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Excitation transfer connectivity in different purple bacteria: A theoretical and experimental study

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ABSTRACT

Photosynthetic membranes accommodate densely packed light-harvesting complexes which absorb light and convey excitation to the reaction center (RC). The relationship between the fluorescence yield (φ) and the fraction (x) of closed RCs is informative about the probability for an excitation reaching a closed RC to be redirected to another RC. In this work, we have examined in this respect membranes from various bacteria and searched for a correlation with the arrangement of the light-harvesting complexes as known from atomic force or electron microscopies. A first part of the paper is devoted to a theoretical study analyzing the $\varphi(x)$ relationship in various models: monomeric or dimeric RC–LH1 core complexes, with or without the peripheral LH2 complexes. We show that the simple "homogeneous" kinetic treatment used here agrees well with more detailed master equation calculations. We also discuss the agreement between information derived from the present technique and from singlet annihilation experiments. The experimental results show that the enhancement of the cross section of open RCs due to excitation transfer from closed units varies from 1.5 to 3 depending on species. The ratio of the core to core transfer rate (including the indirect pathway via LH2) to the rate of trapping in open units is in the range of 0.5 to 4. It is about 1 in *Rhodobacter sphaeroides* and does not increase significantly in mutants lacking LH2—despite the more numerous contacts between the dimeric core complexes expected in this case. The connectivity in this bacterium is due in good part to the fast transfer between the two partners of the dimeric $(RC–LH1–PufX)_2$ complex. The connectivity is however increased in the carotenoidless and LH2-less strain R26, which we ascribe to an anomalous LH1. A relatively high connectivity was found in *Rhodospirillum photometricum*, although not as high as predicted in the calculations of Fassioli et al. (2010). This illustrates a more general discrepancy between the measured efficiency of core to core excitation transfer and theoretical estimates. We argue that the limited core to core connectivity found in purple bacteria may reflect a trade-off between light-harvesting efficiency and the hindrance to quinone diffusion that would result from too tightly packed LH complexes.

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1. Introduction

Photosynthetic membranes are organized so as to absorb efficiently the incident light and transfer the excitation energy to the reaction center (RC). This is achieved by densely packing the membrane with pigmented proteins (light-harvesting complexes, LH) connected to the RC and, on a broader scale, by membrane folding or stacking, thus increasing the trapping section of the whole system (cell or chloroplast). In purple photosynthetic bacteria, the RC is surrounded by a complete or incomplete ring of core antenna (LH1), typically composed of 16 αβ heterodimers, each accommodating a pair of bacteriochlorophylls and one carotenoid. In addition to this RC–LH1 complex ("core complex"), some bacteria have a peripheral antenna complex, LH2 (or sometimes several types of peripheral complexes depending on culture conditions). When present, the amount of LH2 is regulated by light, increasing at low light intensity. The LH2 is also an oligomeric ring of (8–9) αβ heterodimers, each associating three bacteriochlorophylls and one carotenoid.

The photosynthetic excitation energy transfer process has been extensively studied, notably in bacterial systems. Structural, spectroscopic and theoretical investigations have led to important progress for understanding the physics involved at the molecular level (for reviews, see part 3 in [1]). At the supramolecular level, the arrangement of the RC–LH1 complexes, of LH2 (when present) and the other components of the photosynthetic machinery has also been investigated [2–4]. Much valuable structural information has been produced by atomic force microscopy (AFM) imaging of membranes from various bacteria [5].

The migration of excitation energy ("excitonic connectivity") has been studied by several techniques, using measurements of the yield of the

Abbreviations: AFM, atomic force microscopy; Bchl, bacteriochlorophyll; EM, electron microscopy; LH1, core light-harvesting complex (B875); LH2, Peripheral light-harvesting complex (B800-B850); RC, reaction center; *Rb.*, *Rhodobacter*; *Bcl.*, *Blastochloris*; *Rsp.*, *Rhodospirillum*

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fluorescence emitted by the Bchl antenna. One method (the first, historically [6,7]) consists in studying the relationship between the fluorescence yield F and the fraction x of photochemically closed RCs (i.e. centers where the primary donor P is oxidized or the primary quinone acceptor Q_A reduced). The fluorescence yield is proportional to the fraction of excitation which is not used up in photochemistry, thus reflecting, in a complementary way, the photochemical yield. The $F(x)$ relationship is non-linear, which indicates that the excitation visiting closed RCs can be redirected to open RCs, so that the trapping section of open RCs increases as their neighbors become closed. The "photosynthetic units" (i.e., in bacteria, the RC–LH1 complex) are thus somehow connected to each other. A related approach is the study of fluorescence induction kinetics $F(t)$. In a system where the open to closed transition can be observed as a single photochemical turnover during a continuous illumination (e.g., in the presence of an inhibitor blocking multiple turnover), the time course $F(t)$ depends on the degree of connectivity [7]. Separate units will yield an exponential time course, whereas connected units result in sigmoidal kinetics. Other methods for studying connectivity are based on the effect of photochemically generated quenchers, using short and intense laser flashes. In the singlet–triplet fusion technique [8], the quenching is caused by the triplet carotenoid generated in the antenna during an ~20 ns laser flash. A more utilized approach has been singlet–singlet annihilation, with flashes of ~30 ps that can generate several excitations in the same connectivity domain [9–13]. All these techniques indicate that, in purple bacteria as in Photosystem II, the diffusion domain of the excitation energy spreads over several core complexes. However, there has been some debate on the extent of this delocalization. The early investigations supported the notion of free diffusion over very large regions, i.e. a "matrix" or "lake model" picture. On the other hand, Trissl and coworkers [14,15] and Comayras et al. [16] concluded that the excitonic connectivity was more restricted, encompassing typically a few core units, which suggested kinetic and/or structural barriers. In the present work, we revisit this issue, addressing both theoretical and experimental aspects. The theoretical section extends previous work [16,17] and aims at providing a comprehensive toolbox to handle the various types of antenna organization found in purple bacteria: i.e. monomeric or dimeric core complexes, without or with LH2 present. This section is relatively independent from the experimental part and may be skipped by readers primarily interested in the latter. Our theoretical approach relies on a simplified treatment where the migrating excitation and the open or closed core complexes are handled as homogeneously distributed reactants, so that a very small set of rate constants are involved. If a more accurate treatment is desired one should resort to a detailed description of the excitation diffusion, using master equation or Monte Carlo calculations. A long standing question has been: how (in)accurate is the homogeneous treatment? The recent paper by Fassioli et al. [18] provides an opportunity to answer this question. This work applies the master equation method (and Monte Carlo simulations as well) for describing excitation transfer in membranes of *Rhodospirillum photometricum*, using the arrangement of the light-harvesting complexes found by AFM. We could thus compare the results from these calculations with those from the homogeneous treatment and, as will be shown, the homogeneous approach did rather well in this comparison. Furthermore, the modeling of excitation transfer in *Rsp. photometricum* offers the opportunity of a comparison with experimental data, since this bacterium is among those investigated here. This allows a check of the kinetic parameters used in the model and highlights the important fact that the effective rate constants for excitation transfer between light-harvesting complexes are definitely smaller than generally believed.

The experimental section reports data on connectivity in various bacteria. For some of these, independent structural information is available (from EM or AFM), and we searched for a correlation between both types of information. The organization schemes under focus are shown in Fig. 1. The simplest case is a regular array of monomeric core complexes (panel A), as encountered in *Blastochloris viridis* [19,20] and probably in *Rhodospirillum rubrum*. Panel B illustrates the case of a regular array of dimeric core complexes, as encountered in LH2-less mutants of *Rhodobacter sphaeroides* [21,22]. The other panels show membranes were LH2 is present in addition to core complexes, which may be monomeric as in the case of *Rsp. photometricum* (C) [23,24] or dimeric as in *Rhodobacter blasticus* (D) [25]. Panel E shows the organization scheme observed in membranes of WT *Rb. sphaeroides* (another case of dimeric core complexes with LH2) [26].

2. Materials and methods

2.1. Bacterial strains and growth conditions

All strains were grown in Hutner liquid medium, either in anaerobic conditions under illumination (photosynthetic growth), or, in the case of cytochrome deleted strains and their corresponding

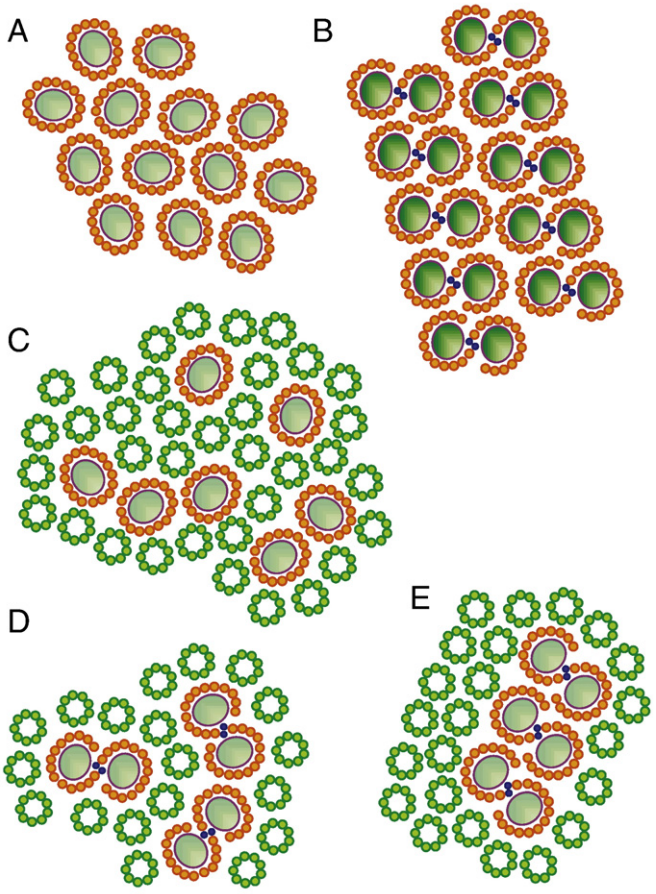

Fig. 1. Various supramolecular arrangements found in membranes from purple bacteria. (A) Hexagonal packing of monomeric core complexes (the RC is depicted as a green oval, the LH1 as an orange ring), as found in membranes of *Blastochloris viridis*. (B) Extended crystalline arrangement of the core dimers observed in tubular membranes in some LH2-less mutants (e.g., RCLH10) of *Rhodobacter sphaeroides*. (C) Monomeric core complexes and LH2 (green rings) as imaged by AFM in membranes of *Rhodospirillum photometricum*. In Refs. [23,24], it was estimated that 27% of the core complexes have zero direct contact with another core and 44% have one core contact; the most frequent core–core distance corresponds to intercalation of one LH2; and besides the type of domain illustrated here with disordered mixed core-LH2 arrangement, adjacent regions of hexagonally packed LH2 are present, whose abundance depends on growth light intensity. (D) The arrangement found in fused membranes of *Rhodobacter blasticus* where 75% of the core complexes are forming dimers. The PufX polypeptide, featured as a dark blue subunit, is believed to play a key role in the dimeric association; its location is debated (see the review [27]). (E) The arrangement found in fused membranes of *Rb. sphaeroides*, presenting short rows of RC–LH1 dimers surrounded by LH2.

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