VISTA Seminar

Seminar 51

May 10, 2023

10:00 am – 11:30 am EST / 3:00 – 4:30 pm GMT London / 4:00 pm – 5:30 pm CET Paris / 11 pm CST Beijing

TOC:

Modeling the photoactivation of photoreceptor proteins: successes and challenges

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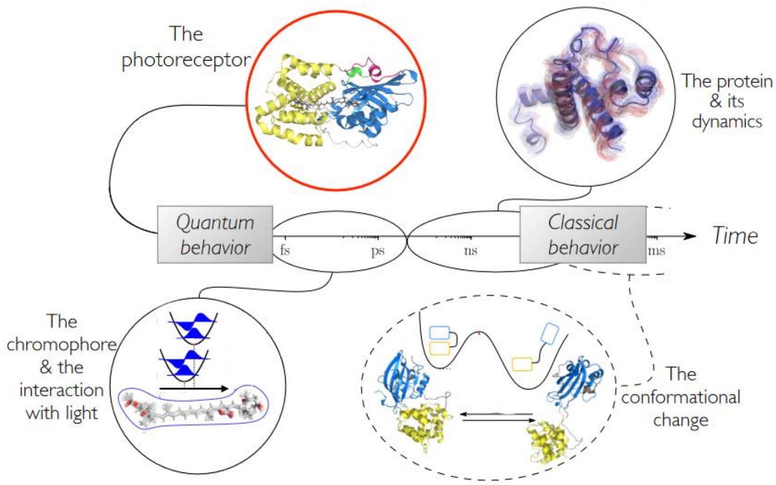

Photoreceptor proteins can respond to light using diverse mechanisms but most commonly they exploit an electronic excitation of a chromophore bound to the protein matrix. This initial excitation rapidly “travels” across space and time finally leading to a protein structural change. However, chromophores behave differently when they are embedded in a protein. The electrostatic fields in the protein cavity and the interactions with the nearby residues can largely affect their response to light. Moreover, their conformational space and vibrational motions can be strongly modified. A possible computational strategy to simulate the specific photophysics/photochemistry of chromophores within a protein is to couple quantum chemistry and classical models and integrate the resulting multiscale approach into a dynamic framework, but many theoretical and numerical critical issues must be faced. In this talk, these issues will be discussed, and possible solutions will be presented together with some applications.

Transport and Dispersion of Exciton-Polaritons in Multimode Cavities

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We investigate ballistic and diffusive transport of polaritons and develop a microscopic theory of polariton dispersion. We shed light on the ambiguity of using a (N+1)×(N+1) vs 2N×2N model for obtaining multimode polariton dispersion and show that ballistic coherent transport is achieved when coupling to cavity which suppresses decoherence due to phonons. We investigate the properties of exciton polaritons formed by coupling a multi-layered material and quantized cavity radiation. First, we develop a microscopic theory to obtain multimode polariton dispersion in materials coupled to cavity radiation modes. We device a general strategy for obtaining simple matrix models based on the structure and spatial location of multi-layered

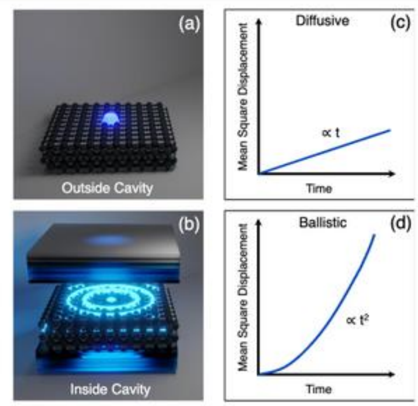

Fig. 1 Schematic illustration (a) diffusive transport in excitons and (b) ballistic motion in exciton-polariton and their time-dependent mean-square displacement in (c) and (d).

2D materials inside the optical cavity by using a rigorous light-matter nonrelativistic quantum electrodynamics (QED) Hamiltonian in the dipole Gauge beyond the usual long-wavelength approximation. We develop a generalized 2N × 2N model (where N is the number of energetically relevant cavity mode branches) that for specific geometries and spatial locations of materials reduces to (N + 2) × (N + 2) or the well-known 2N ×2N and (N + 1) × (N + 1) models. We find that while (N + 1) × (N+ 1) is appropriate for single layer materials, 2N × 2N should be used for filled cavities.

Second, we investigate the transport properties of exciton-polaritons using a mixed quantum-classical approach and non-Markovian master equation approach. Outside cavity, materials exhibit diffusive excitonic transport due to strong electron-phonon interactions which induces dynamic disorder. When placing materials inside an optical cavity, light-matter interactions can strongly modify transport properties by effectively modifying exciton-phonon interactions. We observe that exciting to a polaritonic branch with high photonic character leads to ballistic propagation with velocities that match the expected group velocity from polaritonic dispersion. This is because polariton branches with high photonic character couple weakly to the phonons thereby having a long decoherence time. When initial excitation is of higher excitonic character we observe ballistic motion with velocities that are smaller than expected group velocity from polaritonic dispersion. We show that this is due to a transient localization mechanism. Finally, we observe that initial excitation with significantly high excitonic character leads to diffusive transport. We find that the spatial dependence of the cavity field plays a crucial role in describing these effects and single cavity mode approximation is inadequate for describing polariton transport.

REFERENCES

[1] Xu, D., Mandal, A., Baxter, J. M., Cheng, S. W., Lee, I., Su, H., ... & Delor, M. (2022). Ultrafast imaging of coherent polariton propagation and interactions. arXiv preprint arXiv:2205.01176.
[2] Mandal, A., et. al (2022). Microscopic Theory of Multimode Polariton Dispersion in Multilayered Materials. arXiv preprint arXiv:2303.10815

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 51
Time: May 10, 2023 10:00 AM Eastern Time (US and Canada)

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