

Two-dimensional electronic spectroscopy of Zn-naphthalocyanine derivative

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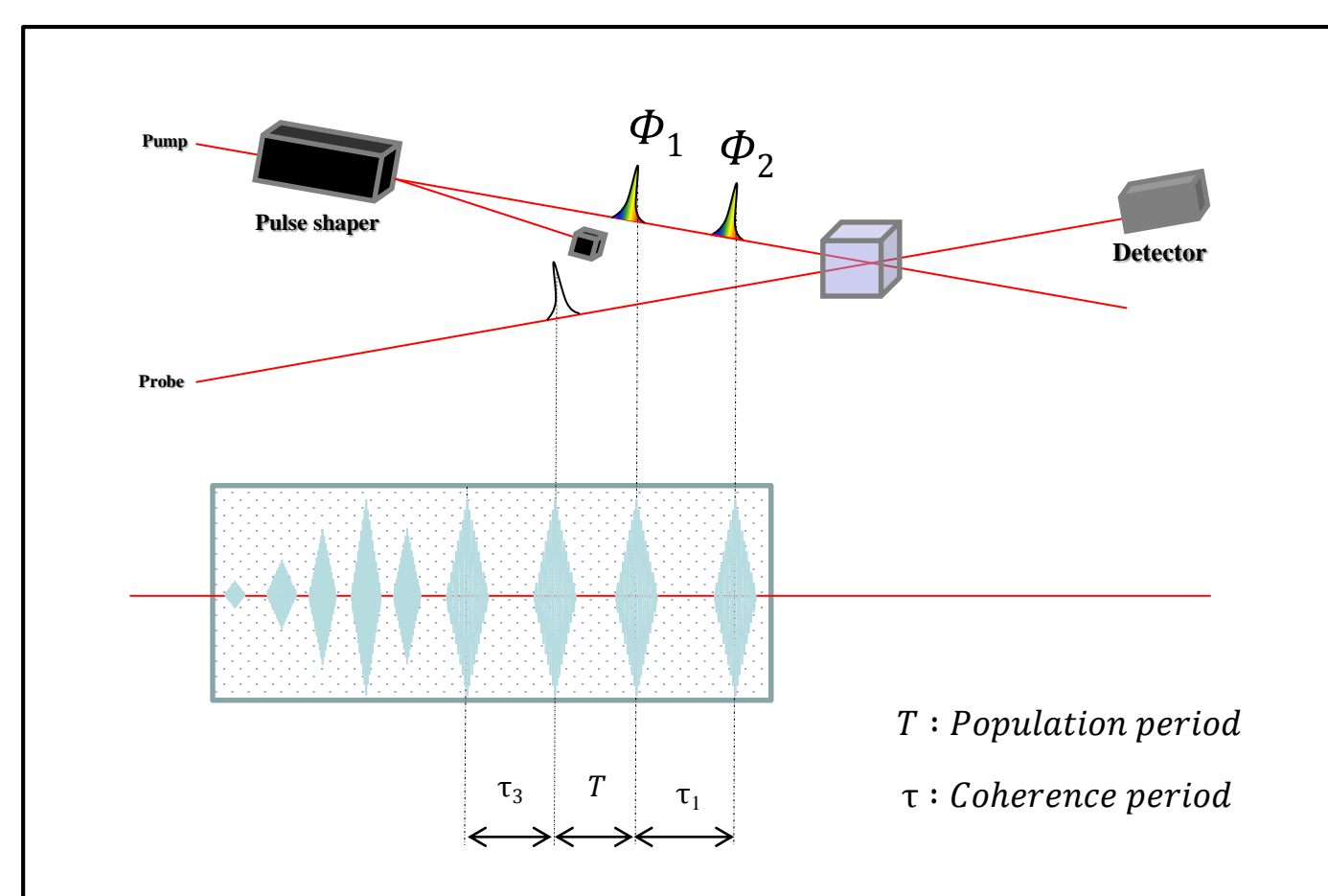
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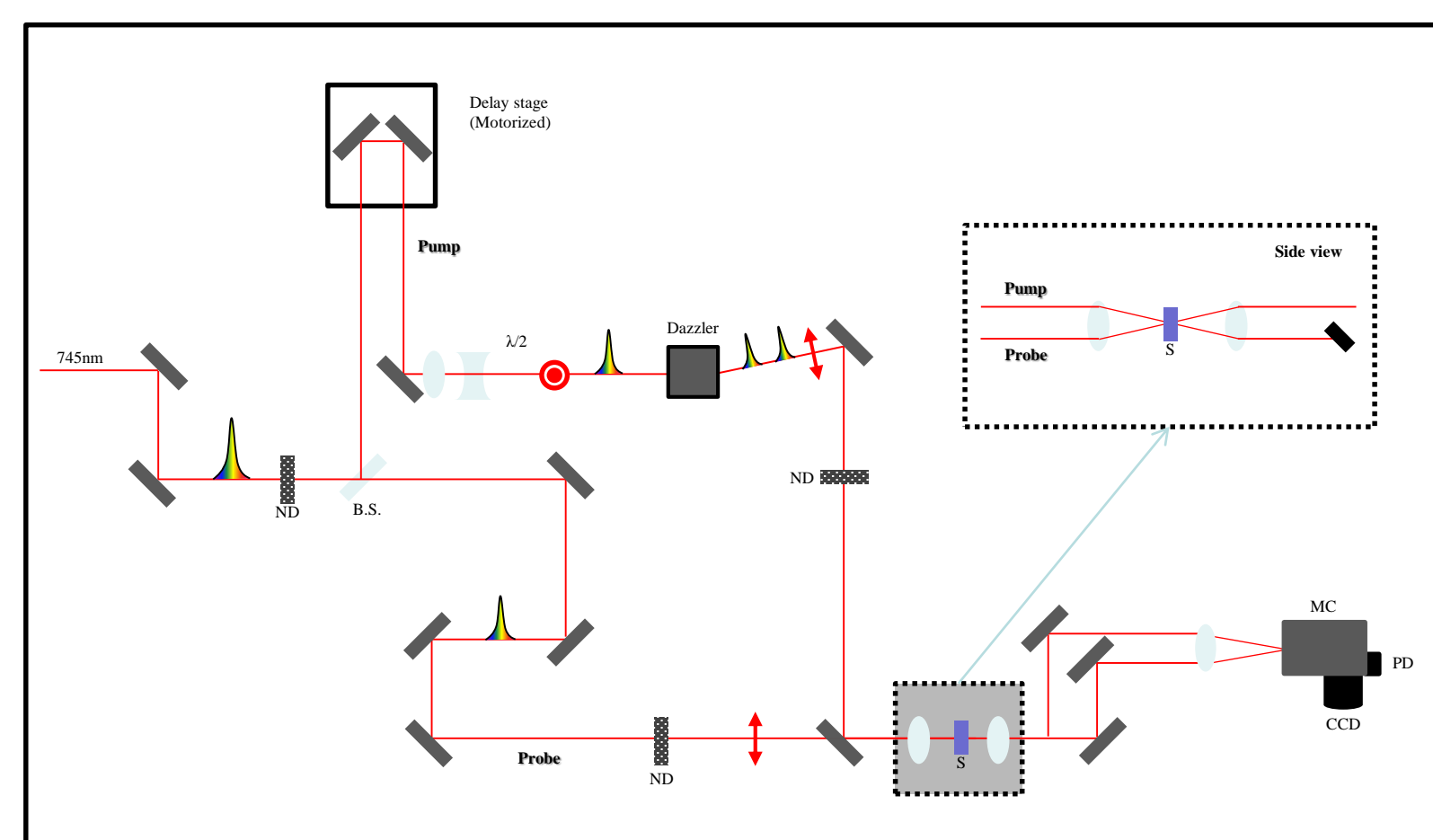
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Introduction

2D electronic spectroscopy



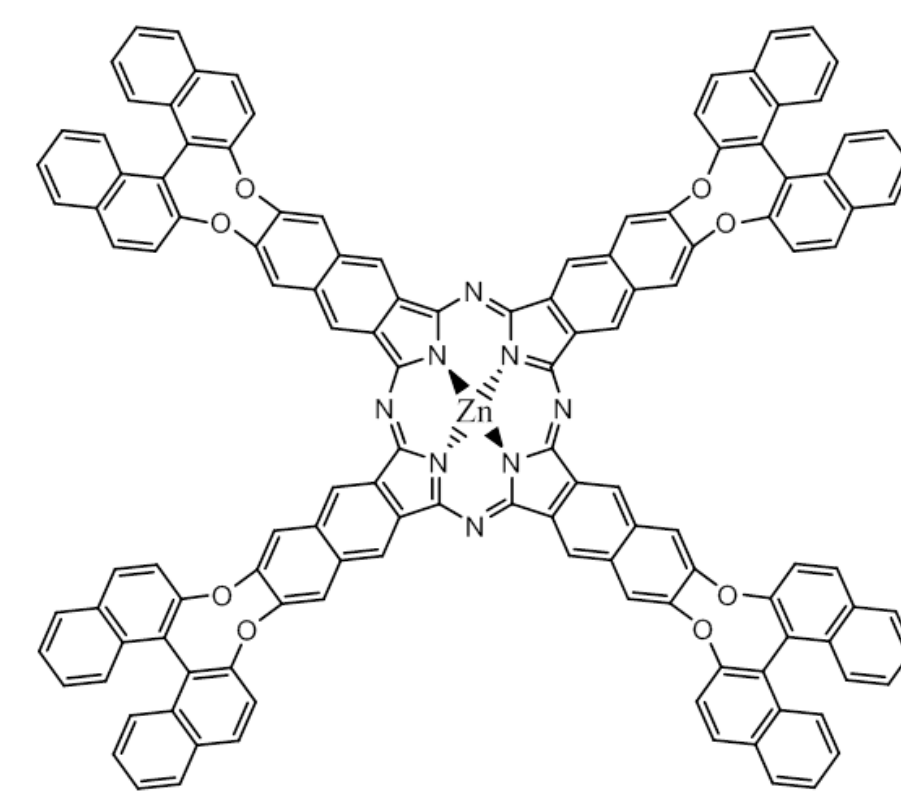
Simple scheme of 2D electronic spectroscopy system



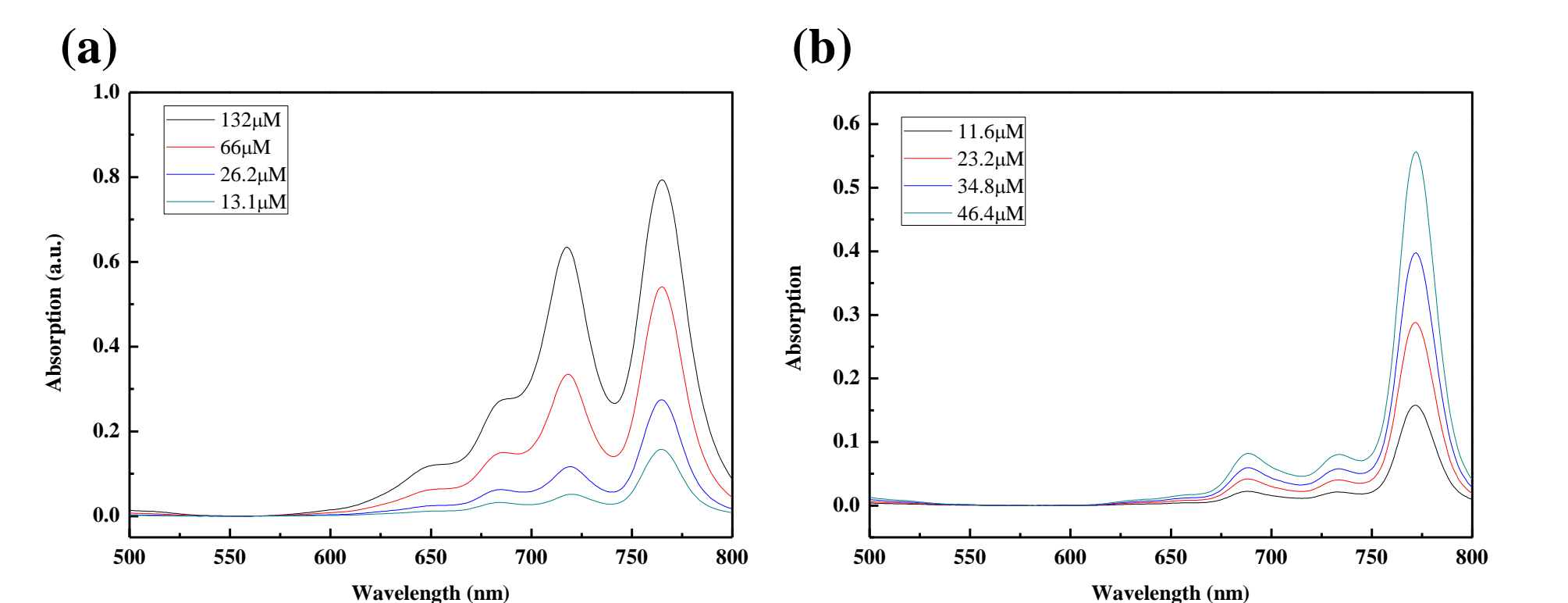
Experimental scheme of 2D electronic spectroscopy system

- Three femtosecond optical pulses are interacted with proper time delay and phase relationship to measure third order nonlinear signal.
- Two beams in pump probe geometry using optical pulse shaper generates two consecutive pump pulses with accurate phase information.

Molecular Information



Molecular structure of ZnNpc

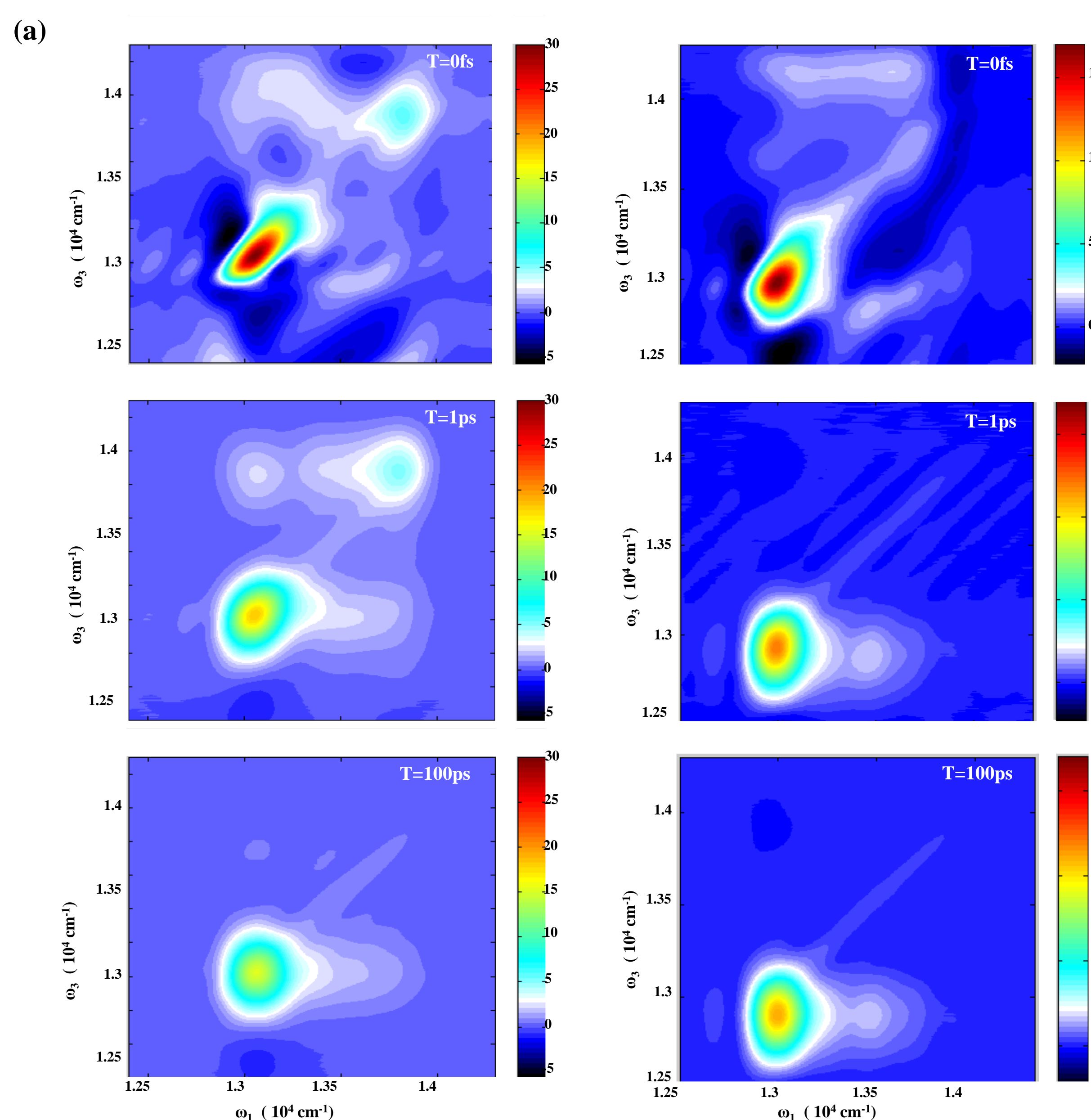


Absorption spectrum of ZnNpc dissolved (a) in THF and (b) in Benzonitrile.

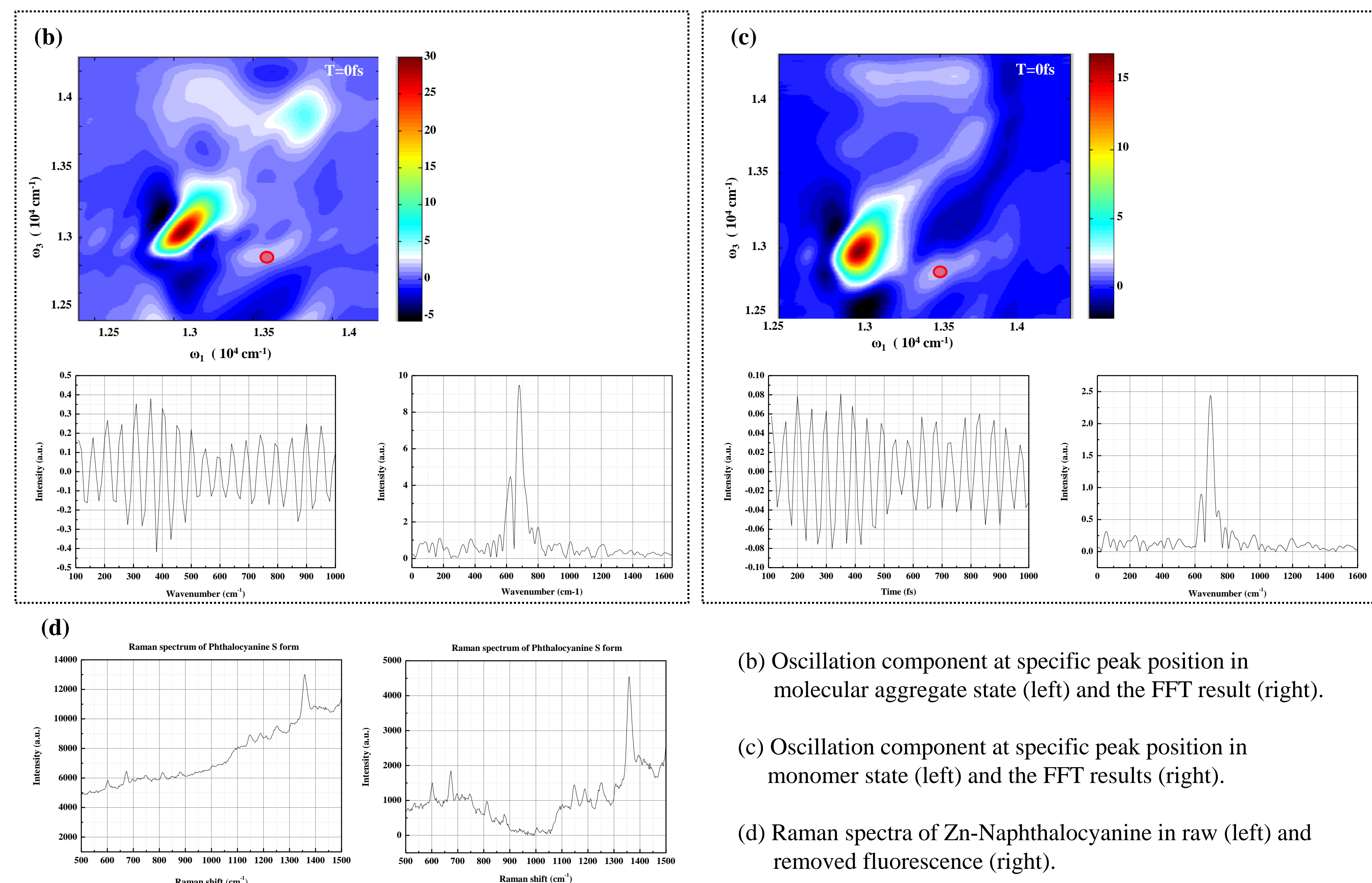
- For the time resolved 2D-ES experiment of ZnNpc in molecular aggregate, Concentration-dependent absorption experiment is conducted.
- Absorption spectra of micro-molar level concentration ZnNpc dissolved in THF and in Benzonitrile are investigated to comparative analysis for the basis of the 2D analysis.

Experiments Results & Discussion

2D electronic spectroscopic results



- Time-resolved 2D spectra of ZnNpc in THF (left) and Benzonitrile (right) are measured at T=0fs, 1ps and 100ps
- 2D spectra are elongated diagonally due to inhomogeneous broadening at T=0 and the inhomogeneity disappear in 100fs.



- In diagonal and cross peaks of the 2D-ES spectra, delicate oscillating signals which is correlated with the vibrational coherence are observed and plotted as a function of T using a hundred of spectrum.
- Slow beating modulation is conspicuously displayed from oscillating signal at cross peak position and their frequency difference of 80cm⁻¹, which is in keeping with result of the Raman spectrum in the ground state, is confirmed

Summary & Perspective

1. We finally confirmed that the origin of slow beating signals at various peaks correlated with the phase relationship of two different mode through analysis of the 2D spectrum in monomer state
2. This approach will be extended to 2D chiroptical spectroscopy in combination with the heterodyne-detected chiroptical method with precise control of optical pulse sequence and polarization state