image the fluorescence from the pump and probe beams. The sample is renewed shot to shot by flowing the sample in an open jet. Raman scatterer light is collected in a conventional spectrometer with high quality liquid nitrogen cooled CCD.

TR^3 PERFORMANCE

The power of this new facility is demonstrated by high quality data obtained from recent experiments. More detailed of the research can be found in the Proceeding of the Conference on Time Resolved Vibrational Spectroscopy VIII^{3,4,5} recently hosted by RAL and in this report.

Figure 4 shows the spectra of the S_1 excited state of quaterphenyl taken using the frequency mixed output from OPA2 at 300 nm to pump and 605 nm probe generated in OPA1. Recent experiments on quaterphenyl⁵ show this to have remarkably simple electronic state features in its excited state and new dynamics hitherto not observed.

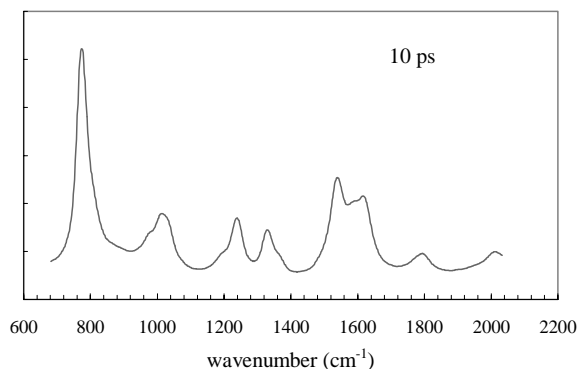


Figure 4: TR^3 spectrum of S_1 quaterphenyl pumped at 300 nm and probed at 605 nm.

Carotenes are known to play a major role in photosynthetic processes. They undergo rapid internal conversion on a femtosecond to picosecond time scale from an initially excited S_2 state to close by S_1 excited state. Truscott (Keele) has used TR^3 to study the evolution of electronic and vibrational mode coupling between these states in different solvents immediately after excitation. The carotenes were pumped at 400 nm and 475 nm and probed between 480 -700 nm using both OPAs. Analysis of data is in progress.

An example that uses only one OPA and the third harmonic of the amplifier is the study of *cis*-stilbene⁴. This molecule has been much studied by conventional pump/probe spectroscopies. It is a model system used to study isomerism from an excited state. The new OPA system has made it possible to probe this process by TR^3 . This system has never been studied by this technique before because of its very short lifetime of approximately 1 ps and the difficulty in accessing its pump and probe wavelengths. Excellent spectra were obtained and these cast new light on the fast relaxation pathways taken by this molecule.

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