

The Ultrafast dynamics of intraband excitons in HgS CQD's

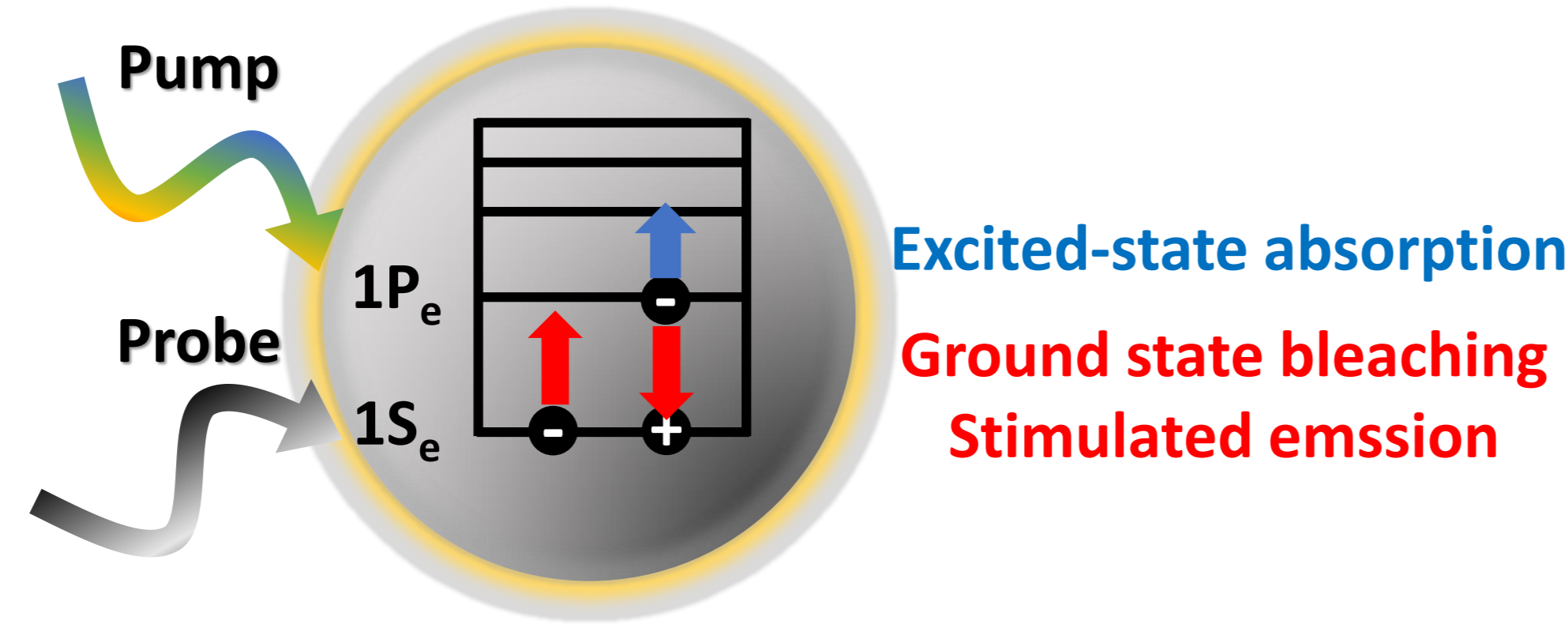
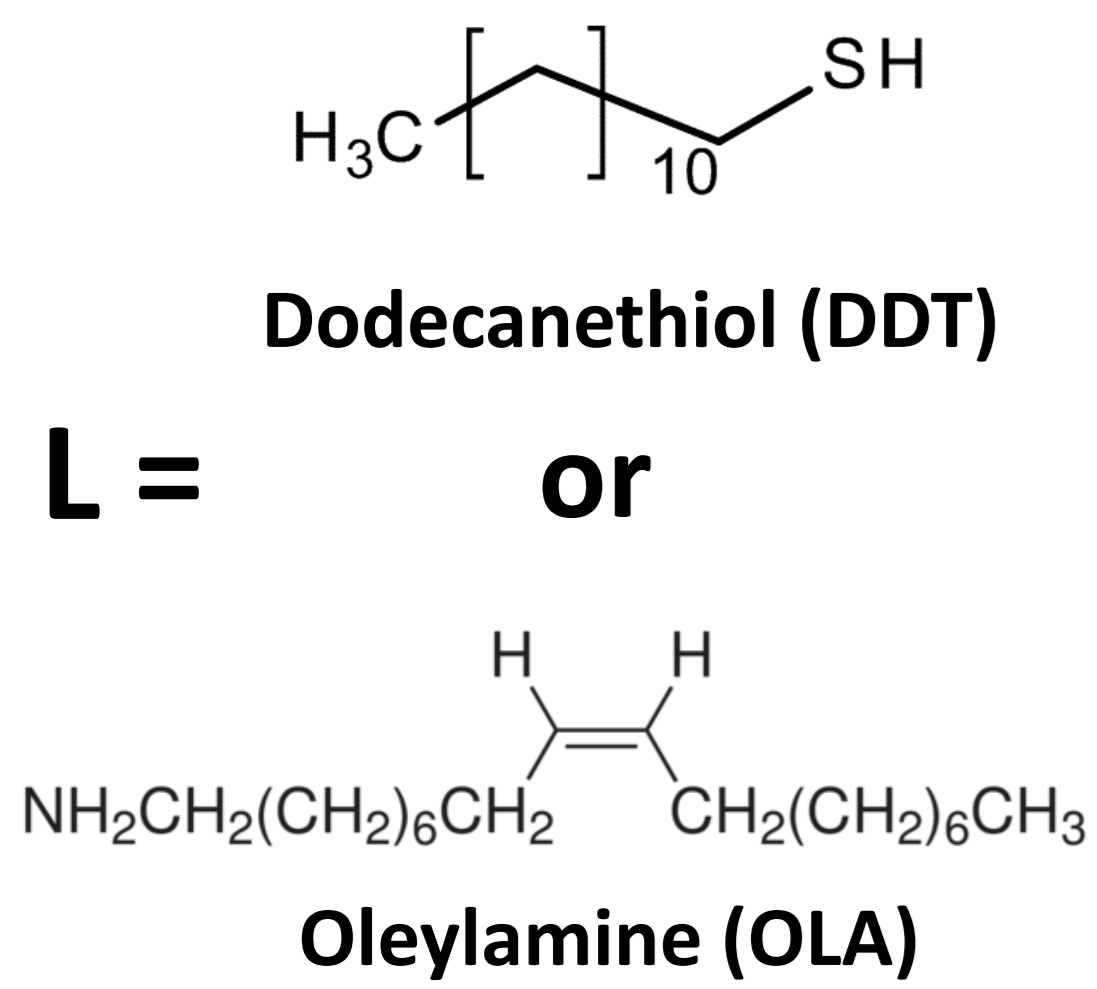
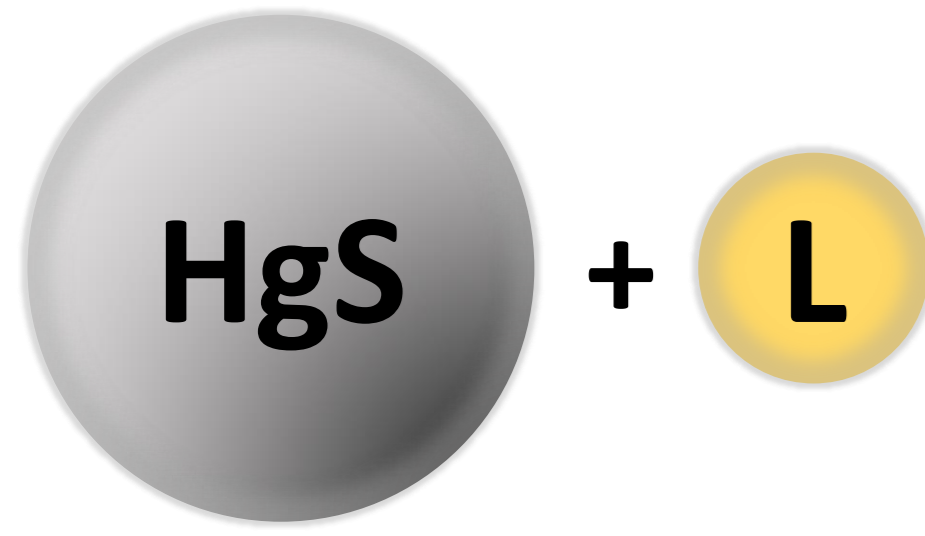
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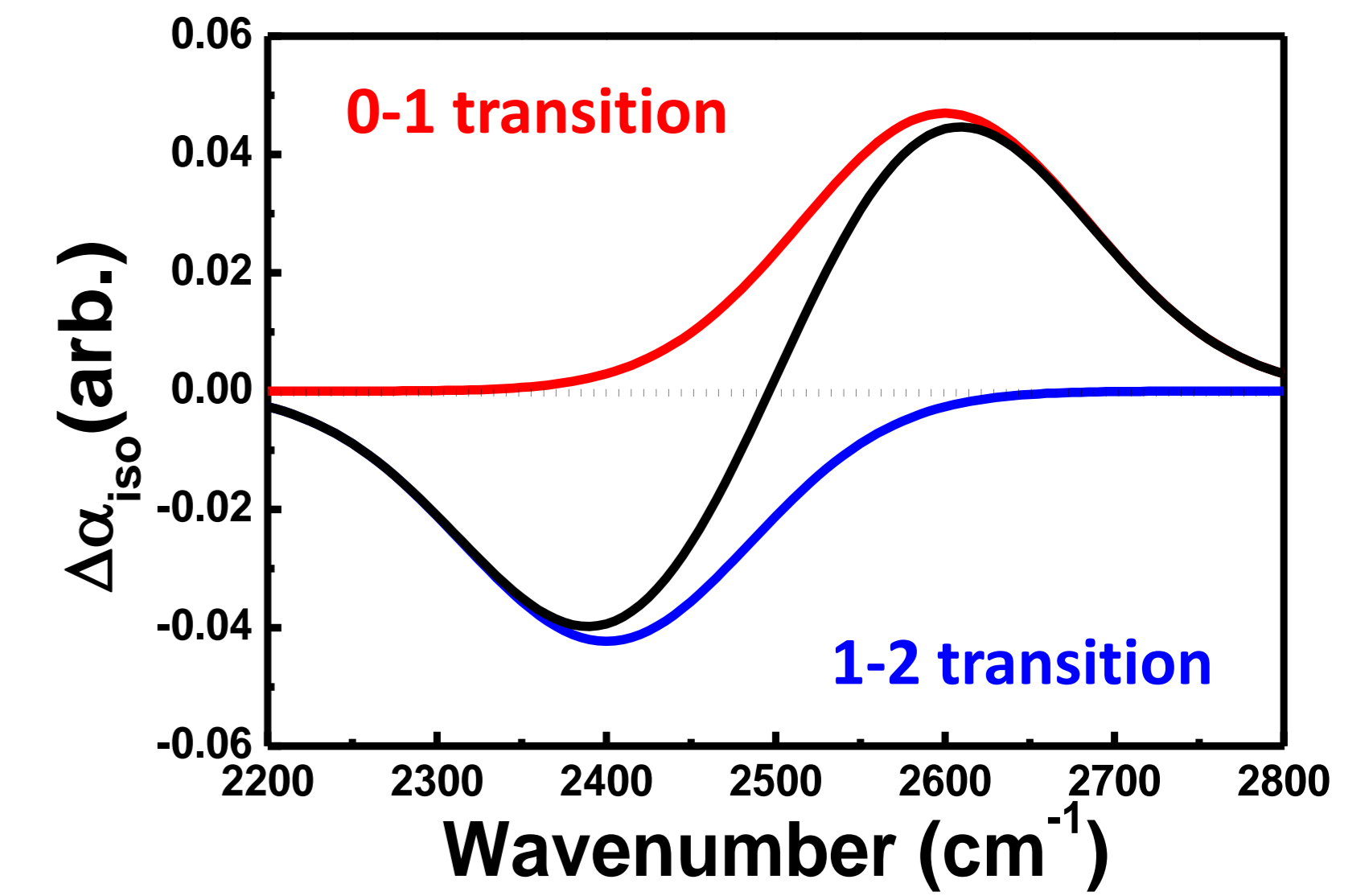
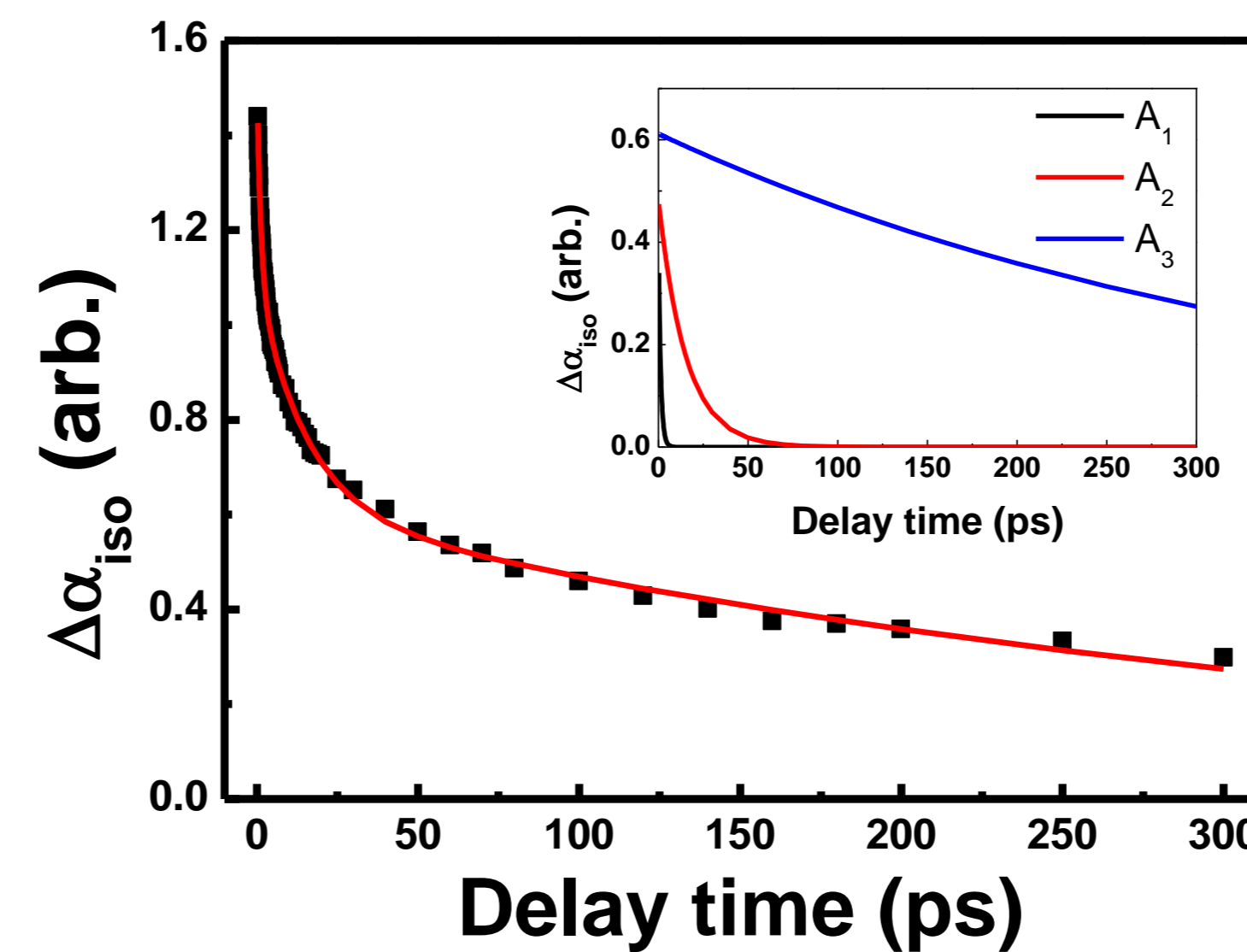


Introduction

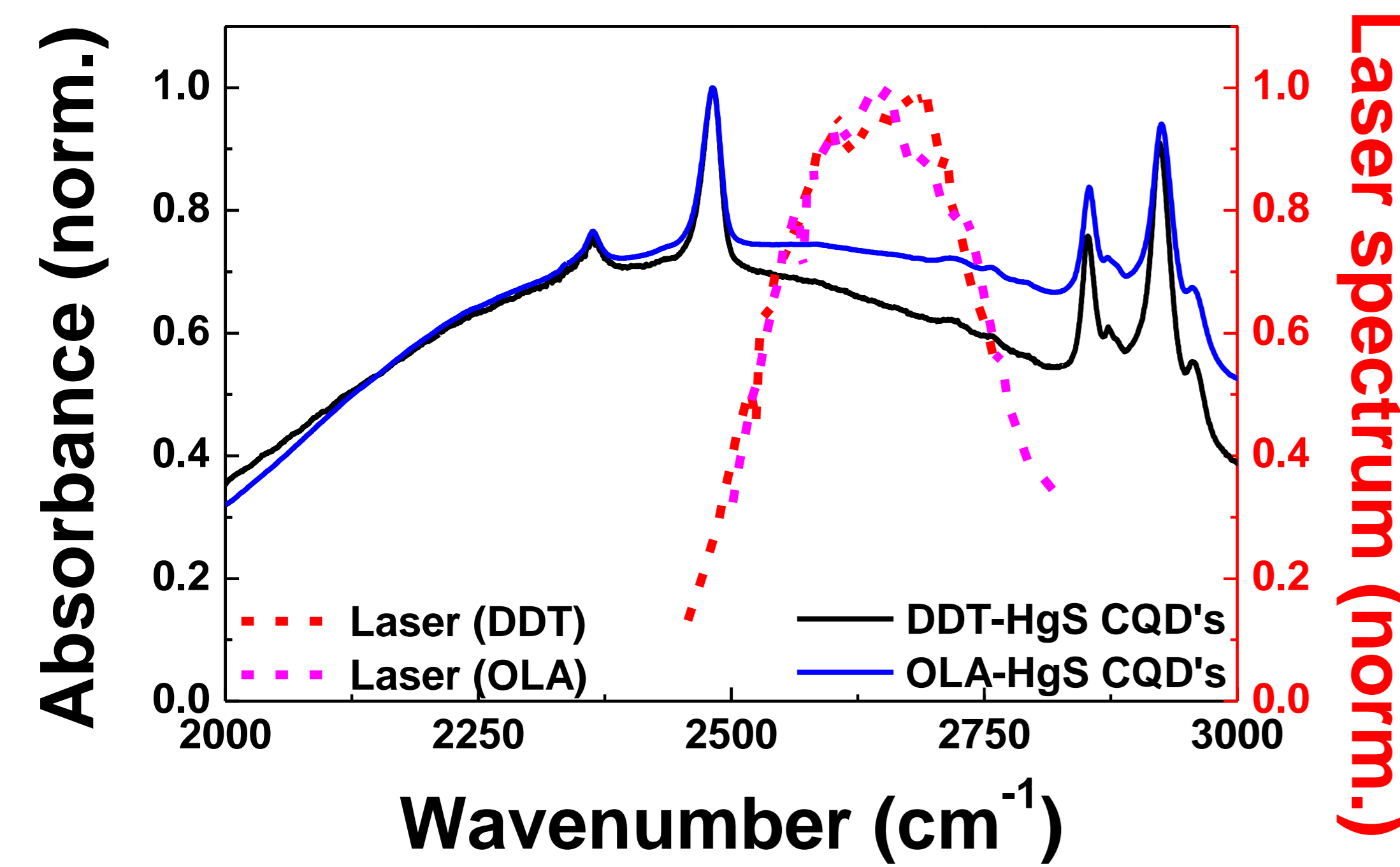
The HgS Collidal Quantum Dots (CQDs) with Mid-IR intraband gap can be fascinating materials for IR photodevices and laser¹. Here, we reported the interesting exciton relaxation dynamics of Dodecanethiol-doped HgS CQD's observed with femtosecond IR pump-probe experiments. With the variation of pump-power, we observed pump power-independent fast decaying dynamics (1.2 ± 0.1 ps) which might be caused by Auger process in CQDs after biexciton generation. Slow decay process (>300 ps) were also measured and assigned as Phonon bottleneck process for CQDs with single exciton state. Moreover, the intermediate component (~20 ps) is also appeared. To address the physical meaning of this component, we did the IR pump-probe experiments varying the ligands of CQD to check the effect of surface states and finally could assigned this intermediate as the surface component



✓ By varying the time between pump and probe pulse, we can get the information of the population of electrons in the conduction band.



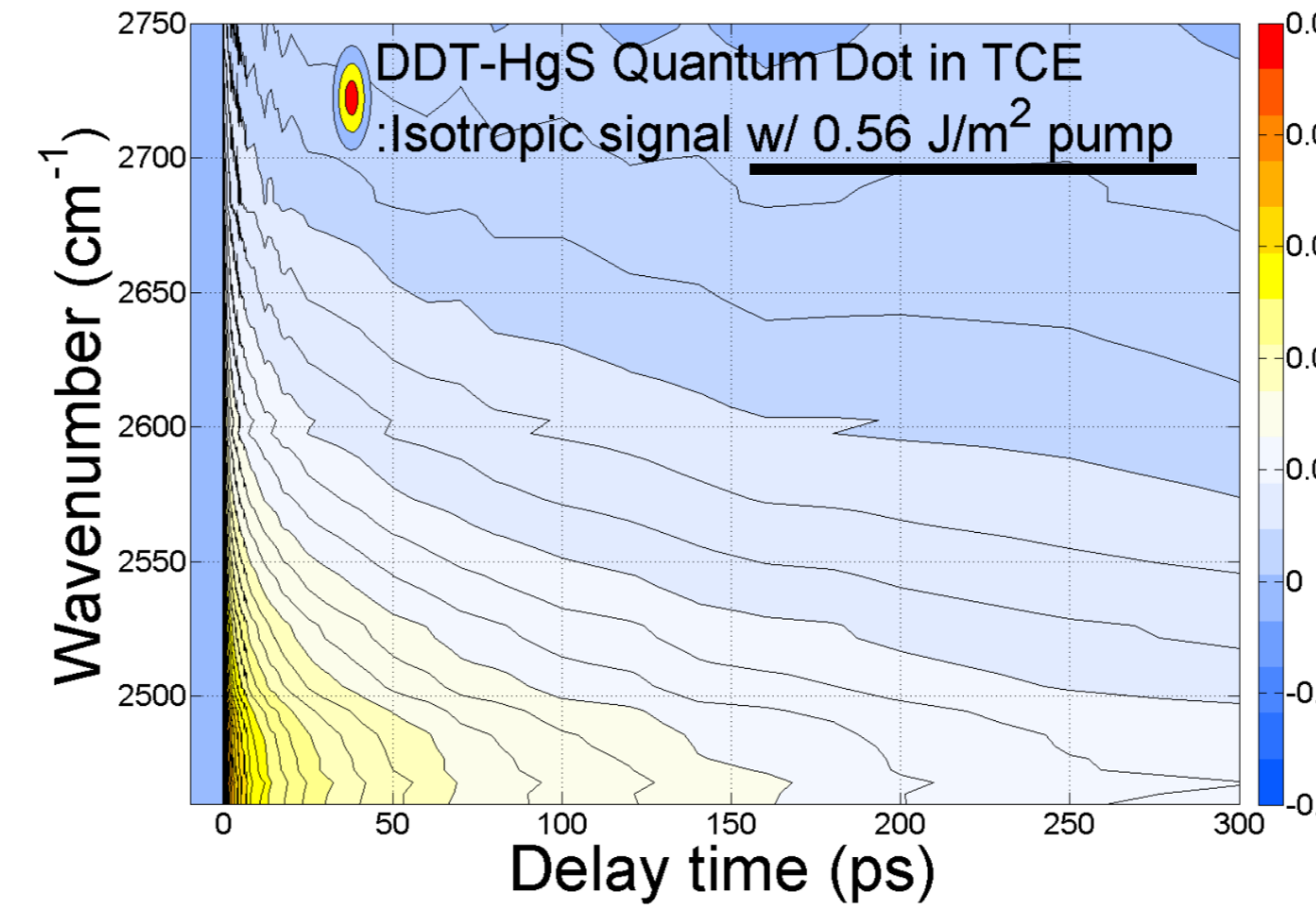
Experimental results



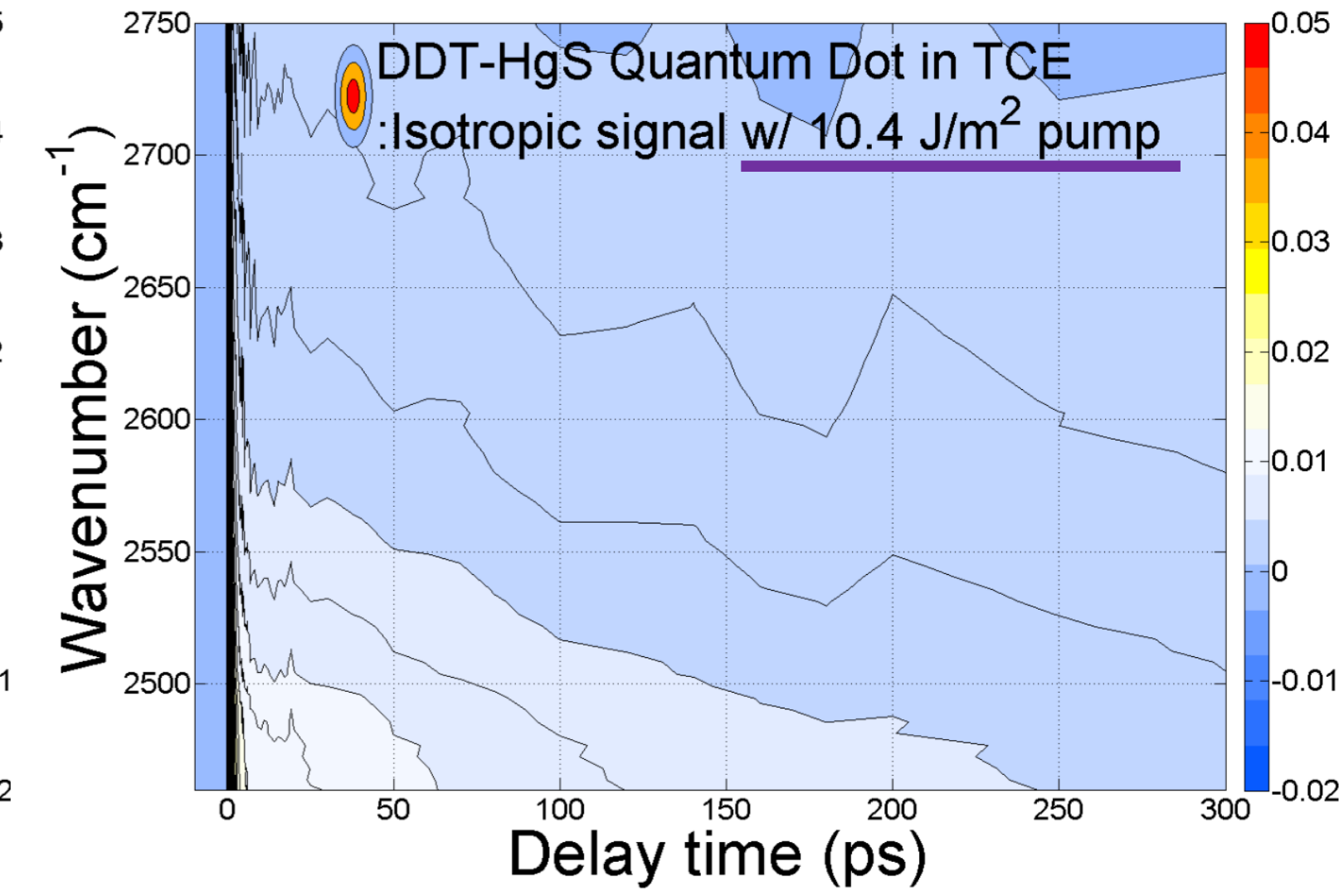
FT-IR spectrum of each samples and laser spectrum used in IR PP

- ✓ The quantum dots are passivated by organic ligand (DDT(-SH) and OLA(-NH₂))
- ✓ By controlling the Fermi level, we can successively put the electrons on conduction band. (it is easily known by FT-IR spectrum; our IR spectrum shows the strong absorption at ~2600 cm⁻¹)
- ✓ We choose the spectrally clean range to study the neat dynamics of them.

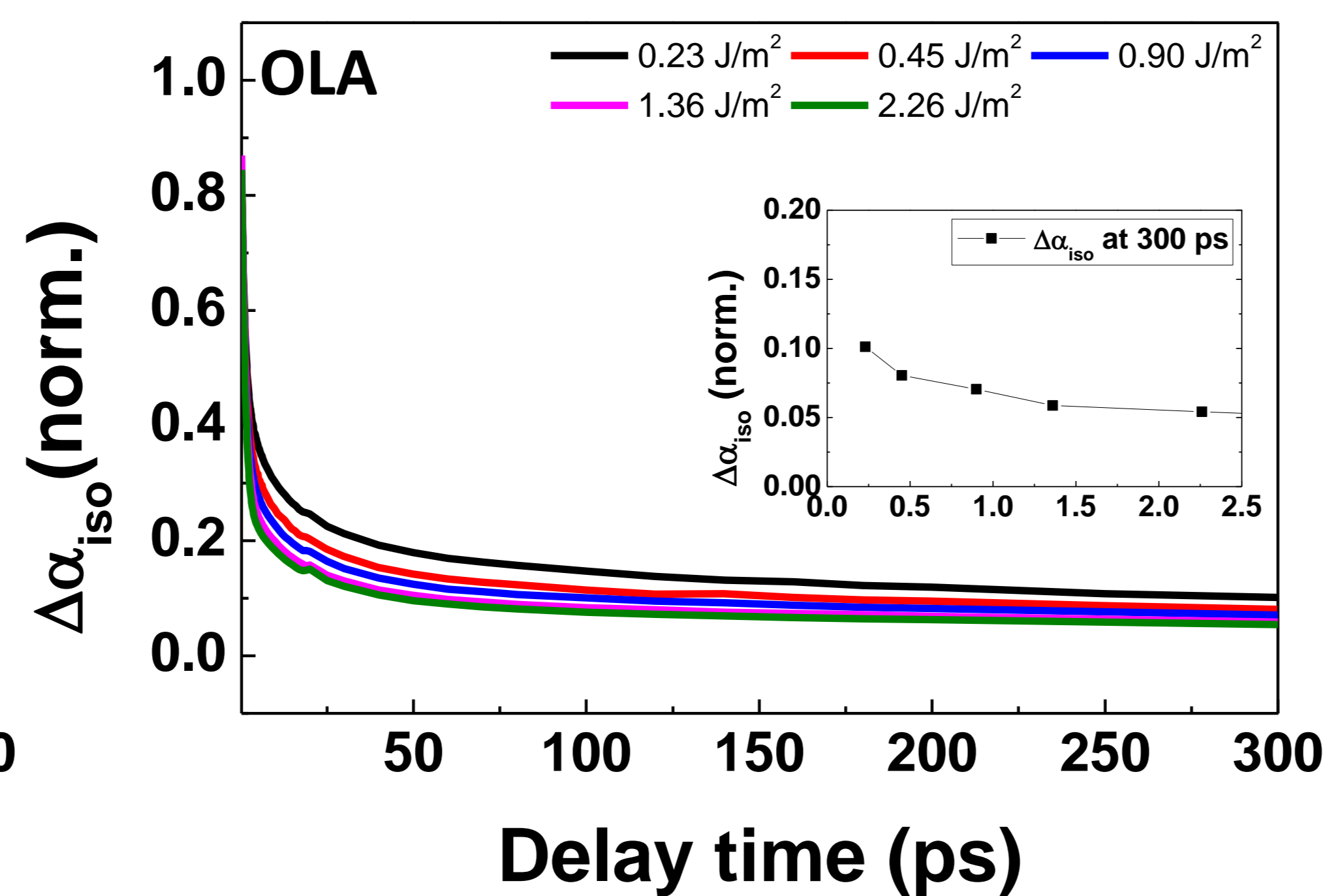
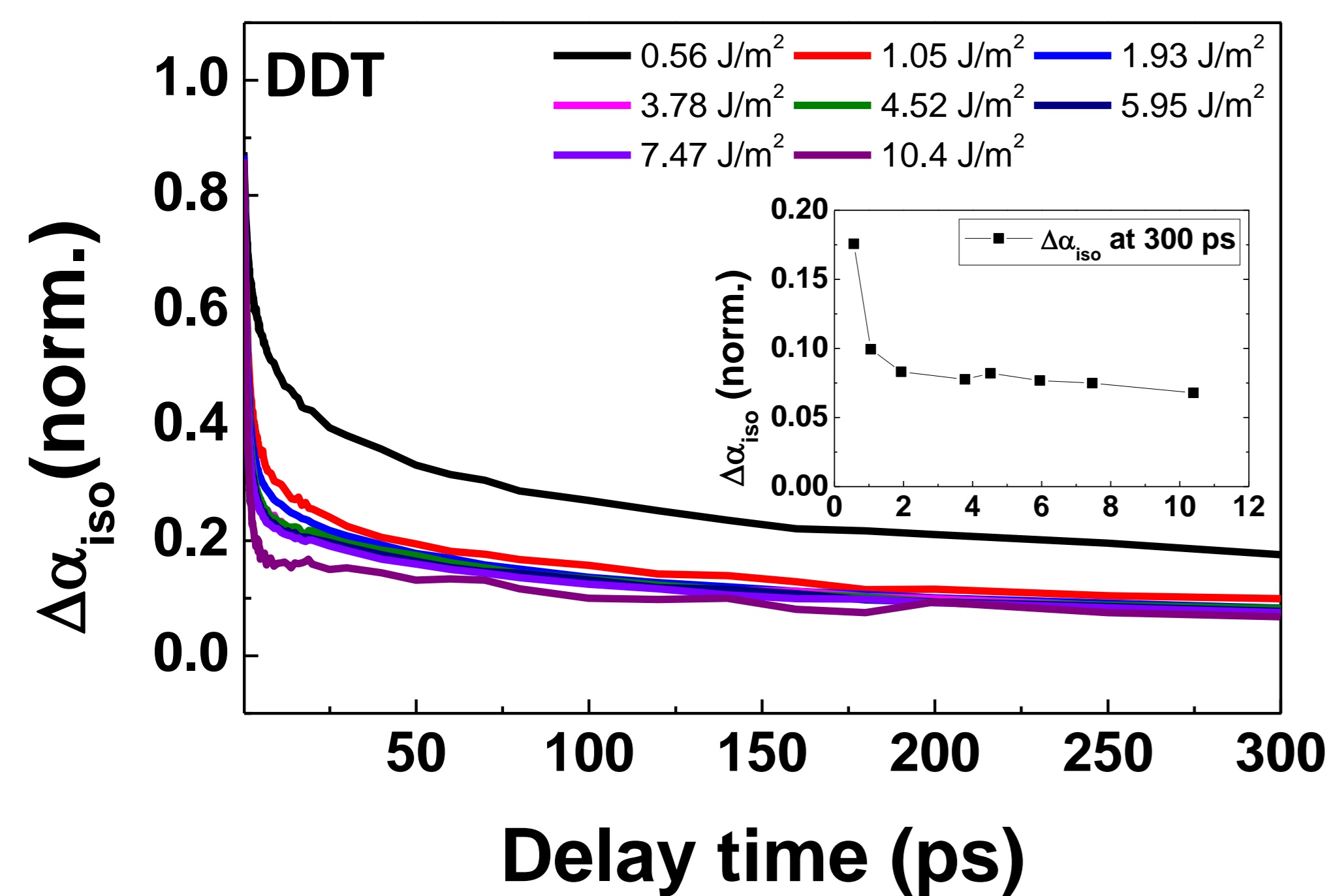
The weakest pump energy



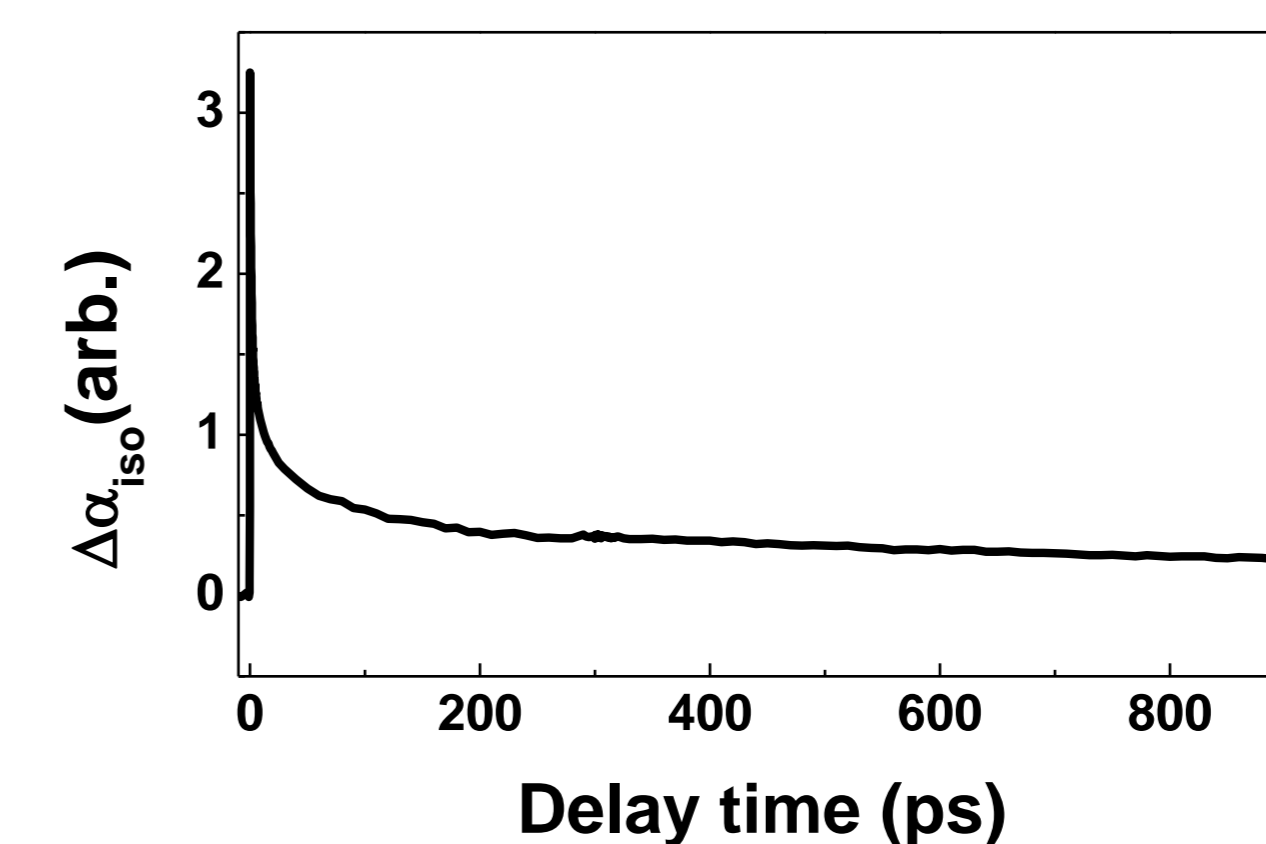
The most intense pump energy



- ✓ The contour plot of isotropic signal with weakest (0.56 J/m²) and the most intense pump (10.4 J/m²)
- ✓ The left panel shows long-lived component clearly, but it is weak in the case of 10.4 J/m²

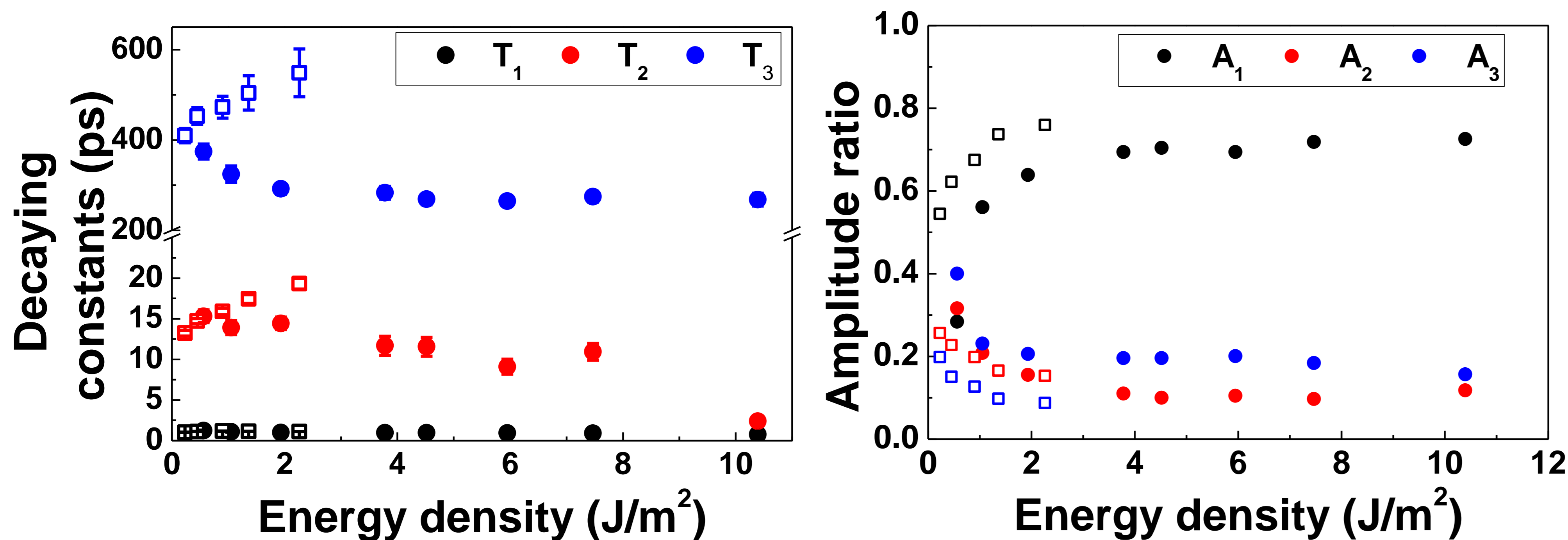


- ✓ The sum of isotropic signal of DDT-HgS colloidal quantum dots (left) and OLA-HgS cqd in TCE (right).
- ✓ The amplitude of the long-lived components decreases as function of the energy density of pump pulses.

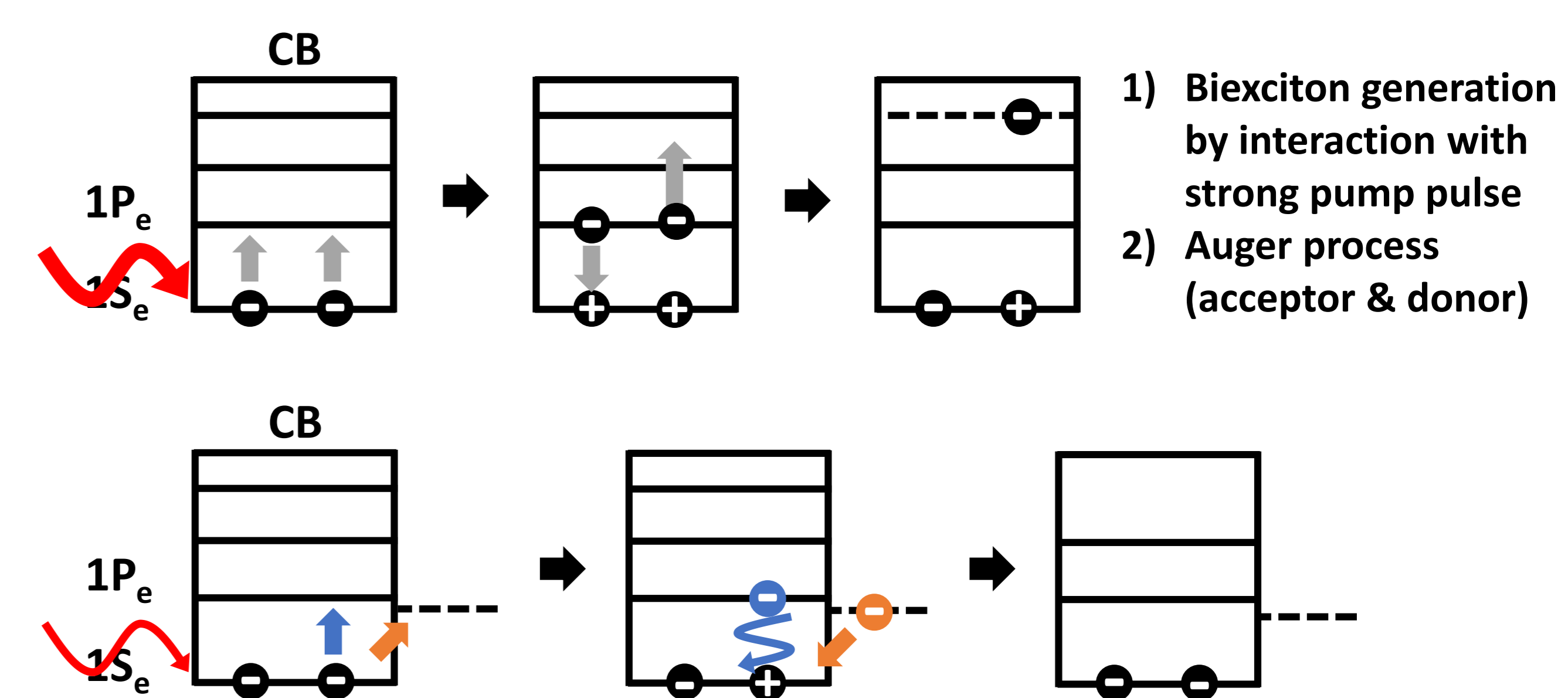


- ✓ The long-delay scan to track the highly excited electron via accepting the energy from its neighbor
- ✓ Unfortunately, it cannot be detected in 900 ps time window

Discussion



- 1) Auger components : Amplitude ratio is proportional to the pump-energy density (~1 ps)
- 2) Surface components : Its amplitude ratio depends on the type of ligand
- 3) Phonon-bottleneck : The lifetime is extremely long (>300 ps) & its amplitude ratio is also dependent on the pump-energy density



- 1) Single exciton generation by interaction with weak pump pulse
- 2-a) Electron is located at 1P_e; there is no phonon-mode who can accelerate the decay by accepting their energy (>300ps)
- 2-b) If there is hole in the surface state (defects), the electron can transfer to their. It will be accelerated by the fluctuations of environments.

Conclusion

With the pump-power dependency and decaying constants of each components, we can successively label the each components. Power-dependence of its amplitude ratio can distinguish the auger component and phonon-bottleneck. And the strength of binding of each ligand let us label the 2nd component as the surface state. We expect the phonon-bottleneck components (slow components) makes this quantum dot as the candidates for photodevices.

Acknowledgement

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