Intra-band relaxation of Frenkel excitons in sexithiophene crystals

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A simple theoretical model [1] is proposed to describe the intra-band relaxation dynamics of the upper Davydov component deriving from the lowest Frenkel exciton in sexithiophene crystals, i.e. to estimate the efficiency of crystal-specific channels of radiationless relaxation of the intense state observed in this crystal. The process is subdivided into three stages: the first one corresponds to the decay of the discrete exciton state into one-phonon continua, followed by a fast energy dissipation through the emission of several high-energy phonons (second stage); ultimately when the remaining excess energy with respect to the lower Davydov component is smaller than one quantum of the relevant vibration the relaxation becomes slower and involves low-frequency phonons (third stage). The first two stages are predicted to be completed within 40fs, while the third (rate-determining) stage takes 500fs or more. The results are in good agreement with available experimental data (femtosecond emission and pomp-probe measurements of Frolov et al. [2]) and suggest further experiments.

[1] P. Petelenz and W. Kulig, Phys. Rev. B, submitted for publication.
[2] S. V. Frolov, C. Kloc, B. Batlogg, M. Wohlgenannt, X. Jiang, and Z. V. Vardeny, Phys. Rev. B 63, 205203 (2001)