An Open Quantum System Model of Electron Bifurcation

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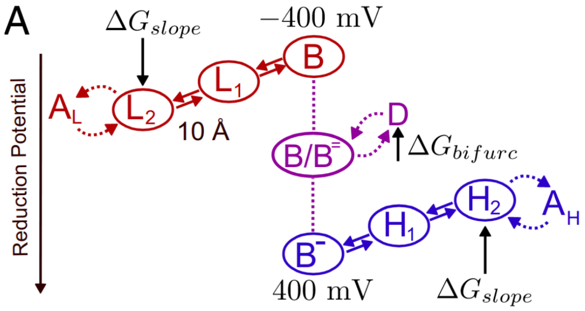

Figure 1: Free Energy Landscape making the extraction of two electrons from the donor B to the acceptors A_L and A_H reversible and protected from short-circuiting dissipative reactions. Adapted from [1].

Electron bifurcation (EB) is a chemical reaction central to a number of essential biological processes such as respiration, photosynthesis, or CO_2 reduction. This reaction spontaneously realises the extraction of two electron from a molecule (called a donor) and transports them along two different pathways (called transfer chains). One of these paths is higher in energy than the donor and the other one is lower in energy. The uphill transport of one of the electron is made possible by leveraging the gain in energy resulting from the downhill flow of the other electron. Surprisingly, EB is a free energy conserving process ($\Delta G \approx 0$) meaning that it is reversible. These properties make EB a unique biochemical reaction and it remained unclear how it could resist fast dissipative processes that would normaly prevent the transport to happen.

Recently, Yuly et al. [1] proposed a thermodynamical model to explain the properties of EB. They devised an equilibrium free-energy landscape (shown in Fig. 1) whose topology has the key properties of being energy conserving (enabling reversibility) and protecting EB from fast short-circuiting dissipative reactions.

However, EB being fundamentally a two-electron process, it should be possible to have a microscopical description complementary to the classical thermodynamical one. This description would be naturally framed as an open quantum system where the system of interest (the electron pair, the donor, and the transfer chains) would be interacting with a vibrational bath and electron reservoirs. For this quantum model to be meaningful, the fact that the charge separation is coordinated – one electron transfer relies on the gain in energy of the other to happen – implies that the dynamics is not solely dissipative. Hence the environment have to play an active role in this dynamics.

The first step in investigating the possibility of a microscopical description of EB is to find out what is the simplest model Hamiltonian exhibiting the key features of this biological process. Initially working at zero temperature enables us to relax the reversibility condition to the conservation of the system's energy (this way we can get rid of consideration around entropy productions); and it might be possible to observe spontaneous separation even after removing the fermionic reservoirs and keeping only a vibrational bath. Building on the simplest model to understand the physics of this system, we could then consider adding layers of complexity (temperature, additional reservoirs) and see whether they are essential to EB or how they affect it.

[1] J. L. Yuly, P. Zhang, C. E. Lubner, J. W. Peters, and D. N. Beratan, *Universal Free-Energy Landscape Produces Efficient and Reversible Electron Bifurcation*, PNAS, 117 (35), 2020, DOI:10.1073/pnas.2010815117.