Faraday Discussions Vol 163

Photo-initiated Quantum Molecular Dynamics

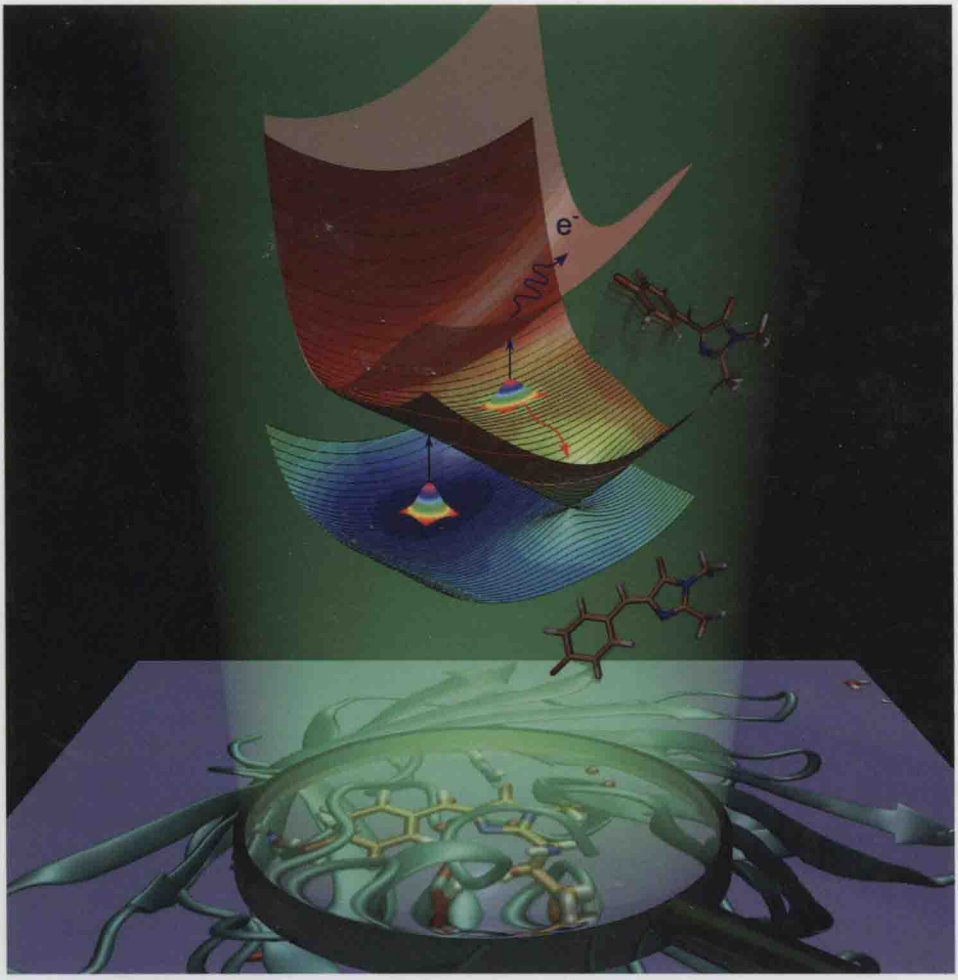

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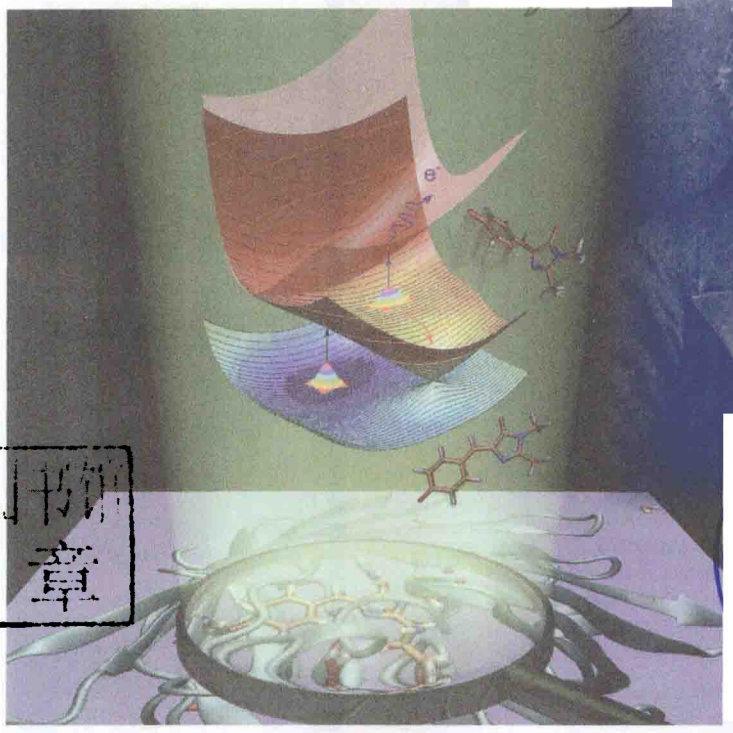
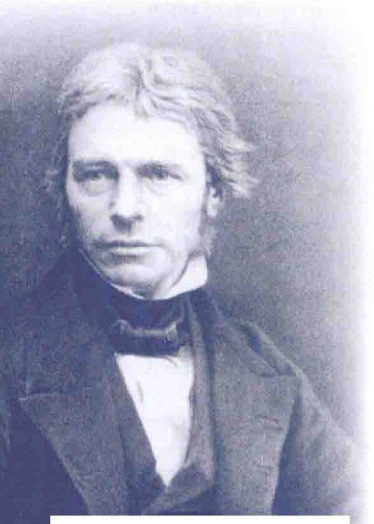
Photo-initiated Quantum Molecular Dynamics

University of Nottingham, UK
15–17 April 2013





馆藏章

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Photo-initiated Quantum Molecular Dynamics

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A General Discussion on Photo-initiated Quantum Molecular Dynamics was held in Nottingham, UK on 15th, 16th and 17th April 2013.

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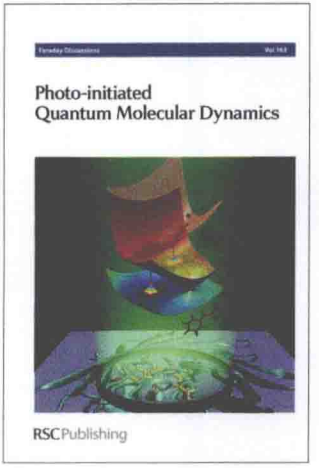

Cover

See Bochenkova and Andersen, *Faraday Discuss.*, 2013, **163**, 297–319.

When taken out of the Green Fluorescent Protein, its anionic light-absorbing molecular unit shows a remarkable fundamental interplay between electronic and nuclear excited-state decay channels in the ultrafast photo-initiated dynamics occurring on a pile of potential energy surfaces.

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INTRODUCTORY LECTURE

PAPERS AND DISCUSSIONS

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PAPER

The three pillars of photo-initiated quantum molecular dynamics

Albert Stolow*

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Studies of the interaction of light with matter, based on spectroscopy and diffraction, yielded a microscopic view of nature. The detailed structure of molecules and solids emerged from this effort and led to many of the advances of the 20th century, with the 'structure-function' paradigm being perhaps paramount. This static view of Nature is a necessary but insufficient advance and, in the 21st century, we will need to develop microscopic yet dynamical views of Nature. We consider here issues in Photo-initiated Quantum Molecular Dynamics raised by Faraday Discussions 163, in the context of three main categories or 'pillars' of light-matter interaction: energy/time, phase/coherence, intensity.

1 Introduction

The subject of Faraday Discussion 163 is 'Photo-initiated Quantum Molecular Dynamics'. The topics under discussion range very broadly, from high resolution spectroscopy,[1,2] to ultrafast dynamics[3–9] and coherence[10,11] quantum control[12–15] and strong field attosecond physics.[16–18] Yet underlying this seemingly disparate collection of topics is the unifying theme of light-matter interactions. Why is this an important subject? One hundred years ago, it was studies of the interaction of light with matter which led scientists to a microscopic view of nature. It was spectroscopy and diffraction, combined with quantum mechanics, which led us to understand the detailed structure of matter—the shapes of molecules, biomolecules and solids. It is hard to overstate how significant a step this was. One might fairly call it the 'great leap forward' of the 20th century. From the point of view of molecular sciences, this led to the central paradigm of 'structure-function relationships', the basis of much of our understanding of chemistry, biochemistry and material science. The canonical example is the structure of double helix DNA, the shape of which clearly determines its function as the template for the storage and transfer of genetic information. As powerful and successful as 'structure-function relations' may be, they remain a necessary but insufficient condition for our understanding of the world around and within us.

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Simply put, this is because Nature is not static. There are dynamical, often fast competing processes which determine the outcomes of many natural phenomena and these cannot be discerned from a static, structural perspective alone.

It is perhaps useful to consider an example. A compelling case is the process of Vision which involves the light-sensitive *trans*-membrane protein rhodopsin, comprised of an opsin protein 'shell' surrounding a photoactive chromophore. The chromophore is the molecular sub-unit retinal which contains a polyene chain that acts, in essence, as the 'light collector'. The first step in Vision, following absorption of a visible photon, is the *cis-trans* isomerisation of *11-cis* retinal about the C11–12 double bond and occurs on the 10^{-13} s time scale.[19] Given that the overall visual response is much slower (10^{-2} s), one could reasonably ask whether this ultrafast time scale is a curiosity or, in fact, is essential for the process of Vision. I argue that it is essential and that if this first step was much slower, Vision would be impossible. The rapid *cis-trans* isomerisation stresses and 'deforms' the *trans*-membrane opsin protein, permitting the now 're-shaped' cytoplasmic side of the protein to catalyze many hundreds of transducin proteins to an activated state.[20] This leads to a closing of cationic *trans*-membrane transport channels, hyperpolarizing the cell membrane and affecting a nearby synaptic terminal, thereby converting visual information to an electrical signal. However, the retinal chromophore is surrounded both by protein and by water—a highly dissipative environment. Dissipative processes are typically fast (ps time scale) and eventually lead to the conversion of photon energy to a simple, local temperature increase. In such a case, the energy associated with photon absorption would not lead to 're-shaping' of the protein: there would be no 'signal transduction'. Given that these fast dissipative mechanisms do exist, Nature was left with only one choice: the signal transduction pathway must be much faster than dissipation in order to compete. The first steps in Vision must be ultrafast, otherwise it would be impossible. We can see that a purely static 'structure-function' perspective would not lead to any understanding of the ultrafast functioning of the rhodopsin protein. In this example, and in many others, dynamics is central to the function. I suggest that

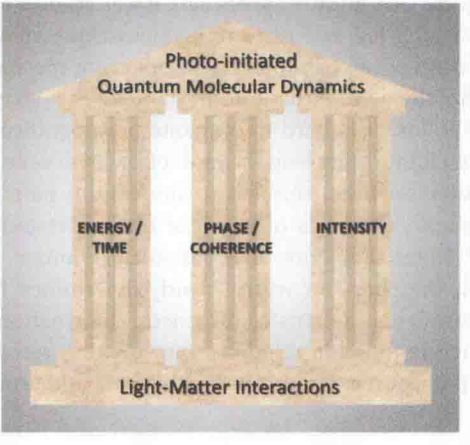

Fig. 1 The three pillars of photo-initiated quantum molecular dynamics. The light-matter interactions which underlie much of this subject can be broadly categorized into these three themes.

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