Mapping of exciton–exciton annihilation in a molecular dimer via fifth-order femtosecond two-dimensional spectroscopy

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We present a theoretical study on exciton–exciton annihilation (EEA) in a molecular dimer. This process is monitored using a fifth-order coherent two-dimensional (2D) spectroscopy as was recently proposed by Dostál et. al. [1]. Using an electronic three-level system for each monomer, we analyze the different paths which contribute to the two-dimensional spectrum. The spectrum is determined by two entangled relaxation processes, namely the EEA and the direct relaxation of higher lying excited states. It is shown that the change of the spectrum as a function of a pulse delay can be linked directly to the presence of the EEA process.

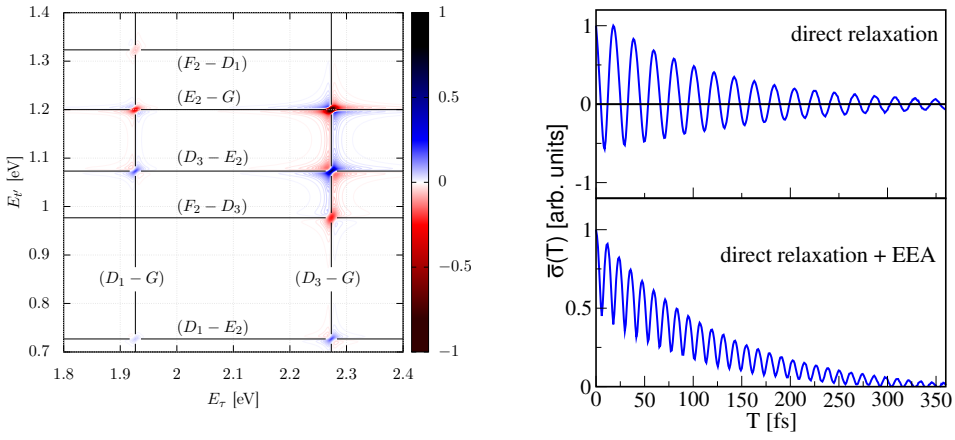

Figure 1: Left: Real part of the 2D spectrum including system-bath interactions. Right: Integrated signal $\overline{\sigma}(T)$ as a function of the population time T. The case of only direct relaxation is compared to the one where additional EEA is present.

References

1. J. Dostál, F. Fennel, F. Koch, S. Herbst, F. Würthner, and T. Brixner, *Nat. Commun.* **9** (2018), 2466.