Coherent 2D spectroscopy of pentacene thin films

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We present our results on the photo induced dynamics inside a prototypical organic thin film semiconductor, namely pentacene. Measurements were performed on our recently commissioned visible 2D photon echo spectrometer, which covers a spectral range between 500 to 950 nm and provides sub-10 fs passively phase stabilized pulses.

Recent research interest in carbon-based semiconductors is sparked by the advance and development of organic electronics, photovoltaics, and light-emitting diodes.(*1*, *2*) One of the main goals is to unravel and comprehend the behavior of charge-carriers in these materials, in order to develop design rules for future applications. 2D spectroscopy has shown to be a powerful technique to investigate energy transfer processes(*3*) and its application to organic semiconductors can reveal new insight into the underlying processes.(*4*)

Coherent 2D spectroscopy covering the spectral range between 540 to 740 nm reveals by which mechanism the energy of the photo excited pentacene thin film funnels down into the lowest singlet excited state on an ultrafast time-scale. These results are accompanied by transient absorption measurements of pentacene singlet crystals, which yield insight into polarization dependences and reveal intermolecular vibrational and librational modes. Ultimately, we gain insight into the mechanism of singlet fission in pentacene thin films, as well as exciton dynamics in organic semiconductors in general.

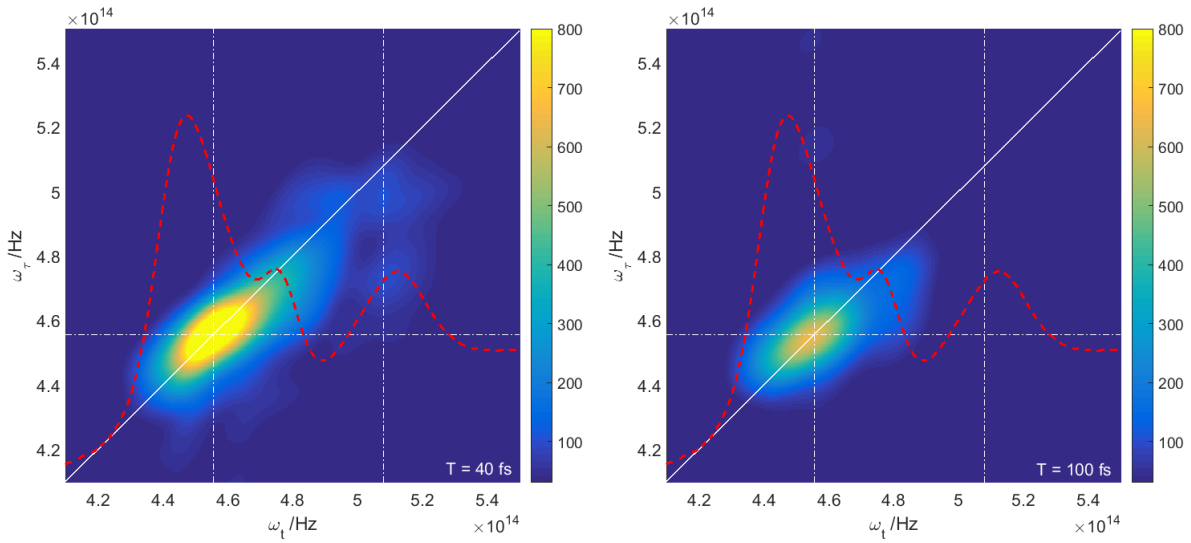

Fig 1: Absolute 2D spectra of Pentacene films at 40 fs and 100 fs population time. The peaks at 520 THz are absent in the 100 fs plot. Static absorption spectra are shown in red.

1. H. E. Katz, J. Huang, *Annual Review of Materials Research* **39**, 71 (2009).
2. Y.-W. Su *et al.*, *Materials Today* **15**, 554 (2012).
3. V. Butkus *et al.*, *Chemical Physics Letters* **545**, 40 (2012).
4. A. A. Bakulin *et al.*, *Nature Chemistry* **8**, 16 (2016).