Electronic Supporting Information (ESI)
Simulating Action-2D Electronic Spectroscopy of Quantum Dots: Insights on the Exciton and Biexciton Interplay from Detection-Mode and Time-Gating

Matteo Bruschi[1], Federico Gallina[1], and Barbara Fresch[1,2]

[1]Dipartimento di Scienze Chimiche, Università degli Studi di Padova, via Marzolo 1, Padua 35131, Italy

[2]Padua Quantum Technologies Research Center, Università degli Studi di Padova, Italy

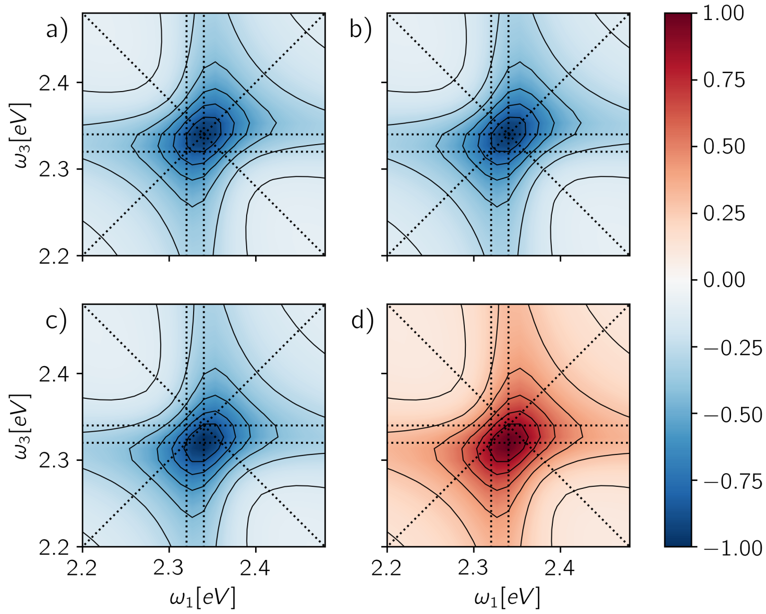

Fig. S1: Spectral representation of the four terms in the perturbative response function for the rephasing component (real part) of a three-level system in A-2DES: a) GSB, b) SE, c) ESAI and d) ESAII pathways. Assuming time-ordering, impulsive-limit and Rotating-Wave Approximation, the signal is given by the sum of the following terms:

$$\begin{aligned}
R_{GSB} &= -(i)^4 \cdot \mathrm{Tr}\{\hat{\mathcal{P}}_1 U_{11}(T_3)\hat{\mu}_{10}U_{00}(T_2+T_1)\rho(0)\hat{\mu}_{01}U^{\dagger}_{11}(T_1)\hat{\mu}_{10}U^{\dagger}_{00}(T_3+T_2)\hat{\mu}_{01}\} \\
R_{SE} &= -(i)^4 \cdot \mathrm{Tr}\{\hat{\mathcal{P}}_1 U_{11}(T_3+T_2)\hat{\mu}_{10}U_{00}(T_1)\rho(0)\hat{\mu}_{01}U^{\dagger}_{11}(T_2+T_1)\hat{\mu}_{10}U^{\dagger}_{00}(T_3)\hat{\mu}_{01}\} \\
R_{ESAI} &= -(i)^4 \cdot \mathrm{Tr}\{\hat{\mathcal{P}}_1 \hat{\mu}_{12}U_{22}(T_3)\hat{\mu}_{21}U_{11}(T_2)\hat{\mu}_{10}U_{00}(T_1)\rho(0)\hat{\mu}_{01}U^{\dagger}_{11}(T_3+T_2+T_1)\} \\
R_{ESAII} &= (i)^4 \cdot \mathrm{Tr}\{\hat{\mathcal{P}}_2 U_{22}(T_3)\hat{\mu}_{21}U_{11}(T_2)\hat{\mu}_{10}U_{00}(T_1)\rho(0)\hat{\mu}_{01}U^{\dagger}_{11}(T_3+T_2+T_1)\hat{\mu}_{12}\}
\end{aligned} \tag{2}$$

where $\rho(0) = |0\rangle\langle 0|$ is the initial density matrix, $\hat{\mu}_{kk'} = \mu_{kk'}|k\rangle\langle k'|$ is the component of the dipole operator inducing a transition in the system, $U_{kk}(T_i)$ are elements of the time-evolution operator, with delay-time between the pulses T_i. For the sake of clarity, the response functions are written in the context of unitary dynamics, while the computation includes the effect of the dephasing. Each map is normalized with respect to its absolute maximum/minimum.

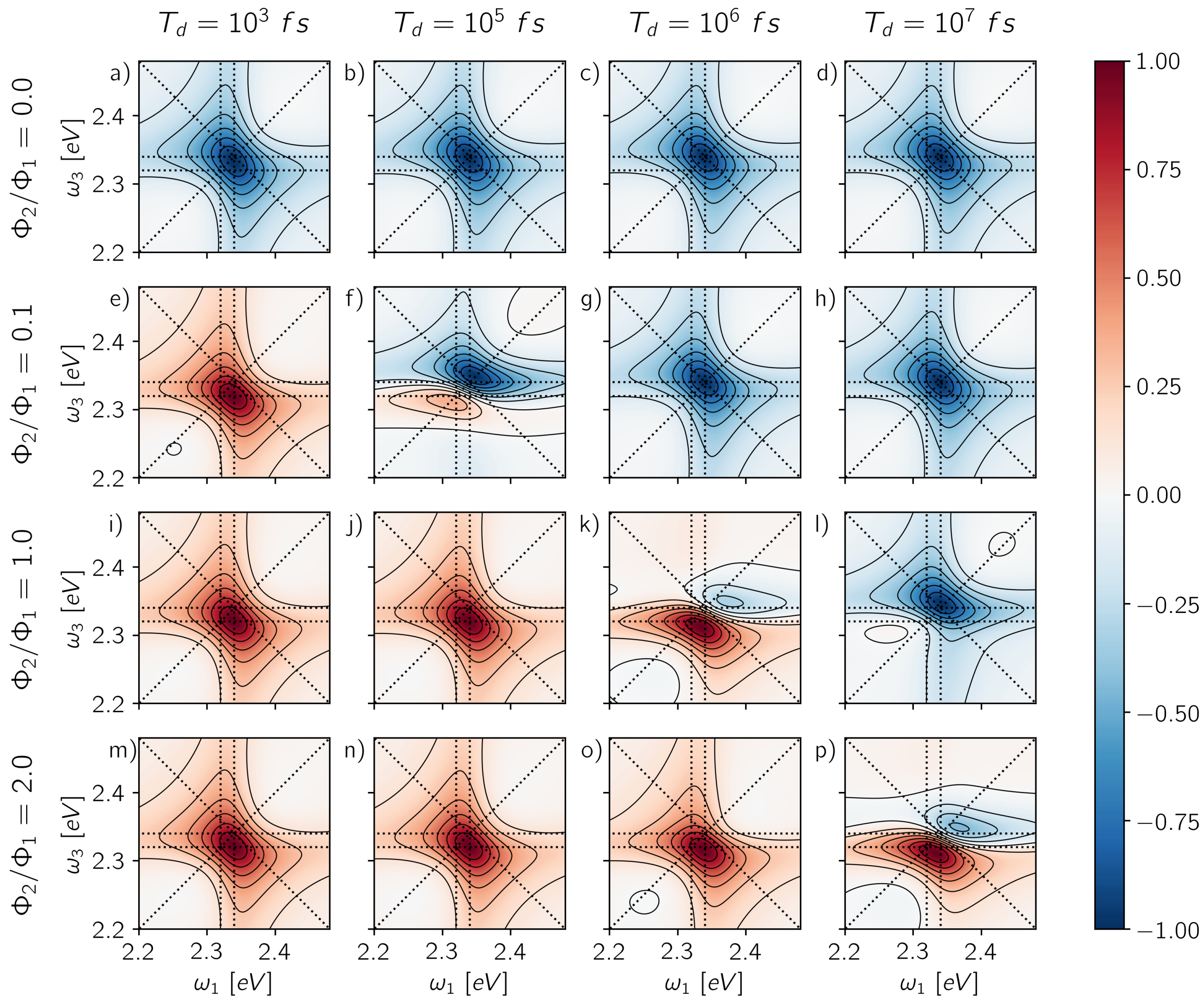

Fig. S2: Non-Rephasing maps for different values of GQY ratio ($\Phi_2/\Phi_1 = 0.0,\ 0.1,\ 1.0,\ 2.0$) on the columns and different detection-times ($T_d = 10^3$ fs, 10^5 fs, 10^6 fs, 10^7 fs) on the rows. Each map is normalized with respect to its absolute maximum/minimum.

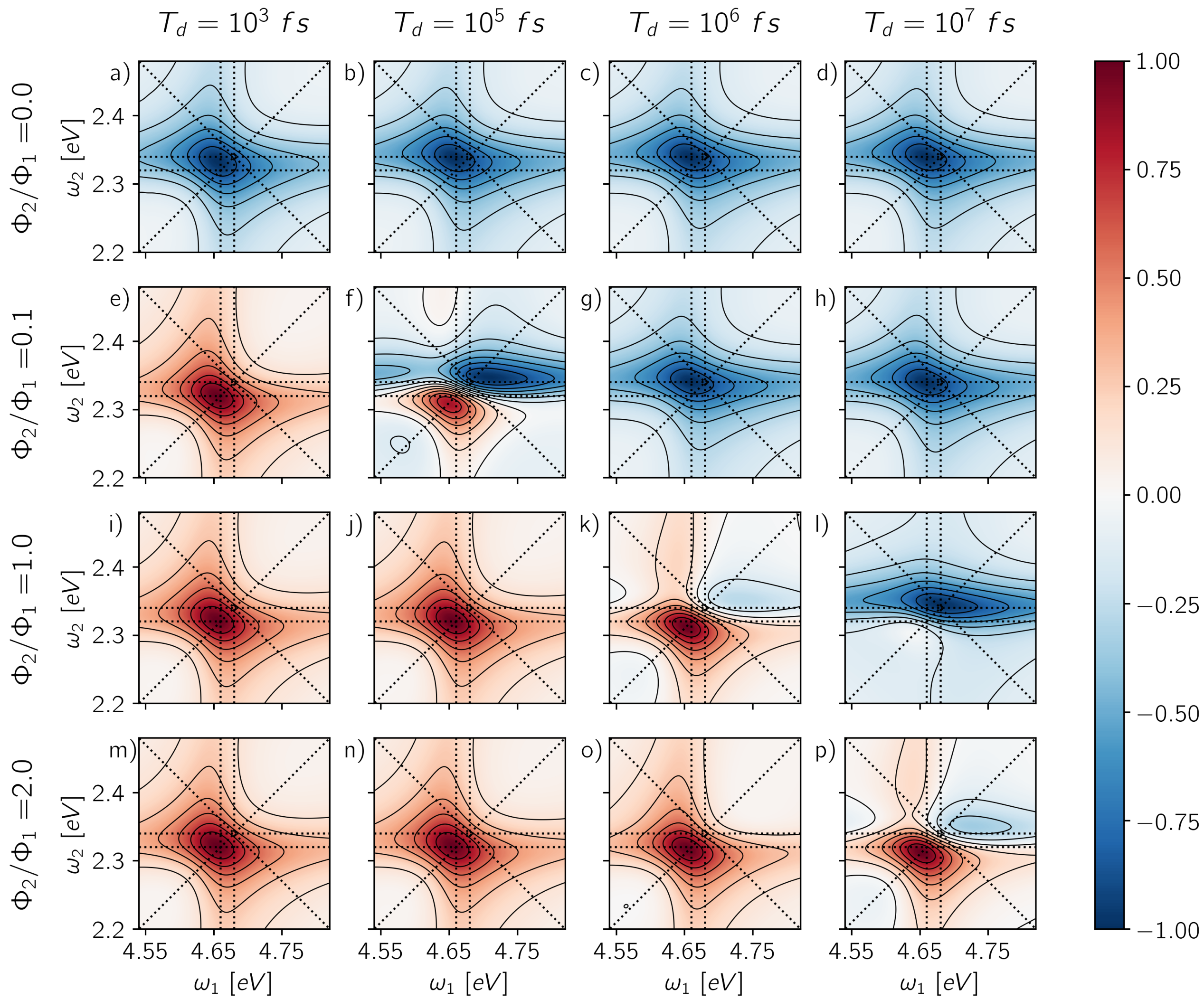

Fig. S3: 2Q1Q maps for different values of GQY ratio ($\Phi_2/\Phi_1 = 0.0,\ 0.1,\ 1.0,\ 2.0$) on the columns and different detection-times ($T_d = 10^3$ fs, 10^5 fs, 10^6 fs, 10^7 fs) on the rows. Each map is normalized with respect to its absolute maximum/minimum.

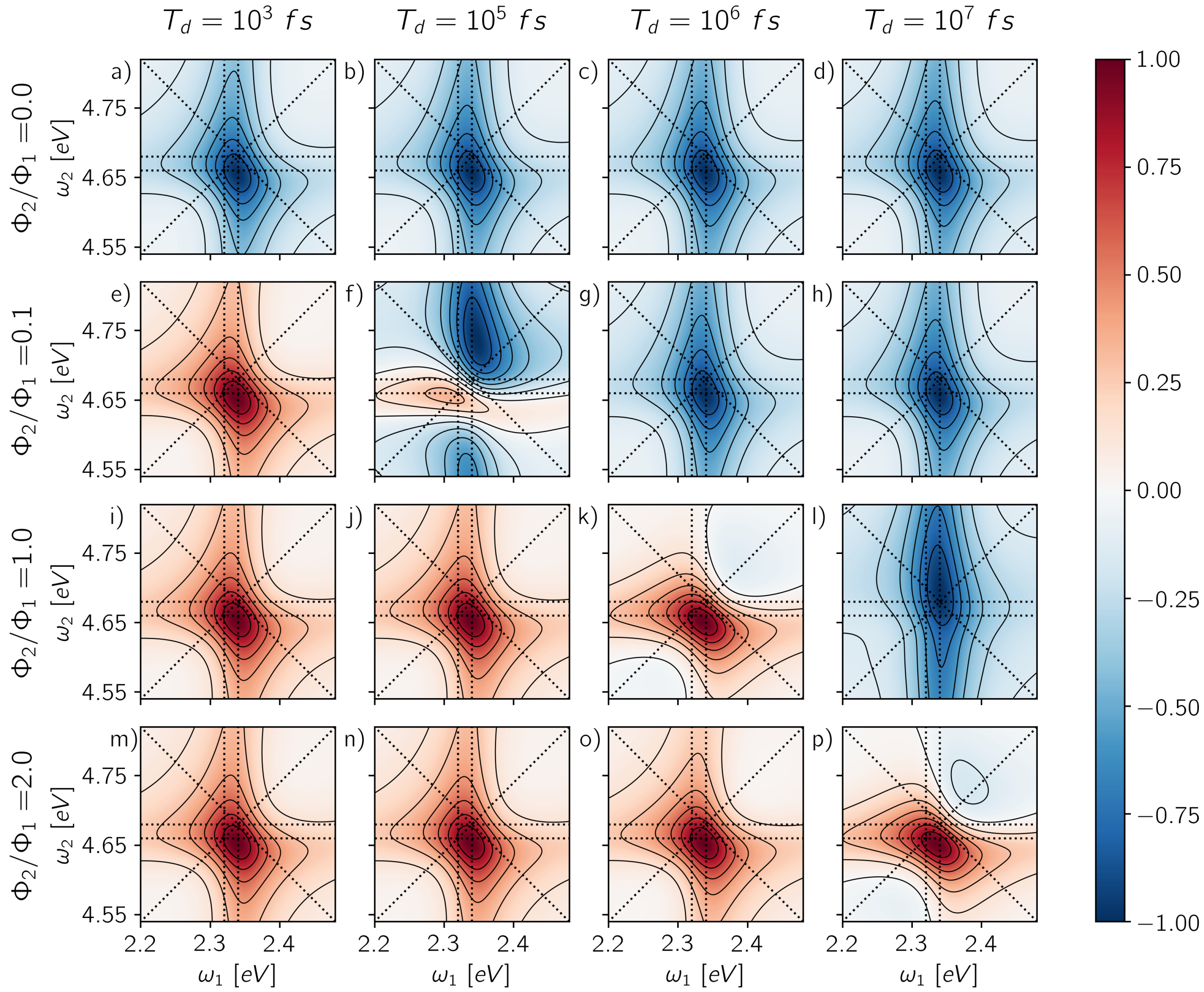

Fig. S4: 1Q2Q maps for different values of GQY ratio ($\Phi_2/\Phi_1 = 0.0,\ 0.1,\ 1.0,\ 2.0$) on the columns and different detection-times ($T_d = 10^3$ fs, 10^5 fs, 10^6 fs, 10^7 fs) on the rows. Each map is normalized with respect to its absolute maximum/minimum.

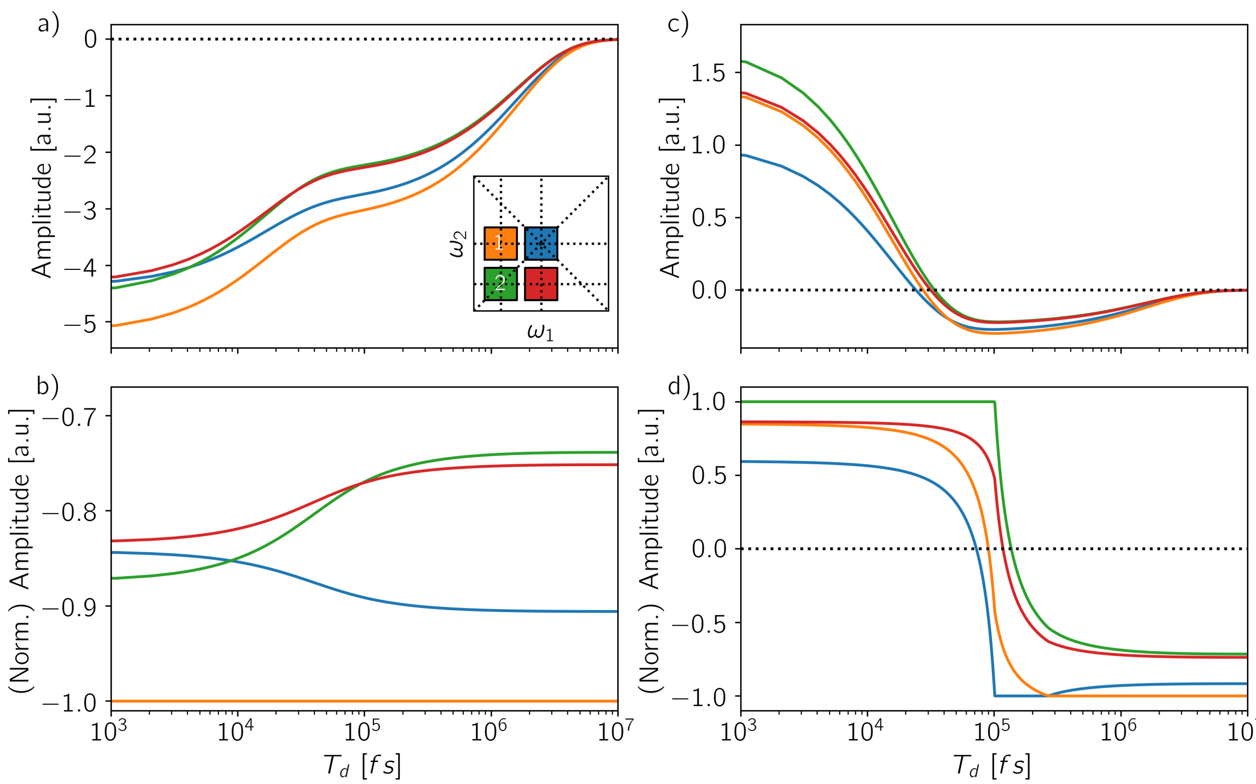

Fig. S5: Temporal evolution of the amplitude along the detection-time T_d for four coordinates in the 2Q1Q map (inset of a): at coordinates $(\omega_{20}, \omega_{10})$ 2Q1QI pathway from the exciton (orange), at coordinates $(\omega_{20}, \omega_{21})$ 2Q1QII pathway from the biexciton (green), while coordinates at $(\omega_{20} + \Delta, \omega_{21})$ (red) and $(\omega_{20} + \Delta, \omega_{10})$ (blue) represent two control positions. a) Time-resolved signal and b) (normalized) time-integrated signal for GQY ratio $\Phi_2/\Phi_1 = 0.0$. c) Time-resolved signal and d) (normalized) time-integrated signal for GQY ratio $\Phi_2/\Phi_1 = 0.1$. The normalization is taken with respect to the absolute maximum/minimum of each map.

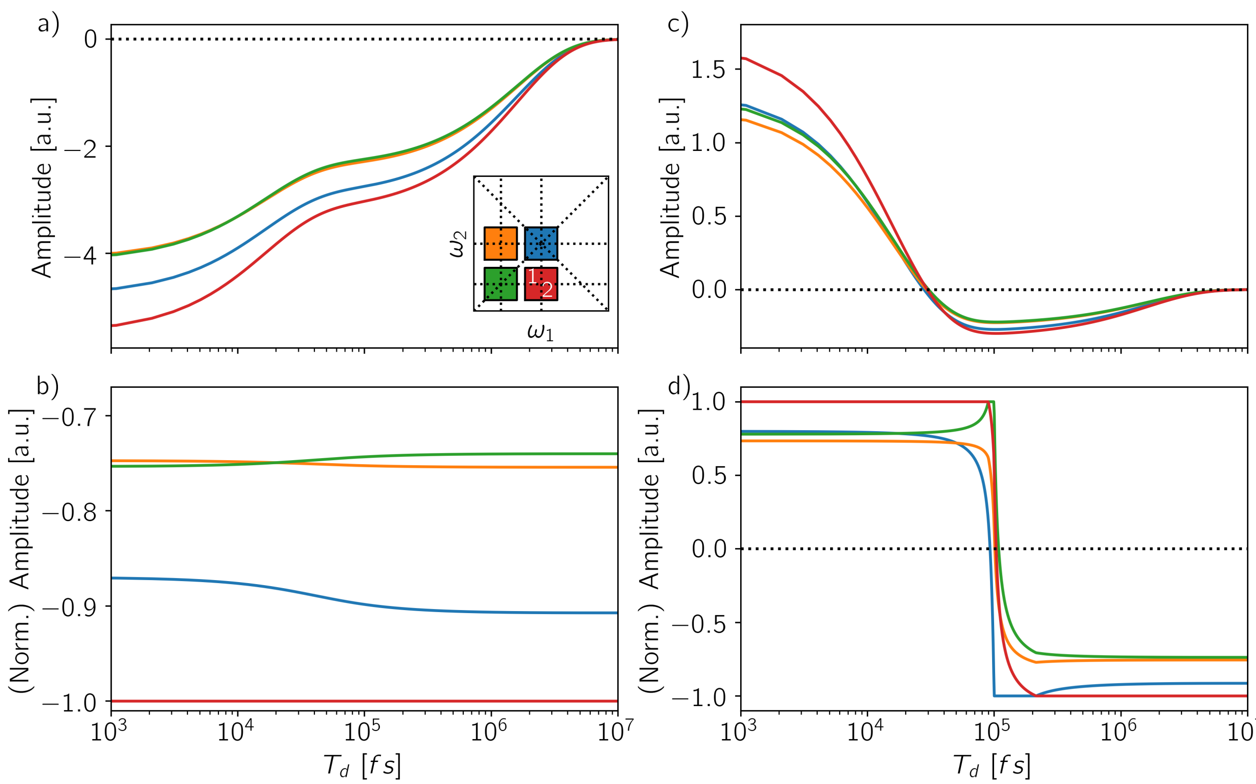

Fig. S6: Temporal evolution of the amplitude along the detection-time T_d for four coordinates in the 1Q2Q map (inset of a): at coordinates $(\omega_{10}, \omega_{20})$ 1Q2QI from the exciton and 1Q2QII from the biexciton (red), while coordinates at $(\omega_{10} - \Delta, \omega_{20})$ (green), at $(\omega_{10}, \omega_{20} + \Delta)$ (blue) and at $(\omega_{10} - \Delta, \omega_{20} + \Delta)$ (orange) represent three control positions. a) Time-resolved signal and b) (normalized) time-integrated signal for GQY ratio $\Phi_2/\Phi_1 = 0.0$. c) Time-resolved signal and d) (normalized) time-integrated signal for GQY ratio $\Phi_2/\Phi_1 = 0.1$. The normalization is taken with respect to the absolute maximum/minimum of each map.