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JCP Emerging Investigator Special Collection 2019

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INTRODUCTION

The Journal of Chemical Physics is invested in recognizing the achievements of early career researchers. In 2019, we launched the first annual JCP Emerging Investigator Special Collection.[1] The collection is a highly selective showcase for excellent work supervised by principal investigators early in their careers (10 years or less since Ph.D. graduation). The collection spans the scope of topics in chemical physics and physical chemistry and encourages contributed papers. Of the excellent papers published in the collection, two are chosen by a subcommittee of the Editorial Advisory Board as winners of the JCP Best Paper by an Emerging Investigator Award. The 2019 winners are as follows:

Jeremy O. Richardson,
Assistant Professor of Theoretical Molecular Quantum Dynamics,
Laboratory of Physical Chemistry, ETH Zurich,
Photo credit: ETH Zurich/Giulia Marthaler

"Instanton formulation of Fermi's golden rule in the Marcus inverted regime,"[2] by Eric R. Heller and Jeremy O. Richardson

Sandeep Shama,
Assistant Professor,
Department of Chemistry, University of Colorado, Boulder

"Multireference configuration interaction and perturbation theory without reduced density matrices,"[3] by Ankit Mahajan, Nick S. Blunt, Iliya Sabzevari, and Sandeep Sharma

The 2019 collection was filled with many impressive contributions. As a service to the community, the JCP editors who handled the papers included in the 2019 collection have summarized them and highlighted their significance below, organized by topic.

ELECTRON TRANSFER

It is well known that quantum tunneling can enhance the rate of an electron transfer reaction by many orders of magnitude in the Marcus inverted regime. It is also well known that semiclassical instanton theory provides an elegant way to understand and compute tunneling rates. However, the instanton theory of electron transfer rates has not previously been extended into the inverted regime. In their award-winning paper, Heller and Richardson[2] show how to do this. They find that the instanton periodic orbit in the inverted regime involves two imaginary time trajectories, one of which travels in negative imaginary time. They also show that despite the computational difficulties that this poses, the instanton provides a practical way to calculate the rate in the inverted regime.

ELECTRONIC STRUCTURE

The accurate calculation of the electronic structure is especially difficult in large, strongly correlated systems. To treat such situations, a suite of so-called "multireference" techniques have been developed, but these approaches are costly and difficult to use for large systems. The award-winning paper by Sharma and co-workers[3] devises a stochastic means to circumvent a major bottleneck in two paradigmatic approaches, paving the way for a more facile treatment of large-scale strongly correlated problems.

A second paper in this area by Blunt[4] uses a hybrid subspace and stochastic approach to combine the best features of the selected configuration interaction (SCI) and full configuration interaction quantum Monte Carlo (FCI-QMC) methods. This leads to faster convergence of these promising approximations to FCI.

Less closely related but also involving stochastic techniques is the paper by Dornheim and co-workers,[5] who use path integral Monte Carlo calculations to train machine learning models that can describe the uniform electron gas under extreme conditions of temperature and pressure. This work will enable efficient and accurate calculations for matter in extreme conditions such as planet interiors.

The collection also contains some interesting contributions that focus on lower levels of electronic structure theory. Brémond and co-workers[6] provide new insight into the relationship between the fraction of exact exchange in a global hybrid density functional and the range-separation parameter in the analogous range-separated hybrid functional. This promises to simplify the construction of accurate range-separated hybrid functionals.

Brandenburg and co-workers[7] present a new benchmark set of high-quality energies, using coupled-cluster and QMC methods, for several ice systems and for the interaction between a water molecule and various carbon nanostructures. A wide array of density functional approximations is assessed against the benchmark values, and the results provide insight into the effectiveness of various recent efforts to improve the accuracy of density functional approximations.

In another development involving density functional theory, Mattiat and Luber[8] present a real-time time-dependent density

functional theory (TDDFT) implementation of resonant vibrational Raman optical activity. They show that the real-time propagation provides a computationally efficient approach for the simulation of Raman optical activity excitation profiles.

Moving beyond density functional theory, Klimeš and Tew[9] compare the random phase approximation (RPA) with second order Møller–Plesset perturbation theory (MP2) for adsorption energies in zeolites. They show that RPA with singles corrections is both more accurate and more efficient than MP2 for this purpose. This work provides a foundation for extensive use of RPA with singles corrections in porous systems.

LIGHT–MATTER INTERACTIONS

The field of strong light–matter interactions is becoming increasingly popular in chemical physics, and this collection contains two separate contributions on this topic.

Li and co-workers[10] present a theoretical study of light–matter interactions in designed nanostructures in which they convincingly demonstrate the feasibility of strong coupling between plasmon-induced magnetic resonance (PIMR) and propagating surface plasmon (PSP) modes at visible frequencies.

Hernández and Herrera[11] use a cavity quantum electrodynamics approach that goes beyond the rotating wave approximation to describe the light–matter interaction of an infrared cavity field with an anharmonic vibration of a non-polar molecule. Their key finding is that the bond length of a vibrational polariton at a given energy is never greater than that of the bare molecule at the same energy. This "polariton bond strengthening" effect might well have implications for the reactivity of vibrational polaritons in the emerging field of polariton chemistry.

PHOTOCHEMICAL REACTIONS

The fate of the first excited state of NO is important in its detection in the atmosphere and during combustion, where it is extensively studied using the laser-induced fluorescence technique. Kidwell and co-workers[12] report an experimental investigation of how this excited state of NO is quenched by collisions with O_2. Their isotropic velocity map ion images provide evidence for a long-lived NO–O_2 collision complex prior to product formation.

MACHINE LEARNING AND ENERGY LANDSCAPES

In addition to the paper by Dornheim and co-workers[5] that we have already mentioned, there is also a second interesting paper on machine learning in the collection. Ceriotti and co-workers[13] use machine learning techniques to analyze a very broad range of hypothetical zeolite structures and show that the smooth overlap of atomic positions (SOAP) method does much better than classical descriptors such as distances, angles, and ring sizes in accurately capturing the structure–property relationships predicted by atomistic force fields. This new approach to describing zeolitic structures may explain the synthesizability (or not) of this important class of materials.

Wang and co-workers[14] present another paper on energy landscapes, which focuses more on global optimization than on structure–property relationships. They show that their recently proposed unbiased "fuzzy global optimization" (FGO) algorithm can be used to identify candidate lowest-energy structures of large Lennard-Jones clusters with both high reliability and efficiency.

MOLECULAR SIMULATION

A number of contributions in the collection are concerned with the development and application of simulation methods to both molecular and biomolecular systems.

Das and Limmer[15] describe a variational method for computing the likelihood of a rare event within a non-equilibrium molecular dynamics simulation, relevant to the study of driven systems undergoing dynamical fluctuations. An application of the method to a system undergoing a dynamical phase transition, reflecting competing ballistic and diffusive modes of transport, demonstrates its accuracy and promise.

Cossio and co-workers[16] address the challenge of extracting equilibrium and dynamical information, such as free energies and transition rates, from trajectories under constant force. They demonstrate limitations of the promising committor inversion method when applied to barrier estimation that must be addressed to fully realize the promise of this method in the interpretation of single molecule experiments.

Tiwari and co-workers[17] revisit the classic problem of droplet nucleation in a simple fluid through the application of modern approaches to rare event dynamics. Surprisingly, they find that as supersaturation decreases, it becomes important to explicitly consider local density fluctuations, which correlate with shape and density variations in the nucleus, in the definition of the reaction coordinate.

Hoy and co-workers[18] explore the competition between crystallization and glass formation for a family of bent trimer molecules in which the bond angle is varied systematically. They find that the local cluster structure is a key predictor of whether a crystal or a glass is formed on cooling.

Savoie and co-workers[19] quantify the errors incurred in coarse-grained (CG) computer simulations of molecular liquids in terms of "representability errors" (arising from the limitations of the functional form of the CG model) and "information errors" (arising from the limited data that are used to parameterize the model). These concepts are illustrated with example CG simulations of butane, butanol, and 1,3-propanediol.

Whitford and co-workers[20] use all-atom molecular dynamics simulations to extract the diffusion coefficient characterizing tRNA motion along a specific reaction coordinate, observing an order-of-magnitude decrease in the diffusion coefficient for the process by which tRNA enters the ribosome. Considering the role of electrostatic and solvation interactions, they demonstrate how steric effects induce large length scale barriers, while short length scale roughness determines the rate of diffusive movement across the energy landscape.

SOFT MATTER

In addition to the simulation studies described above, the collection contains a number of studies that focus specifically on soft matter systems. Many of these are theoretical, but the last stands out in that it involves an experimental validation.

Recent studies of chiral active fluids have discovered anomalous transport properties, in particular the so-called odd viscosity. This has been explained in general terms as emerging from symmetry principles; however, a microscopic, molecular understanding has been lacking. The analysis in the paper by Vaikuntanathan and co-workers[21] provides this missing microscopic understanding. This work will be of great importance to the non-equilibrium statistical mechanics community as it provides an illustrative example of how transport can be controlled and modulated far from equilibrium.

Another paper on transport is that of Palmer and co-workers,[22] who study the transport of small tracer molecules in media formed by colloidal particles in a glassy state obtained by supercooling. They show that the tracer transport in these media is influenced by the structure of the supercooled liquid. Depending on the interactions between the colloidal particles that form the supercooled matrix, one may obtain quite different dynamic behavior for the tracer. This study may provide some hints as to the mechanism of the delivery of drug molecules encapsulated in hydrogels or the movement of DNA in crowded media.

Finally, there are two papers in the collection by the same group, one theoretical and the other experimental. In their theoretical paper, Yurchenko and co-workers[23] analyze the many-body interactions in colloidal suspensions that are subjected to external rotating electric or magnetic fields. Their robust and systematic approach to these tunable interactions will make the manuscript useful to future researchers working in both molecular and colloidal physics. In their experimental paper,[24] the authors use a recently developed interpolation method to successfully extract the interaction potential between two particles from the observed structures of colloidal crystals and dusty plasmas, thereby confirming the applicability of this method to a variety of crystalline systems.

2020 COLLECTION AND AWARDS

The 2020 Emerging Investigator Collection is open for submissions and already publishing. The deadline for acceptance into the 2020 collection is December 31, 2020. Papers accepted after that date will be included in the 2021 collection and considered for the 2021 awards. For more information, please see the journal website: https://aip.scitation.org/jcp/info/awards. To go directly to how to submit and eligibility requirements, please see the call for paper page: https://publishing.aip.org/publications/journals/special-topics/jcp/2020-jcp-emerging-investigators-special-collection/.

We look forward to many more annual collections recognizing early career researchers. If you have any questions or comments, please contact us at jcp-journalmanager@aip.org.

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