Supporting Information

Vibronic coupling in the excited-states of carotenoids

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1 – The 1800 cm^{-1} stretching mode of Neurosporene

Figure S1 shows the evolution of the frequency of the mode at about 1800 cm^{-1} for neurosporene. The initial dynamics is dominated by a strong asymmetric and weak mode, which evolves and finally takes a central frequency at about 1800cm^{-1} at T>1ps. Due to the weak signal amplitude associated with the asymmetric band, it is challenging to fit unambiguously the central frequency. In spite of that, a tendency of the central frequency to up-shift with the T-delay can be observed similarly to the other carotenoids (Fig.6).
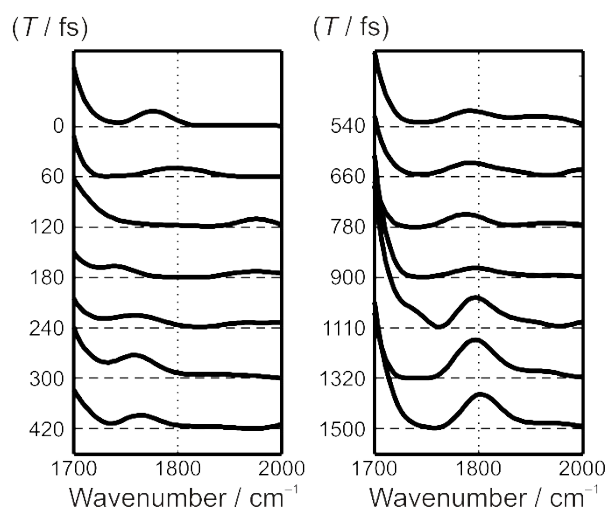

Figure **S1**. FFT spectra of C=C stretching mode (S$_1$) for neurosporene

2 – Solvent-Induced Shift of the Absorption of Lutein

Figure S2 shows the absorption spectra of lutein for three solvents investigated in the main manuscript. The more polar the solvent is, the stronger is the red-shift of the absorption. As an ionic state, the $1B_u^+$ state of lutein is stronger stabilized in comparison to other electronic states of carotenoids, which show a covalent character. Thus, the shift of the absorption spectra is mainly due to the energy shift of $1B_u^+$.
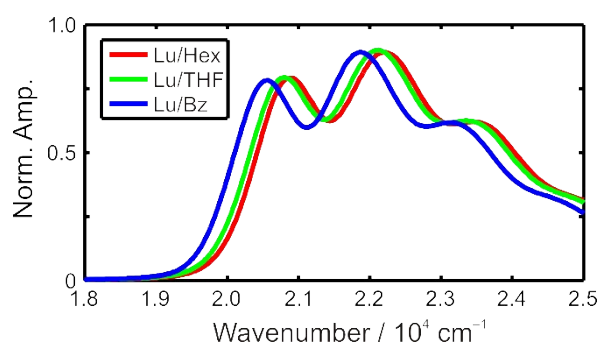

Figure S2. Stationary absorption spectra of Lutein in hexane, THF and benzene

3 – Accuracy of the Time Constants for the Sequential Model

The accuracy of the time constants for the sequential model shown at Fig.10 was determined in the following way. (I) The inverse number of root mean square of the fitting error are plotted for each time constants as fitting quality in Fig. S3. The error values are calculated for all possible combinations of the time constants. (II) Standard deviation is obtained by Gaussian fitting, which gives the following precisions: $\tau_1 = 42.6 \pm 5.4$ fs and $\tau_2 = 35.1 \pm 1.9$ fs in hexane, $\tau_1 = 38.1 \pm 2.5$ fs and $\tau_2 = 32.2 \pm 2.7$ fs in THF, and $\tau_1 = 34.7 \pm 4.8$ fs and $\tau_2 = 22.6 \pm 5.0$ fs in benzene.
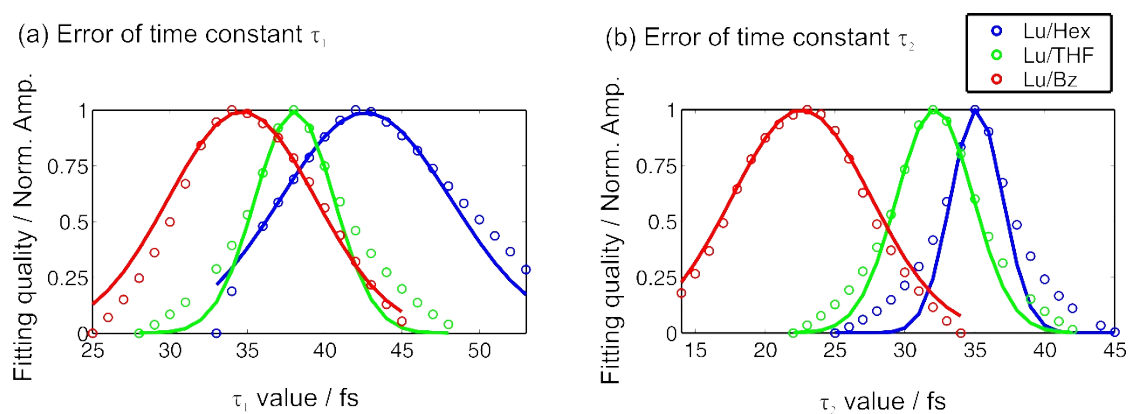

Figure S3. Fitting quality for (a) τ_1 and (b) τ_2 in three different solvents (circle).