CFEL SCIENCE

SEMINAR

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CFEL-bldg. 99, seminar room IV

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Emergent coherent dynamics in the relaxation of photo-excited molecules

The description of quantum systems interacting with their environment is key to the understanding of the dynamics of photo-excited molecules in condensed phase, ranging from the initial steps of vision to singlet fission. On the other hand, the theoretical description of such dissipative quantum dynamics is numerically challenging, in particular in regimes where inter-site and system-bath couplings of similar magnitude render the Born–Markov approximation invalid. I will introduce the recently developed, non-perturbative MACGIC-QUAPI method [1]. The method provides a rigorous framework for the investigation of a vast range of complex dynamics, ranging from initially coherent oscillatory dynamics to the incoherent trapping of population in low energy states where medium reorganization occurs on a comparable timescales to system dynamics [2].

Going beyond the Condon approximation, where a constant electronic coupling is assumed [3], I will demonstrate pronounced effects on the electronic decay. Numerical simulations reveal the existence of two distinct dynamical regimes: an ultrafast non-exponential initial decay is followed by relatively slower late-time decay that shows exponential dynamics. The findings allow to impose conditions where electronic decay exhibits coherent dynamics even at room temperature (Fig. 1), characterized by an oscillation frequency that increases with coupling strength. Such coherences are evidently different from the ones observed in the weak coupling regime.

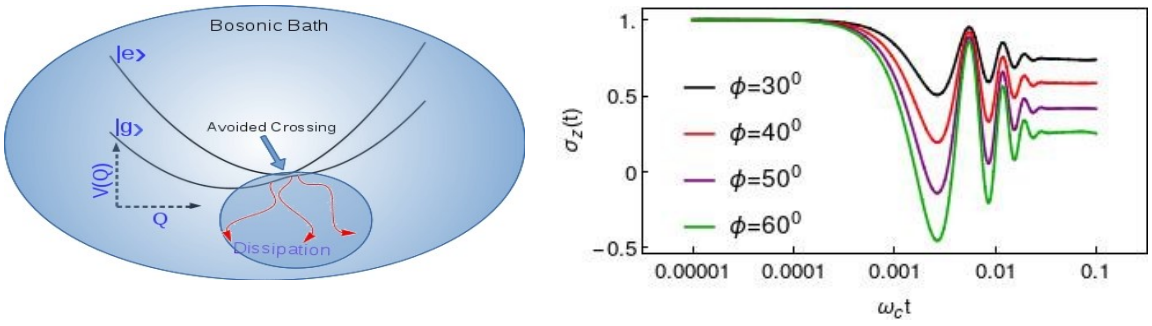

Fig 1: Schematic of system-bath **interaction** and the emergent oscillatory dynamics at strong coupling.

Host: Robin Santra – CFEL-DESY Theory Division