Minireview

Pigment–protein architecture in the light-harvesting antenna complexes of purple bacteria: does the crystal structure reflect the native pigment–protein arrangement?

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Received 23 May 2000; accepted 14 June 2000

Edited by Vladimir Skulachev

Abstract Structural analysis of crystallized peripheral (LH2) and core antenna complexes (LH1) of purple bacteria has revealed circular aggregates of high rotational symmetry (C_8, C_9 and C_{16}, respectively). Quantum-chemical calculations indicate that in particular the waterwheel-like arrangements of pigments should show characteristic structure-sensitive spectroscopic behavior in the near infrared absorption region. Laser-spectroscopic data obtained with non-crystallized, isolated LH2 of *Rhodospirillum molischianum* are in line with a highly symmetric (C_8) circular aggregate, but deviations have been found for LH2 of *Rhodobacter sphaeroides* and *Rhodopseudomonas acidophila*. For both the latter, C-shaped incomplete circular aggregates (as seen only recently in electron micrographs of crystallized LH1–reaction center complexes) may be a suitable preliminary model.

Key words: Photosynthetic antenna complex; Circular aggregate; Crystal structure; *Rhodobacter sphaeroides*; *Rhodopseudomonas acidophila*; *Rhodospirillum molischianum*

1. Introduction

Nature accomplishes primary photosynthetic processes like light absorption, electronic excitation energy transfer as well as primary charge separation in the broad manifold of plant and bacteria species by employing essentially two types of pigments, (bacterio)chlorophylls and carotenoids. As monomers in dilute solution, these molecules deactivate after electronic excitation via radiating as well as non-radiating internal conversion and intersystem crossing channels. With respect to the mentioned photosynthetic processes however, these deactivation paths result in losses which potentially may reduce overall photosynthetic efficiency. To overcome this drawback nature has organized chromophores in pigment–protein networks.

Recent X-ray crystal structure analyses of two basic types of photosynthetic pigment–protein antenna complexes from purple bacteria, the peripheral light harvesting (LH2) [1,2] and the core light harvesting antenna complexes (LH1) [3], respectively, have revealed a highly symmetric arrangement of the pigments. In contrast, electron crystallography of the main light harvesting complex of higher plants (LHC II) [4] indicates a remarkably lower symmetry. Nevertheless, light harvesting and energy transfer in all these antenna complexes in native (non-crystallized) systems occur with high efficiency in ultrafast steps. As is well known, crystallization of membrane-intrinsic proteins requires extraction from their native membranes and stabilization and thus, prolonged exposure to detergents which may lead to structural rearrangement of pigments or the respective pigment-carrying apoproteins. Its is conceivable, too, that crystallization itself can have similar effects.

Therefore it is a question of topical interest, whether the crystal structure reflects the native molecular arrangement of the protein-embedded pigments.

Here we compare crystal structure-based theoretical predictions of spectroscopic properties of LH2 and LH1 with related experimental results as obtained with non-crystallized complexes in the near-infrared spectral region (NIR). Conclusions establishing a (preliminary) answer to the title question will be given.

2. Structure of crystallized antenna complexes of purple bacteria

A high resolution (to 2.2 Å) X-ray crystallography study [1] of LH2 of *Rhodopseudomonas (Rps.) acidophila* suggests a concentric arrangement of two hollow cylinders built from the transmembrane helices of nine α- and nine β-apoproteins, respectively. Within this protein-network, 27 bacteriochlorophyll *a* (BChl *a*) molecules are arranged in two distinct C_9-symmetric circular aggregates as shown in Fig. 1: 18 BChls *a* are located in-between the protein cylinders and nine BChls *a* are inserted between the helices of the β-chains of the outer wall. In this paper, we will refer mainly to the waterwheel-like aggregate of 18 BChls *a* because of its sensible structure–function relationship. This aggregate is called B850, because its main spectroscopic feature (in chromatophores as well as in isolated LH2) is an absorption band peaking at about 850 nm. It is assumed that this band belongs to an aggregate similar or identical to the waterwheel-like BChl *a* arrangement found in

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Abbreviations: LH1, LH2, core and peripheral light harvesting antenna complex of photosynthetic purple bacteria; LHC II, main light harvesting complex of higher plants; NIR, near-infrared spectral region; NLPF, non-linear polarization spectroscopy in the frequency domain; RC, reaction center; BChl *a*, Bacteriochlorophyll *a*; *Rb., Rhodobacter*; *Rps., Rhodopseudomonas*; *Rsp., Rhodospirillum*

0014-5793/00/$20.00
PII: S0014-5793(00)01892-5

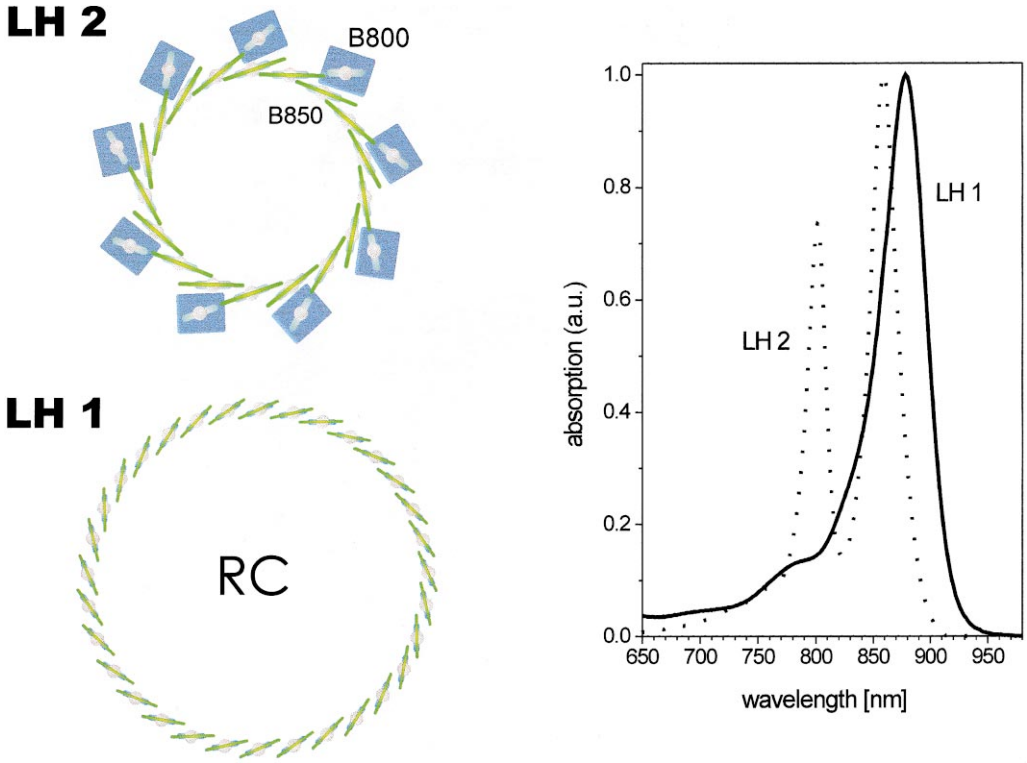

Fig. 1. Schematic representation of crystal structure-based BChl *a* arrangement (left) and NIR absorption spectra (right) of LH1 from *Rb. sphaeroides* and LH2 from *Rps. acidophila*. LH2: There is one smaller ring with waterwheel-like arrangement of 18 closely spaced BChls *a* (dimeric 'unit cell') with center-to-center distances of about 9 Å, and a larger ring of nine more distantly spaced BChls *a*, lying parallel to the ring plane (Mg–Mg separated by about 21 Å). This plane is aligned parallel to the membrane plane. The two rings are separated from each other by about 17 Å along the C_9 symmetry axis. The normal vectors of the BChl molecular planes of the larger ring (nine BChl) are aligned parallel to the C_9 axis, those of the smaller ring (18 BChl) are oriented perpendicular to this symmetry axis [36]. It is assumed that the 850-nm band belongs to an aggregate similar or identical to the waterwheel-like BChl arrangement in the crystal. This pigment ensemble will be called B850. Correspondingly, the ring of nine BChls is called B800. LH1: Analogous to B850, the circular aggregate of 30–34 BChls is called B875 according to the main NIR absorption band of the LH1 antenna. In usual pictures of the LH1–RC complexes each LH1 ring encircles one RC [5].

crystals. Correspondingly, the ring of nine (monomeric) BChls *a* is called B800 (see also Fig. 1).

For LH2 of *Rhodobacter (Rb.) sphaeroides* it has been demonstrated recently by electron microscopy [5], that the BChl *a* arrangement is very similar to that of *Rps. acidophila*. But notably, LH2 of *Rb. sphaeroides* shows a deviation from perfect C_9 symmetry, probably caused by a small tilt of the individual LH2 rings relative to the membrane normal [5].

X-ray crystallography of LH2 from *Rhodospirillum (Rsp.) molischianum* revealed an architecture analogous to that of both species mentioned before, but with C_8 symmetry [2]. The also waterwheel-like aggregate consists of 16 BChls *a*, arranged in eight dimeric building blocks.

It is important to note that in LH2 of all three species a network of hydrogen bonds and ligations to histidine residues are essential factors for stabilization of the waterwheel-like structure.

Two-dimensional crystals of LH1 and LH1–reaction center (RC) complexes from *Rb. sphaeroides* have been investigated recently by electron microscopy [5]. A waterwheel-like ring of 15–17 protein subunits, with two BChls *a* attached to each subunit, has been identified (see Fig. 1). Analogous to B850, this aggregate of 30–34 BChls *a* is called B875 according to the main NIR absorption band. In the LH1–RC complexes, each LH1 ring encircles one RC.

3. Theoretical predictions on excitation spectra of circular aggregates

Based on the crystal structural data, several theoretical calculations concerning the expected spectroscopic behavior of the B850 and B875 circular aggregates have been published [6–8]. Notably, there were already such calculations before the first X-ray analysis data for LH2 were published [9]. Dense packing of the BChls in these aggregates results in strong dipole–dipole interaction among them, expected to be reflected in spectroscopic behavior very different from that of monomeric BChl *a*.

The predicted excitation level structure [6,10–13] of the lowest-energetic one-excitation band of an 18 BChls *a* numbered, completely ordered circular aggregate (i.e. identical Q_y excitation energy of the individual BChls *a*) of C_9 symmetry is shown in Fig. 2. The only allowed radiating transitions from the ground state are those to the degenerate $k = \pm 1$ states in the lower manifold and to the $k = 9$ state in the upper manifold. The former transitions carry by far most of the oscillator strength of the 18 individual BChl Q_y transitions (more than 97%, see e.g. [6]). Correspondingly, both these degenerate absorption transitions have a transition dipole moment enhanced by a factor of about $\sqrt{9}$ as compared to monomeric BChl *a*. The excitation density in the experimentally indistinguishable states $k = \pm 1$ will be practically the

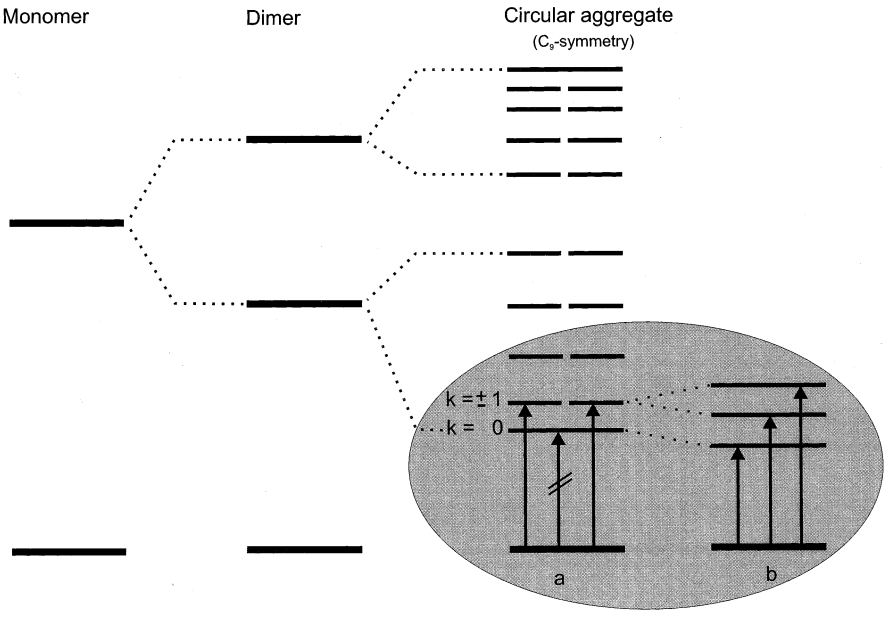

Fig. 2. excitation level structure of a circular aggregate of 18 strongly interacting BChls with C_9 symmetry (a) and schematic representation of possible changes of lowest-energetic level structure and transitions in case of distortion of symmetry (b). a: Because of the dimeric building blocks of the circular aggregate, there are two forms of one-excitation state. They contain nine levels each. Eight of them in each manifold belong to four pairs of degenerate excitation states (denoted $k = \pm 1$, ± 2, ± 3, ± 4 in the lower manifold; $k = \pm 5$, ± 6, ± 7, ± 8 in the upper manifold). Besides there is in each manifold one non-degenerate excitation ($k = 0$ and $k = 9$) level. The only allowed radiating transitions from the ground state are those to the degenerate $k = \pm 1$ states in the lower manifold and to the $k = 9$ state in the upper manifold.

same at all 18 BChls *a*, i.e. there is an (initial) excitation delocalization over the entire ring.

Theoretical investigations [14] predict similar spectroscopic features for a corresponding ring of 16 BChls *a* (as seen in the crystal structure of *Rsp. molischianum*) and for the 32-numbered circular aggregate of LH1.

From the calculated spectroscopic properties of the tightly packed, highly symmetric BChl *a* arrangements in the crystallized complexes, two basic facts are remarkable with respect to their role in photosynthetic energy transfer: (i) the lowest excited state is 'optically dark'. This means that besides radiationless relaxation there is no radiating transition to the ground state, i.e. this process competing with energy transfer is eliminated. (ii) Excitation in the dominating transition(s) to $k = \pm 1$ makes the energy immediately available at each site of the entire ring of chromophores.

If one assumes that the absorption of isolated LH2 and LH1 results from highly symmetric circular aggregates as seen in the respective crystals, than one has to identify their main NIR-absorption bands at about 850 nm and 875 nm, respectively, (see Fig. 1) with transitions from the ground state to the lowest degenerate ($k = \pm 1$) levels in both cases.

With respect to the topic of the present review, theoretical expectations about spectroscopic consequences of various possible deviations from the above described highly symmetric circular aggregates (with zero energetic disorder) are of special interest. Such deviations of BChl *a* arrangements in native complexes from that in the crystals may include:

1. Variations of the protein environment along the BChl binding sites in the circular aggregate, which may result in a Gaussian distribution of individual BChl *a* Q_y energies, while the respective symmetry is conserved (called diagonal disorder in the density matrix description [14]). Calculations show, that with increasing disorder split of the originally degenerate excitation levels may become significant (see Fig. 2). In connection with this, the oscillator strengths of the transitions to the two levels resulting from splitting of $k = \pm 1$ decrease, but their equality is conserved. The remaining oscillator strength is distributed to other, originally not allowed transitions, among them the transition to $k = 0$. The respective absorption bands become increasingly inhomogeneously broadened [14].
2. Variations of BChl *a* positions and orientations along the circular aggregate, which result in changes of dipole–dipole interactions (off-diagonal disorder). The expected modifications in excitation level structure and distribution of oscillator strengths are similar as in case (1) [15]. However, there are reasons to assume that off-diagonal disorder is of minor importance [16].
3. Principal deviations, such that the native system has a well defined but different structure as compared to that obtained by crystal structure analysis. In these cases one can expect quite different spectra critically depending on the shape of the complex. If the native LH2 rings were not circular but elliptical aggregates, the degeneracy of certain excitation levels would be lifted and one would expect a split of the B850-band into two subbands, each containing the same oscillator strength [17]. For 'broken' LH2-rings (which means that the distance between two of the unit cells of the circular aggregate is considerably enlarged) a lift of level degeneracy would be expected, too. In this case, however, the oscillator strength is not symmetrically distributed to the two subbands. Some of the oscillator strength of the transition with dipole moment di-

rected to the 'cut' is redistributed to the lowest excitation level [14].

4. The overall geometry of the native systems may deviate stochastically from the crystal structure (global disorder). For example, individual LH2 rings may be elliptically deformed to a varying degree. In this case one would expect an inhomogeneously broadened B850-band (instead of two distinct subbands as described in (3).

Summarizing, theoretical expectations concerning spectroscopic consequences of possible deviations in native BChl *a* arrangements as compared to those in waterwheel-like crystallized systems, are changes in the energetic positions of excitation levels, lift of level degeneracy and changes in the distribution of oscillator strengths.

Crucial experimental observations for answering the question of whether the crystal structure of LH2 as well as LH1 reflect the pigment arrangement in the native systems therefore include: number, spectral position and oscillator strengths/transition dipole moments of absorption transition(s) between ground state and levels of the lower one-excitation manifold. Of particular importance are these parameters as determined under physiological conditions in chromatophores (membrane fragments with the respective pigment–protein complexes in their native environment) or at least with complexes extracted from them as mild as possible. One has to focus on the following questions: Are the energy level pairs really degenerate? Are transitions to all other but $k = \pm 1$ excitation levels really completely forbidden?

Solving these questions implies search for a substructure of the main NIR-absorption bands of the compartments B850 (of LH2) and B875 (of LH1) and for further bands in their spectral neighborhood. In addition, the amount of inhomogeneous broadening of the transitions is important in this respect – this will be the subject of Section 4.

4. Experimental results and conclusions concerning NIR-absorption transitions in B850 of LH2 from three species of purple bacteria

According to the theoretical calculations, the lower manifold of energy levels of the one-excitation band of an 18- or 16-numbered ring with C_9 or C_8 symmetry spans over a region of at least 600 cm^{-1} [7,18] to about 1400 cm^{-1} [14,19]. Accordingly, the spectral range that should be inspected for respective absorption transition(s) in B850 is between 780 and 880 nm (see also Fig. 2). To accomplish this, one has to overcome the problem, that natural LH2 consists of two different circular compartments. Specifically, in the spectral range of interest there is at least one more major absorption transition (at about 800 nm), which does not belong to the B850 aggregate, but to B800, see Fig. 1. The use of B800-depleted complexes is helpful in this respect [20,21]; these complexes are referred to as p(pure)B850 in this review.

4.1. B850 in LH2 of Rb. sphaeroides

The 850-nm band in the absorption spectrum of the isolated LH2 does not show any indication of spectral substructure, not even at 4 K [18]. However, with non-linear polarization spectroscopy in the frequency domain (NLPF, a special optical mixing technique [22,23]) in B850 of *Rb. sphaeroides* such substructure can be clearly resolved already at room temperature [24]: there are two transitions with maxima at 847 and 857 nm (i.e. the splitting amounts to 140 cm^{-1}). The 847-nm subband is the stronger one by about one order of magnitude. For the discussion given below it is important to note that the inhomogeneous width of the main subband is below 100 cm^{-1}, while the homogeneous width is 470 cm^{-1}. The splitting in two subbands becomes more prominent at low temperatures (e.g. with maxima at 837 and 860 nm at 20 K, as shown in Fig. 3)[1].

The oscillator strength/dipole moment for the dominating transition in the 850 nm region has been determined to $3.3 \pm 0.6/25.5 \pm 2.5$ D based on the determination of the absorption cross section in the 850 nm region by non-linear absorption as well as of the homogeneous width by NLPF [25]. The obtained values correspond to a $\sqrt{16 \pm 4}$-fold enhancement as compared to the respective transition in monomeric BChl *a*. The results indicate an initial excitation delocalization over 16 ± 4 BChl *a* molecules. Besides the giant transition dipole moment, there is also strongly enhanced resonant third-order optical non-linearity [25].

Hole burning spectroscopy (at cryogenic temperatures) indicates the existence of a weak transition at about 870 nm. Extrapolation to room temperature (if allowed at all) would result in a blue shift by about 5 nm. Unluckily, at present NLPF at room temperature does not allow a decision about such a weak transition at about 865 nm, because it is too close to the spectral limit of the measuring range (B. Voigt, unpublished results). To the blue of the 850-nm band, there is a further small band (located at about 800 nm). This excitation transition has been identified in pB850 [21]. In native (complete) LH2 this B850 side band is masked by the main B800 transition, but has also been detected there [18,21,26].[2]

The given experimental facts indicate that at room temperature in the isolated B800–850 antenna of *Rb. sphaeroides* the molecular structure responsible for the 850-nm absorption (B850) does not correspond to the ideal waterwheel-like structure in the crystal: If the transitions at 847 and 857 nm are interpreted in terms of lift of degeneracy of the $k = \pm 1$ level, ring models with increased disorder fail because of the experimentally found large difference in oscillator strength between these transitions. Moreover, because of the measured small inhomogeneous broadening, the 857-nm transition can not be assigned to the transition to $k = 0$. (Note, that by lowering the temperature this transition even gains in intensity relative to the 847-nm transition). On the other hand, the experimental facts are generally in line with theoretical expectations for

[1] Note, that work at cryogenic temperatures requires addition of glycerol to the buffer to obtain a glass, and this addition of glycerol results in a significant deviation of B850 band shape from that in normal buffer already at room temperature (see Fig. 3 for details), which indicates changes in antenna architecture. Therefore we avoid reviewing in detail related spectroscopic results as obtained at cryogenic temperatures in this paper.

[2] Noteworthy, simulation of NIR-difference absorption spectra of LH2 (measured with 150-fs pulses at 830-nm excitation) on the basis of a regular C_9 circular aggregate [27] seems to indicate that in the 850-nm region there are 3–5 transitions to the excitation levels $k = 0$, $k = \pm 1$ and $k = \pm 2$ with dipole moments of 2.41, 6.40, 6.52, 0.95 and 0.68 (as normalized to the Q_y-transition dipole moment in the monomer). However, assumption of level degeneracy seems to be questionable.

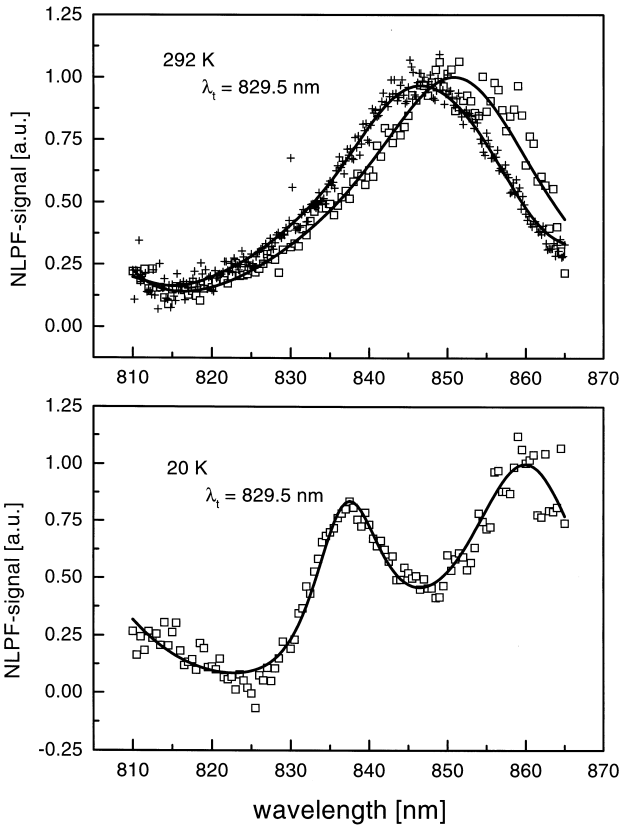

Fig. 3. NLPF spectrum of the isolated LH2 from *Rb. sphaeroides*, suspended in LDAO-buffer (+) and in a LDAO/glycerol (1:2) mixture (□) at 292 K (top), and the latter at 20 K (bottom). The smooth lines represent simulations according to NLPF lineshape theory [37] with model parameters as described in the text.

the case of a disruption of the circular arrangement [14]. In such a model both the aforementioned transitions may be either the result of lift of degeneracy of $k = \pm 1$, or (much less probable) the 857-nm subband may correspond to the transition to a lowest level analogous to $k = 0$ in the system with C_9 symmetry (see Fig. 2). Preliminary results for B850 in chromatophores indicate similar spectroscopic behavior as in isolated LH2, i.e. suggest also deviations from the ideal architecture in crystals [24].

4.2. B850 in LH2 of Rps. acidophila

Most of the spectroscopic features mentioned above for B850 of *Rb. sphaeroides* are observed also in B850 of *Rps. acidophila*: The red-most absorption band in the isolated B800–850 complex (which peaks at 859 nm at room temperature, see Fig. 1), does not show any indication of substructure, not even at cryogenic temperatures [28]. NLPF at room temperature identifies a dominant subband at 853 nm with a homogeneous width comparable to the width of the absorption band (410 cm^{-1}), and an inhomogeneous width smaller than 100 cm^{-1}. The transition dipole moment is 24.6 ± 4.6 D, indicating initial excitation delocalization over 14 ± 6 BChl [29]. NLPF also indicates the existence of a red-shifted, less pronounced subband [24]. The present conclusion is that also for the B850 compartment of LH2 from *Rps. acidophila*, the assumption of an ideal C_9-symmetric waterwheel-like aggregate as in the corresponding crystals can not be maintained. Symmetry is lowered, and the model of disordered rings fails. Also here, a dissected 'circular' aggregate is a suitable working model.

4.3. B850 in LH2 of Rsp. molischianum

The NIR-absorption band of B850 of LH2 has its maximum at 846 nm at room temperature, and there is no indication of substructure even at 4.2 K [18]. Unlike in the two other species described before, a closer inspection of this absorption band of the *Rsp. molischianum* complex indicates that its shape is nearly Lorentzian. Careful NLPF investigations also do not show any indication of a substructure of this band, i.e. there appears to be only a single transition, with a homogeneous line width of 474 ± 10 cm^{-1} and an inhomogeneous line width smaller than 120 cm^{-1} (W. Beenken, unpublished results). The absorption cross section at band maximum, determined by non-linear absorption, is 3.4×10^{-15} cm^{-2} (H. Stiel, unpublished results). This results in a transition dipole moment for the 850-nm band of *Rsp. molischianum* of 20 ± 3 D, indicating initial excitation delocalization over 9 ± 2 BChl (see also [30]). The theoretically predicted dipole moment for the transition(s) to the degenerate $k = \pm 1$ levels in nearly perfect C_8-symmetry (of the respective crystals) is 17.4 D [14]. Moreover, for the first time the absorption cross section of a strongly coupled aggregate could be determined independently by another, new method, based on the intensity dependence of NLPF [31]. The technique, which is sensitive to the orientation of the transition dipoles, suggests for the 850-nm band two perpendicular transition dipole moments, each of 17.8 ± 0.9 D. Furthermore, the experimental data indicate a fast relaxation to a 'dark' state, which may be identified as the $k = 0$ level.

We conclude therefore, that at room temperature the BChl arrangement in B850 of LH2 of *Rsp. molischianum* is very much the same as that in the crystal.

5. First experimental results concerning B875 (LH1) of *Rb. sphaeroides*

The aforementioned results indicate that in native B850 of LH2 of *Rb. sphaeroides* and *Rps. acidophila*, the waterwheel-like, closed circular arrangement of BChls *a* as seen in the crystal structure analyses seems to be modified towards open ring structures. The following is focused on the detection of such modified structures in LH1.

For the red-most absorption band of LH1 of *Rb. sphaeroides* (peaking at 875 nm) substructure analysis with NLPF does not exist, yet. Investigations with other techniques at 77 K seem to indicate, that there are no distinct subbands (contrary to the situation with the BChl *b*-containing LH1 antenna of *Rhodopseudomonas (Rps.) viridis* [27]). Moreover, the homogeneous width of this band amounts to 75–90% of the overall absorption band width [32]. Recent fs pump-probe and fluorescence quenching experiments with LH1 reconstituted from a LH1-only mutant (in which varying amounts of BChl *a* had been substituted by Ni-bacteriopheophytin *a*) suggest that the size of the absorbing unit is 20 BChls (with very little size distribution). Upon fs-pulsed excitation there is initial excitation delocalization over the whole unit [33,34]. With this relatively small number of 20 BChls *a* it appears to be impossible to place a RC inside a corresponding small closed circular aggregate. The latter possibility was inferred

from crystal structure data suggesting ring dimensions incorporating 32 pigments (see above). This model of a 32-numbered ring enclosing the RC is underlying theoretical descriptions of light harvesting and energy transfer in the whole photosynthetic unit of purple bacteria until now [14]. Calculations also suggested that the minimum number of BChls for an RC-enclosing ring is 32 [14]. Interestingly, a recent electron micrograph analysis of native tubular membranes from *Rb. sphaeroides* showed S-shaped LH1–RC supramolecular complexes. The observations can be rationalized as a dimer of two C-shaped LH1, each containing 24 BChls and 'embracing' a RC [35]. The C-shaped structures of LH1 seem to be related to the presence of the PufX polypeptide [35].

6. Concluding remarks

Generalization of results implying high rotational symmetry of BChl *a* arrangements in B800, B850 as well as B875 compartments of crystallized light harvesting complexes of purple bacteria had resulted in a development of a model of the complete purple bacterial photosynthetic unit [8,14,19], which may function as a valuable frame to fit in as much as possible from the wealth of related experimental results. Moreover, it may generate new questions and stimulate investigations, e.g. on possible limits of its validity. The title question of the present paper is of this kind. Answers, which we have attempted to formulate above, are of a preliminary nature. Surprisingly enough, the model of a highly symmetric LH2 antenna as derived from crystal structure analyses holds true (according to tailor-made spectroscopic investigations) for the native LH2 of *Rsp. molischianum* until now. For the respective isolated antenna complexes of *Rb. sphaeroides* and *Rps. acidophila,* differences to the crystal structures are obvious, they strongly suggest incomplete ring structures.

Acknowledgements: The authors thank Prof. Hugo Scheer (Botanical Institute, University of Munich) for a long period of inspiring cooperation. Comments and suggestions by Dr. Heiko Lokstein (Institute of Biology, Humboldt University of Berlin) are gratefully acknowledged.

References

[1] McDermott, G., Prince, S.M., Freer, A.A., Papiz, M.Z., Hawthornwaite-Lawless, A.M., Cogdell, R.J. and Isaacs, N.W. (1995) Nature 374, 517–521.
[2] Koepke, J., Hu, X., Muenke, C., Schulten, K. and Michel, H. (1996) Structure 4, 581–597.
[3] Karrasch, S., Bullough, P.A. and Ghosh, R. (1995) EMBO J. 14, 631–638.
[4] Kühlbrandt, W., Wang, D.N. and Fujiyoshi, Y. (1994) Nature 367, 614–618.
[5] Walz, T., Jamieson, S.J., Bowers, C.M., Bullough, P.A. and Hunter, C.N. (1998) J. Mol. Biol. 282, 833–845.
[6] Sauer, K., Cogdell, R.J., Prince, S.M., Freer, A., Isaacs, N.W. and Scheer, H. (1996) Photochem. Photobiol. 64, 564–576.
[7] Alden, R.G., Johnson, E., Nagarajan, V., Parson, W.W., Law, C.J. and Cogdell, R.G. (1997) J. Phys. Chem. B 101, 4667–4680.
[8] Hu, X. and Schulten, K. (1997) Phys. Today 1, 28–34.
[9] Dracheva, T.V., Novoderezhkin, V.I. and Razjivin, A.P. (1995) Chem. Phys. 194, 223–235.
[10] Bakalis, L.D., Coca, M. and Knoester, J. (1999) J. Chem. Phys. 110, 2208–2218.
[11] Barvik, I., Warns, C., Neidlinger, T. and Reinecker, P. (1999) Chem. Phys. 240, 173–189.
[12] Kühn, O. and Mukamel, S. (1997) J. Phys. Chem. B 101, 809–816.
[13] Meier, T., Chernyak, V. and Mukamel, S. (1997) J. Phys. Chem. B 101, 7332–7342.
[14] Hu, X., Ritz, T., Damjanovic, A. and Schulten, K. (1997) J. Phys. Chem. B 101, 3854–3871.
[15] Wu, H.-M. and Small, G.J. (1997) Chem. Phys. 218, 225–234.
[16] Wu, H.-M. and Small, G.J. (1998) J. Phys. Chem. B 102, 888–898.
[17] van Oijen, A.M., Ketelaars, M., Köhler, J., Aartsma, T.J. and Schmidt, J. (1999) Science 285, 400–402.
[18] Wu, H.-M., Reddy, N.R.S., Cogdell, R.J., Muenke, C., Michel, H. and Small, G.J. (1996) Mol. Cryst. Liq. Cryst. 291, 163–173.
[19] Ritz, T., Hu, X., Damjanovic, A. and Schulten, K. (1998) J. Lumin. 76/77, 310–321.
[20] Bandilla, M. (1995), thesis, University of Munich, Munich.
[21] Leupold, D., Stiel, H., Ehlert, J., Nowak, F., Teuchner, K., Voigt, B., Bandilla, M., Ücker, B. and Scheer, H. (1999) Chem. Phys. Lett. 301, 537–545.
[22] Song, J.J., Lee, J.H. and Levinson, M.D. (1978) Phys. Rev. A. 17, 1439–1447.
[23] Voigt, B., Nowak, F., Ehlert, J., Beenken, W.J.D., Leupold, D. and Sandner, W. (1997) Chem. Phys. Lett. 278, 380–390.
[24] Nowak, F.R. (2000), thesis, University of Potsdam, Potsdam.
[25] Leupold, D., Stiel, H., Teuchner, K., Nowak, F., Sandner, W., Ücker, B. and Scheer, H. (1996) Phys. Rev. Lett. 77, 4675–4678.
[26] Koolhaas, M.H.C., Frese, R.N., Fowler, G.J.S., Bibby, T.S., Georgakopoulou, S., van der Zwan, G., Hunter, C.N. and van Grondelle, R. (1998) Biochemistry 37, 4693–4698.
[27] Novoderezhkin, V., Monshouwer, R. and van Grondelle, R. (1999) Biophys. J. 77, 666–681.
[28] Wu, H.-M., Reddy, N.R.S. and Small, G.J. (1997) J. Phys. Chem. B 101, 651–656.
[29] Stiel, H., Leupold, D., Teuchner, K., Nowak, F., Scheer, H. and Cogdell, R.J. (1997) Chem. Phys. Lett. 276, 62–69.
[30] Razjivin, A., Leupold, D., and Novoderezhkin, V. (1998) in: Photosynthesis: Mechanisms and Effects (Garab, G., Ed.), vol. 1, pp. 15–20, Kluwer, Dordrecht.
[31] Beenken, W. and May, V. (1997) J. Opt. Soc. Am. B 14, 2804–2810.
[32] Westerhuis, W.H.J., Hunter, C.N., van Grondelle, R. and Niederman, R.A. (1999) J. Phys. Chem. B 103, 7733–7742.
[33] Fiedor, L., Leupold, D., Teuchner, K., Voigt, B., Hunter, C.N., Scherz, A. and Scheer, H. (2000) Biochemistry, in press.
[34] Fiedor, L., Scheer, H., Hunter, C.N., Tschirschwitz, F., Voigt, B., Ehlert, J., Nibbering, E., Leupold, D. and Elsässer, T. (2000) Chem. Phys. Lett. 319, 145–152.
[35] Jungas, C., Ranck, J.-L., Rigaud, J.-L., Joliot, P. and Vermeglio, A. (1999) EMBO J. 18, 534–542.
[36] Cogdell, R.J., Isaacs, N.W., Freer, A.A., Arrelano, J., Howard, T.D., Papiz, M.Z., Hawthornthwaite-Lawless, A.M. and Prince, S. (1997) Progr. Biophys. Mol. Biol. 68, 1–27.
[37] Beenken, W. and Ehlert, J. (1998) J. Chem. Phys. 109, 10126–10137.