Visualization of Energy Conversion Processes in a Light Harvesting Organelle at Atomic Detail

Melih Sener*†, John E. Stone*, Angela Barragan*, Abhishek Singharoy*, Ivan Teo*†, Kirby L. Vandivort*, Barry Isralewitz*, Bo Liu*, Boon Chong Goh*†, James C. Phillips*, Lena F. Kourkoutis‡§, C. Neil Hunter¶, and Klaus Schulten*†
*Beckman Institute, University of Illinois at Urbana-Champaign
†Department of Physics, University of Illinois at Urbana-Champaign
‡School of Applied and Engineering Physics, Cornell University
§Kavli Institute at Cornell for Nanoscale Sciences, Ithaca, New York
¶Department of Molecular Biology and Biotechnology, University of Sheffield

***Abstract*—The cellular process responsible for providing energy for most life on Earth, namely, photosynthetic light-harvesting, requires the cooperation of hundreds of proteins across an organelle, involving length and time scales spanning several orders of magnitude over quantum and classical regimes. Simulation and visualization of this fundamental energy conversion process pose many unique methodological and computational challenges. We present, in an accompanying movie, light-harvesting in the photosynthetic apparatus found in purple bacteria, the so-called chromatophore. The movie is the culmination of three decades of modeling efforts, featuring the collaboration of theoretical, experimental, and computational scientists. We describe the techniques that were used to build, simulate, analyze, and visualize the structures shown in the movie, and we highlight cases where scientific needs spurred the development of new parallel algorithms that efficiently harness GPU accelerators and petascale computers.**

I. Introduction

Solar energy, directly or indirectly, powers almost all life on Earth through photosynthesis. Nature has evolved devices that utilize quantum mechanics for conversion of light energy into stable chemical energy, which can be stored and consumed by living cells to power physiological processes [1]. Studies of natural light-harvesting systems provide insights for the engineering of bio-hybrid and artificial solar devices and may contribute substantially to the current 15 TW power demand of our civilization by harvesting better Earth's 120,000 TW average solar irradiance [2].

Light-harvesting in biological cells poses substantial computational challenges, since it involves many overlapping time and length scales, ranging from electronic excitation transfer on a picosecond timescale [3] to diffusion events on a millisecond timescale [4]. In the case of purple photosynthetic bacteria, the photosynthetic apparatus is the so-called chromatophore, now structurally known at atomic detail [4], [5]; the chromatophore is a spherical membrane of 50 nm inner diameter, comprising over a hundred proteins and $\sim$3000 bacteriochlorophylls for photon absorption [6] (see Fig. 1). The study of the chromatophore and its component proteins has been the subject of simulation efforts for three decades, driving software development and new methodologies in collaboration with experimental studies (see sec. III and Table I below).

In an accompanying movie, all the primary energy conversion events in the chromatophore, from light absorption to ATP synthesis, are shown in a contiguous structural narrative that seamlessly connects cell-scale organization to atomic-scale function. The structural models and supra-molecular organization shown in the movie were experimentally determined by atomic force microscopy, cryo-electron microscopy, electron tomography, crystallography, optical spectroscopy, mass spectroscopy, and proteomics data (see [4] and references therein). The movie represents molecular dynamics (MD) simulation and modeling of the entire chromatophore as well as of its constituent protein complexes made possible by petascale systems such as Blue Waters, demonstrating, for the first time, the complete physiological sequence of a basic photosynthetic apparatus.

In the following, first, the energy conversion processes in the chromatophore are outlined as illustrated in the movie; second, the computational challenges and representative milestones for modeling the chromatophore function are recounted; last, the visualization techniques are discussed that enable the rendering of protein function across multiple scales.

II. Visualization of Chromatophore Function

The accompanying movie presents a narrative of the photosynthetic function of the chromatophore of purple bacteria as a clockwork of interlocked processes for the purpose of energy utilization, culminating in ATP synthesis. Purple bacteria experience, in their habitat, often low illumination levels, such as $\sim$1% of full sunlight, and have developed an adaptation to light starvation by overpopulating the cell interior with chromatophores (Fig. 1G). The primary step of light-absorption is carried out by pigments (bacteriochlorophylls and carotenoids) contained in the light harvesting complexes 1 and 2 (LH1 and LH2) that form a network delivering the absorbed light energy in the form of electronic excitation energy to the reaction center (RC) [7]. The RC initiates a sequence of charge transfer events, ultimately transferring two electrons and two protons to a mobile charge carrier, quinone, converting it to quinol. The quinol diffuses through the chromatophore membrane to the bc_1-complex, which processes quinols for generating a proton gradient across the membrane [8]. The soluble charge carrier cytochrome c_2 returns electrons back from the bc_1-complex to the RC, thereby completing a circuit. The proton gradient drives ATP synthesis at the ATP synthase [9]. (See Fig. 1H for a schematic of the energy conversion processes.)

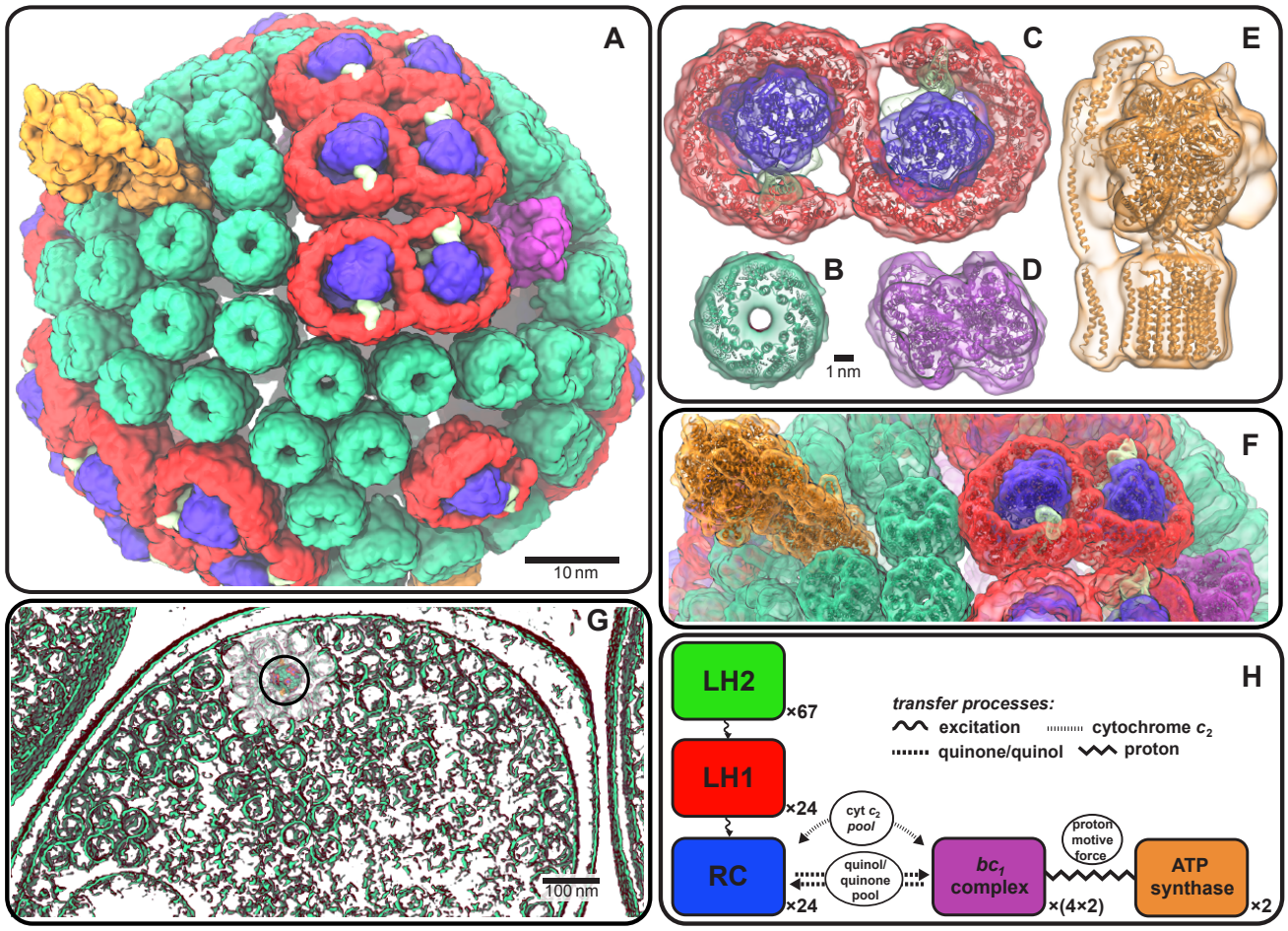

Fig. 1. The chromatophore from the purple bacterium *Rba. sphaeroides* (A) harvests solar energy for ATP production via a network of hundreds of cooperating proteins [4] (see accompanying movie). In order of energy utilization, these proteins are: the LH2 (B) and LH1-RC-PufX (C) complexes for light capture, electronic excitation transfer, and charge separation; the bc_1 complex (D) for generation of a proton gradient across the vesicle membrane; and the ATP synthase (E) for using the resultant proton gradient for ATP production (B, C, D: top view, i.e. normal to the membrane; E: side view). A minimal subunit of the four aforementioned primary protein groups is shown in (F) (proteins in cartoon representation; same colors as in B-E). The chromatophores densely populate the cell under low-light conditions, occasionally forming vesicular connections with one another (G) (the chromatophore shown in (A) is indicated by a circle). The energy conversion processes are summarized schematically in (H) (see sec. II for details).

III. Computational Challenges

The simulation of chromatophore components shown in the movie has been the subject of nearly 40 publications by the senior author over the last three decades [6] (see Table I), driving development efforts for simulation and visualization software packages NAMD [10] and VMD [11]. The chromatophore model shown in Fig. 1A was developed using VMD [12] by combining crystallographic structures of constituent proteins with supramolecular organization data [4]. Molecular dynamics (MD) simulation of a spherical chromatophore as shown in Fig. 1A contains approximately 100 million atoms, including lipids and water. A lamellar chromatophore patch containing 20 million atoms has already been simulated [13] determining excitation transfer properties of the pigment network. Current simulation efforts involve chromatophore models that contain not only the primary light harvesting complexes (LH2 and LH1-RC), but also bc_1 and ATP synthase complexes [4] responsible for energy conversion, as well as diffusible charge carriers [13].

A 100 million-atom NAMD simulation was specified by the NSF initially in 2006 as a petascale target application, and later the chromatophore (based on a model comprising only LH2 and LH1-RC complexes [5]) specifically was made an acceptance test for the Blue Waters supercomputer, launching a multi-year effort both to prepare the model and to improve NAMD performance and parallel scaling. The extreme size of the chromatophore structure to be modeled required the development of new molecular structure and trajectory file formats, new model building tools and algorithms, techniques for parallelization of existing analysis scripts, and new visualization techniques and rendering approaches within VMD [14]–[16]. At SC11 Mei et al. [17] showed the utility of memory usage optimization and multithreading in NAMD to scale 100 million atoms to the full Jaguar PF Cray XT5 (224,076 cores). Building on that work, at SC12 Sun et al. [18] explored techniques for improving fine-grained communication on the Cray Gemini network, demonstrating scaling to all 300,000 cores of ORNL Titan (before GPUs were installed on all nodes). Finally, at SC14 Phillips et al. [19] now describe the mapping of NAMD algorithms and data structures to toroidal network topologies, efficiently scaling a 224-million-atom system to 16,384 nodes of both Blue Waters and ORNL Titan (with GPUs).

TABLE I. REPRESENTATIVE COMPUTATIONAL MILESTONES. (See reviews [3], [6] for a comprehensive list.)

system & computational achievement	*publication (1st author)*
magnetic field dependence of primary photochemical reactions in the RC	Werner, 1978 [20]
time varying electrostatic properties of the RC	Treutlein, 1988 [21]
structure solution of the LH2 complex	Koepke, 1996 [22]
excitation transfer in a photosynthetic unit consisting of LH2 & LH1-RC complexes	Ritz, 1998 [23]
dynamic disorder and thermal effects on excitonic properties in LH2 based on polaron models	Damjanović, 2002 [24]
structural modeling and excitonic properties of a chromatophore containing LH2 & LH1-RC complexes	Sener, 2007 [5]
open quantum dynamics and thermal disorder in LH1 and LH2 based on hierarchy equations	Strümpfer, 2012 [25]
simulation of a 20M atom lamellar chromatophore	Chandler, 2014 [13]
integration of energy and electron transfer processes across a spherical chromatophore	Cartron, 2014 [4]

IV. VISUALIZATION TECHNIQUES

All of the molecular structure and cellular tomography renderings in the movie were produced by VMD [11], using combinations of many common molecular graphics structural representation techniques such as ball-stick, ribbon, secondary structure, and surface representations, but on a much larger-scale biomolecular complex than is typically visualized by such methods [14]–[16]. The fast GPU-accelerated "QuickSurf" molecular surface representation in VMD was used extensively throughout the movie, enabling emphasis to be placed on overall chromatophore architecture or on atomic detail as needed in different contexts [26].

The movie is composed of a series of short clips and transitions that were produced using the VMD ViewChangeRender plugin, hereafter referred to as "VCR". The VCR plugin allows researchers to create a list of visualization "viewpoints" that combine the camera view orientation, the molecular representations and their settings, and the associated simulation trajectory frame or other time series data. Individual movie clips are generated using previously defined viewpoints and associated transition times that allow the VCR plugin to interpolate viewing orientations, graphical representation properties, and simulation timestep indices as it renders individual frames. The VCR plugin provided the movie team with live dry-run movie visualization using the VMD OpenGL renderer, and final production rendering was performed using the built-in GPU-accelerated ray tracing engine described below.

The VCR plugin was significantly modified beyond its original capabilities to enable a large team of researchers to work together to create movie content, render movie clips, and assemble clips into a complete movie for final editing. Support for the script-driven definition of movie clips and complex scene transitions was added, along with export of a human-readable annotated "edit list" for use in the final video editing process. The VCR plugin was also extended to allow rendering of large groups of clips and to support batch mode parallel rendering on petascale computing systems.

Historically, MD simulation trajectories were generated at supercomputer centers and then transferred to the researcher's home lab, where they were ultimately analyzed. Petascale MD simulations generate terabytes of output that must be analyzed. The sheer size of such simulation trajectories is creating a shift in visualization and analysis practice, leading us to adapt our software to perform analytical and visualization tasks on the supercomputer where the data is generated, thereby avoiding days or weeks of off-site data transfer [12], [15], [16]. Movie rendering was performed using a new GPU-accelerated variant of the Tachyon ray tracing engine that is built into VMD [15], [16], [27]. Embedded ray tracing engines allow VMD to efficiently render molecular geometry in-situ without disk I/O, while enabling the use of high quality rendering techniques such as shadow filtering and ambient occlusion lighting [16].

All of the VMD parallel rendering work required for production of the movie was performed on Blue Waters, using the Cray XK7 GPU-accelerated compute nodes which are operating in a fully graphics-enabled "GPU operation mode", allowing the use of OpenGL for rapid turnaround of preview visualizations [28], and the built-in Tachyon/OptiX GPU ray tracing engine for production renderings [16], [29]. Movie frames were rendered in 16:9 aspect ratio at HD 1920×1080 resolution, with 12 antialiasing samples per pixel, 144 ambient occlusion shadow feeler rays per pixel, with direct lighting contributions and shadows from two directional lights; transmission rays and shadow filtering were performed for transparent geometry. One complete parallel rendering of the movie frames using 96 GPU-accelerated Cray XK7 compute nodes consumed $\sim$290 node hours with a wall-clock turnaround time of 3 hours, and produced $\sim$7,500 frames which used $\sim$45 GB of disk space.

Non-linear video editing was employed to compose the complete set of rendered VMD movie clips with hand-drawn figures, transitions, captions and annotations, and other materials. Cross-fade scene transitions were designed in the VMD VCR plugin using the live dry-run visualization mode, but rather than being rendered by brute force within VMD itself, the "edit list" exported by the VMD VCR plugin was used to guide generation of the transitions within the video editing software. Several clips in the movie utilize animated sequences produced within Final Cut Pro and Motion 5 to illustrate excitation migration between pigment clusters in modified Förster formalism, involving rapid delocalization and thermal equilibration of excitonic states within one LH-protein prior to transfer to neighboring proteins [3], [5], [25]. The animated illustrations were composited with VMD imagery through the use of multiple renderings, transparency, and depth layering. The final movie was exported in both 16:9 and cropped 4:3 aspect ratios for presentation on diverse display hardware.

V. CONCLUSIONS

A complete description of the energy harvesting and conversion processes in a photosynthetic pseudo-organelle is achieved for the first time through a combination of petascale simulations, theoretical modeling, and experimental collaborations, visualized in a comprehensive movie. Both the modeling and visualization tasks currently utilize the capabilities of the Blue Waters petascale computer. Simulation and visualization of these fundamental cellular processes constantly pose new software and hardware challenges due to their size and complexity. Future simulations of cell-scale integration and long time-scale behavior (e.g., milliseconds for diffusive and minutes for assembly processes) will require a significantly greater computational capability than is available today, along with development of new methods and computational algorithms.

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