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Management Strategies for the Nanoscale

Managing light is emerging as one of the crucial issues in creating efficient photovoltaic cells for solar energy conversion, as well as for optimizing optical devices such as light-emitting diodes. The different effects that have to be considered are coupling light into and out of the devices and how to enhance absorption or emission of light by the components of the device. The Perspective article by Fan and co-workers in this issue of *The Journal of Physical Chemistry Letters* provides a detailed review of recent research in this area of science.¹ The topics covered include calculations of the electric fields around nanostructures in photovoltaic devices, the use of whispering gallery modes to increase interaction lengths,² using plasmonic nanostructures to enhance absorption and/or emission,^{3,4} and designing nanostructured coatings to minimize surface reflections.⁵ This area of research requires a strong connection between theory and experiment. Photovoltaic devices often have many components, and how the components are arranged and the distances between the different components can affect their performance.⁶ Thus, theory is typically needed to design the device, and once the device has been made and tested, more detailed calculations are needed to understand what it all means.¹ As demonstrated by Fan and co-workers, when this comes together, the results can lead to very high impact publications.¹

In many cases, the calculations that are required are “simply” using Maxwell’s equations to simulate the fields in the devices. This can be done using a variety of standard approaches, such as finite element calculations or by using codes based on the discrete dipole approximation.^{7,8} However, there are some problems in this research field that are not straightforward applications of Maxwell’s equations. One such topic is how the electromagnetic fields at metal surfaces (“surface plasmons”) interact with molecular systems.⁹ The problem here is that the surface plasmons are typically treated using classical physics, while accurately describing processes such as absorption and emission of light in molecules, or electron transfer to and from the surface requires quantum mechanics. Meshing these two together, particularly for the strong fields at plasmonic surfaces, is a difficult problem.

Progress in this area of research has been made in a variety of ways. For example, Masiello and co-workers have developed a many-body Green’s function formalism to describe how the electronic structure of molecules changes in the presence of the field at a metal surface.¹⁰ This approach is important for understanding surface-enhanced spectroscopies, such as surface-enhanced Raman spectroscopy (SERS),¹¹ as well as for the device applications discussed by Fan and co-workers. Although at first glance SERS may not seem to be related to photovoltaic devices, a crucial issue in SERS is the distribution of the electromagnetic fields around nanostructures, the so-called SERS “hot spots”, which is one of the major issues addressed in the Perspectives by Fan and co-workers.¹ Recently, a number of clever experimental approaches have been developed for mapping these fields at high spatial resolution. Examples include cathode luminescence and/or electron energy loss

measurements^{12–14} and super-resolution SERS imaging using single molecules.¹⁵ These experiments are providing new and detailed information about the fields around metal nanostructures, and it is certainly possible that a similar level of detail could be obtained for the fields in photovoltaic devices.

Another way to attack the problem of coupling between plasmons and molecular resonances is to treat the molecular system semiclassically.¹⁶ This approach has been used to explain experiments where molecular fluorescence is used to measure the propagation lengths of surface plasmon polaritons in metal nanowires.^{17,18} While these studies represent fundamental physical chemistry research, the plasmonic fields at metal surfaces may potentially be useful for confining and guiding light in photovoltaic devices.^{19,20} Thus, this is an area of research that brings together researchers in applied physics, experimental and theoretical physical chemistry, as well as materials science.

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Notes

Views expressed in this Editorial are those of the author and not necessarily the views of the ACS.

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