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Theory of relaxation dynamics within carotenoids via high frequency stretching modes VYTAUTAS BALEVICIUS, DARIUS ABRAMAVICIUS, Vilnius University — Carotenoids are ubiquitous natural pigment molecules acting as light harvesters in the blue-green region of the spectrum, and at the same time ensuring the photoprotection against excessive light by quenching the triplet state of chlorophylls and singlet oxygen. However, their photophysics is still not fully understood, because the absorption takes place not into the optically dark lowest excited state S_1, but to the short-lived higher-lying state S_2. This leads to complicated intramolecular energy redistribution schemes within carotenoids. From the transient absorption experiments it is known that the S_1 state is populated shortly after the excitation of the S_2 state (on the time-scale of tens of femtoseconds). The corresponding excited state absorption signal is blue-shifting and narrowing at early times, which is attributed to the vibrational cooling of the S_1 state. We apply the secular density matrix theory to take into account both the internal conversion from the S_2 into the S_1 state and the subsequent relaxation within the manifold of high-frequency vibrational states corresponding to the carbon-carbon stretching modes (C-C and C=C). It allows us to obtain relevant pump-probe spectra in the time range from femto- to picoseconds.

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