

Physical Chemistry Seminar

Embrace the darkness From singlet fission to exciton-polaritons

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Polaritons are increasingly touted as a promising tool to ‘rewrite’ the functional behavior of molecular systems. Polaritons are mixed states formed from the hybridization of molecular transition dipoles with a confined electromagnetic field. Originally the purview of ultra-cold physics, when this concept is applied to molecular vibrational or electronic absorption transitions polaritons can be attained at room temperature. Studies over the last decade have revealed a host of weird and wonderful effects that result, from enhanced energy transport and charge carrier mobility to changes in the selectivity of chemical reactions – all by simply enclosing the materials between a pair of mirrors. Yet, while the field has become better and better at identifying exciting polaritonic phenomena, we lack a fundamental understanding of their underlying mechanisms. The temptation is strong to explain their exotic behavior in terms of the bright, strongly coupled states that we can easily observe. However, the bright polariton states are not alone. When we peer into optical cavities with ultrafast spectroscopy, we see that they are accompanied by a host of dark states that can dominate the photophysical response. An improved model, then, frames their photophysics in terms of an interplay between bright and dark states. But to make matters worse, we find that most systems studied today don’t even fit this neat bright-dark dichotomy. The ‘grey’ states in these materials mix the properties of both manifolds, whether due to disorder or higher-order state couplings that are frequently overlooked. Our results from simple molecular dimers to complex thin-film microcavities force us to reevaluate our basic pictures of molecular photophysics and present new opportunities for materials design, from the optical generation of entangled spins to polaritonic structures with orders-of-magnitude enhanced donor-acceptor transfer.



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10:30am



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James Tarpo Jr. and Margaret Tarpo
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Musser Bio

Andrew originally trained as a physicist with an eye towards solving global problems. Not infrequently accused of being a spy, he spent two years working and studying in Russia and tried his hand at organic synthesis at the Max-Planck Institute in Mainz, Germany, and genetic engineering at the Manchester Interdisciplinary Biocentre in the UK. He fully returned to science in 2008, enrolling in the Nanoscience master's programme at the Zernike Institute for Advanced Materials, where he worked with Prof. Andreas Herrmann on the synthesis and characterisation of DNA-organic hybrid materials for drug delivery, sensing, and bioelectronics. Equipped with this understanding of how to design and make functional materials, Andrew turned to the main task for his PhD: saving the world with green energy (pending). In 2010 he joined the Optoelectronics group at the Cavendish Laboratory (Cambridge, UK), under Prof. Sir Richard Friend. Working in the laser lab during his PhD and a subsequent postdoc, he used ultrafast spectroscopy to unravel the mechanism of singlet fission – this 2-for-1 deal for charge carrier generation has the potential to revolutionise the solar energy sector if we can just work out how to harness it. In 2016 Andrew moved to the University of Sheffield Department of Physics for a postdoc with Prof. David Lidzey to study organic exciton-polaritons and their impact on molecular spin physics.

In summer 2019 Andrew finally found his natural home as a physical chemist, joining the faculty of the Department of Chemistry and Chemical Biology at Cornell. His group weaves together the many strands of his past research, exploiting light-matter interactions in complex materials with an emphasis on organic exciton-polaritons, ultrafast vibronic dynamics, and electron-primed photocatalysis. His work has been recognized with a DOE Early Career Award and a Sloan Research Foundation Fellowship.