Fully coherent 2D electronic spectrometer with polarization shaping capabilities

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We present a 2D spectrometer for visible spectroscopy based on two acousto-optic pulse shapers arranged in a Mach-Zehnder geometry. The setup enables the production of fully coherent, polarization-shaped pulse trains. Ultimately this feature will be exploited to observe multi-quantum coherences in molecules and colloidal nanostructures.

In a multi-dimensional spectroscopy experiment, the polarizations of the exciting and probing fields can be used to significantly enhance or suppress specific light-matter interaction pathways [1]. To serve this purpose, two acousto-optic pulse shapers (Dazzler, Fastlite) are arranged in a Mach-Zehnder geometry to create fully collinear trains of pulses with well-defined and controllable carrier-envelope phases, delays and polarization states [2]. The interferometer is stabilized to λ/225. In order to probe systems in the pulse shapers' diffracting bandwidth (480-720 nm), a visible super-continuum is generated by focusing roughly 1.3 mJ of 800 nm, 100 fs pulses into a 2.5 m hollow-core fibre of 400µm inner core diameter. Before operating the spectrometer in the fully co-linear geometry, its basic capabilities are demonstrated in the pump-probe geometry [3].

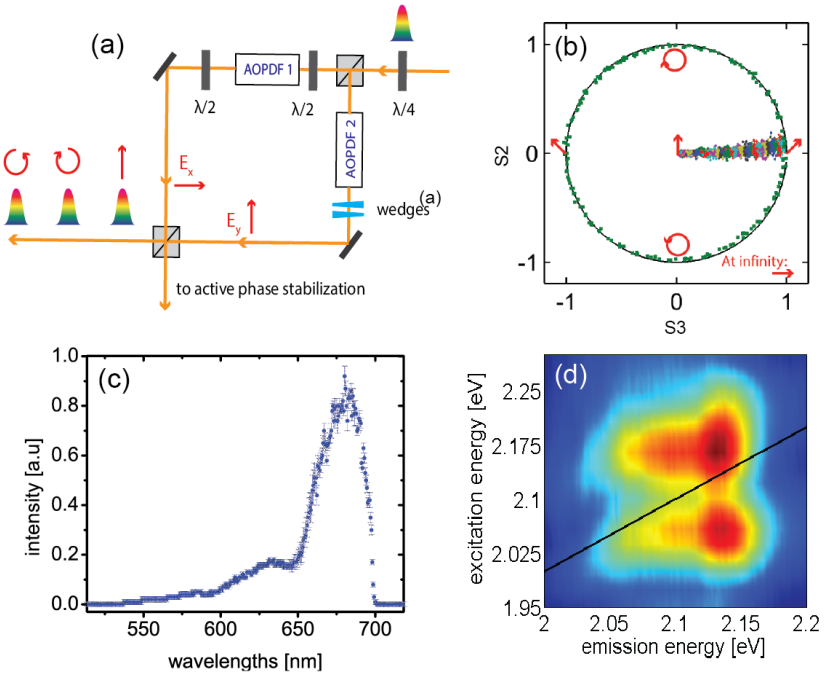

Fig.1 (a) Scheme of the dual acousto-optic pulse shapers setup. (b) Demonstration of the polarization shaping capabilities of the setup, represented on a projected Pointcaré sphere (adapted from [2]). (c) Typical spectrum obtained from the hollow core fibre (after a short-pass filter) as used for the experiments. Error bars indicate one standard deviation. The total energy over this spectral region is close to 100µJ/pulse. (d) Preliminary 2D electronic spectrum obtained on CdSe nanocrystals in the pump-probe geometry [3], exhibiting the strong coupling between the lowest two S-like transitions in the quantum dots.

[1] M.T.Zanni *et al*., PNAS **98,** 11265 (2001).
[2] H.Seiler *et al*., JAP **118,** 103110 (2015).
[3] J.A.Myers *et al*., Optics Express **16**, 17420 (2008).