Theoretical Studies of Chemical Dynamics in Condensed and Biomolecular Systems

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2008 D.Sc. Kyoto University

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2006 JPSP Research Fellow, Kyoto University
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Awards
2015 10th Condensed-Matter Science Prize, Japan
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Photosynthesis provides the energy source for essentially all living things on Earth, and its functionality has been one of the most fascinating mysteries of life. Photosynthetic conversion of the energy of sunlight into its chemical form suitable for cellular processes involves a variety of physicochemical mechanisms. The conversion starts with the absorption of a photon of sunlight by one of the light-harvesting pigments, followed by transfer of electronic excitation energy to the reaction center, where charge separation is initiated. At low light intensities, surprisingly, the quantum efficiency of the transfer is near unity. A longstanding question in photosynthesis has been the following: How does light harvesting deliver such high efficiency in the presence of disordered and fluctuating dissipative environments? Why does not energy get lost? At high light intensities, on the other hand, the reaction center is protected by regulation mechanisms that lead to quenching of excess excitation energy in light harvesting proteins. The precise mechanisms of these initial steps of photosynthesis are not yet fully elucidated from the standpoint of molecular science. Particularly, recent observations of long-lived beating phenomena in two-dimensional electronic spectra of photosynthetic pigment-protein complexes stimulated a huge burst of activity in an interdisciplinary community of molecular science and quantum physics.

Selected Publications

- A. Ishizaki and Y. Tanimura, "Quantum Dynamics of System Strongly Coupled to Low-Temperature Colored Noise Bath: Reduced Hierarchy Equations Approach," *J. Phys. Soc. Jpn.* **74**, 3131–3134 (2005).
- A. Ishizaki and G. R. Fleming, "Unified Treatment of Quantum Coherent and Incoherent Hopping Dynamics in Electronic Energy Transfer: Reduced Hierarchy Equation Approach," *J. Chem. Phys.* **130**, 234111 (10 pages) (2009).
- A. Ishizaki and G. R. Fleming, "Theoretical Examination of Quantum Coherence in a Photosynthetic System at Physiological Temperature," *Proc. Natl. Acad. Sci. U.S.A.* **106**, 17255–17260 (2009).
- A. Ishizaki, T. R. Calhoun, G. S. Schlau-Cohen and G. R. Fleming, "Quantum Coherence and Its Interplay with Protein Environments in Photosynthetic Energy Transfer," *Phys. Chem. Chem. Phys.* **12**, 7319 (2010). [Invited perspective article]
- A. Ishizaki and G. R. Fleming, "Quantum Coherence in Photosynthetic Light Harvesting," *Annu. Rev. Condens. Matter Phys.* **3**, 333–361 (2012). [Invited review article]
- G. D. Scholes, G. R. Fleming, L. X. Chen, A. Aspuru-Guzik, A. Buchleitner, D. D. F. Coker, G. S. Engel, R. van Grondelle, A. Ishizaki, D. M. Jonas, J. S. Lundeen, J. K. McCusker, S. Mukamel, J. P. Ogilvie, A. Olaya-Castro, M. A. Ratner, F. C. Spano, K. B. Whaley and X. Y. Zhu, "Using Coherence to Enhance Function in Chemical and Biophysical Systems," *Nature* **543**, 647–656 (2017).

1. Using Coherence to Enhance Function in Chemical and Biophysical Systems

Coherence phenomena arise from interference, or the addition, of wave-like amplitudes with fixed phase differences. Although coherence has been shown to yield transformative ways for improving function, advances have been confined to pristine matter and coherence was considered fragile. However, recent evidence of coherence in chemical and biological systems suggests that the phenomena are robust and can survive in the face of disorder and noise. We surveyed the state of recent discoveries, present viewpoints that suggest that coherence can be used in complex chemical systems, and discuss the role of coherence as a design element in realizing function.[1)]

2. Modeling of High Frequency Vibrational Motion in Quantum Dynamics of Singlet Fission Process

Singlet fission is a spin-allowed energy conversion process whereby a singlet excitation splits into two spin-correlated triplet excitations residing on adjacent molecules and has a potential to dramatically increase the efficiency of organic photovoltaics. Recent time-resolved nonlinear spectra of pentacene derivatives have shown the importance of high frequency vibrational modes in efficient fission. In this work, we explored impacts of vibration-induced fluctuations on fission dynamics through quantum dynamics calculations with parameters from fitting measured linear and nonlinear spectra. We demonstrated that fission dynamics strongly depends on the frequency of the intramolecular vibrational mode. Furthermore, we examined the effect of two vibrational modes on fission dynamics. Inclusion of a second vibrational mode creates an additional fission channel even when its Huang-Rhys factor is relatively small. Addition of more vibrational modes may not enhance fission *per se*, but can dramatically affect the interplay between fission dynamics and the dominant vibrational mode.[2)]

3. Spin Hall Effect without Space Inversion Symmetry

A master equation approach based on an optimized polaron tranThe spin Hall effect (SHE), first proposed almost half a century ago, has been investigated and observed in a large variety of systems including electrons in semiconductors such as GaAs and In GaAs, photons passing through an interface with a refractive index gradient, and atomic Bose-Einstein condensates subject to synthetic gauge fields. In all of these studies, the SHE arises from the spin–orbit interaction which requires space inversion symmetry (SIS) breaking. In this work, we showed that the SHE can appear in a totally different but equally broad class of many-body systems with SIS. In particular, we demonstrated that the SHE can emerge from the dipole–dipole interaction (DDI), which preserves SIS. The DDI-based SHE is expected to occur in, for example, heteronuclear molecules, strongly magnetic atoms, Rydberg excitations, electron gases, etc. The SHE arising from the DDI offers a complementary tool to generate spin currents, which constitutes an essential ingredient in spintronics.[3)]

References

1) G. D. Scholes, G. R. Fleming, L. X. Chen, A. Aspuru-Guzik, A. Buchleitner, D. D. F. Coker, G. S. Engel, R. van Grondelle, A. Ishizaki, D. M. Jonas, J. S. Lundeen, J. K. McCusker, S. Mukamel, J. P. Ogilvie, A. Olaya-Castro, M. A. Ratner, F. C. Spano, K. B. Whaley and X. Y. Zhu, *Nature* **543**, 647–656 (2017).
2) Y. Fujihashi, L. Cheng, A. Ishizaki J. Wang and Y. Zhao, *J. Chem. Phys.* **146**, 044101 (11 pages) (2017).
3) T. P. Nguyen and M. Ueda, submitted.

Awards

ISHIZAKI, Akihito; 18th Sir Martin Wood Prize (2016).
ISHIZAKI, Akihito; Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology The Young Scientists' Prize (2017).