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ORIENTATION OF THE BACTERIOCHLOROPHYLL TRIPLET AND THE PRIMARY UBIQUINONE ACCEPTOR OF *RHODOSPIRILLUM RUBRUM* IN MEMBRANE MULTILAYERS DETERMINED BY ESR SPECTROSCOPY (I) *

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Summary

Chromatophores from *Rhodospirillum rubrum* were oriented as multilayers on quartz slides under reducing conditions. Irradiation of these multilayers in the resonance cavity of an ESR spectrometer at 6 K yielded the spectrum of the bacteriochlorophyll dimer triplet. The relative intensities of the main six lines of the triplet were dependent on the angle subtended by the direction of the external magnetic field with plane of the multilayers. The angular dependence of the intensities of these transitions can best be interpreted in terms of one of the principal axes of the triplet lying along the plane of the membrane while the other two axes are tilted 10—20° away from the parallel to and normal to the membrane directions. If we assume the porphyrin planes of the dimer to be parallel and the largest splitting of the triplet transitions to correspond to those transitions in a direction normal to this plane, then these data imply that the dimer planes are nearly perpendicular to the membrane plane.

Purified iron-depleted phototrap complexes were similarly oriented in reconstituted phosphatidylcholine multilayers and the angular dependence of the light-induced spectrum recorded at room temperature. A computer analysis of this angular dependence suggests that the plane of the primary ubiquinone acceptor molecule is parallel to the plane of the membrane and therefore, perpendicular to the donor.

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* This paper was already published as a preliminary report (Ref. 1).
Abbreviation: BChl, bacteriochlorophyll.

Introduction

Membranes serve a multifunctional role in living cells. Model systems such as those proposed by Singer and Nicholson [2] and Jain and White [3] depict the membrane as a viscous lipid fluid containing islands or plates of less-mobile protein-lipid aggregates. It is hypothesized that the functional purpose of this aggregation is an efficient facilitation of membrane enzymic and transport functions.

Undoubtably one of the more important biological processes occurring in the membrane is the series of electron transport reactions linked to phosphorylation. Although a large number of electron transport proteins have been identified, few have been isolated and even fewer sequenced. In order to better understand the structure of the biological electron transport system, it would be useful to know the relative orientation of each protein as well as the protein's redox group. This paper discusses the use of oriented membrane multilayers to determine the orientation of the primary photochemical electron transport components in photosynthetic bacteria.

Membrane multilayers were first investigated by ESR spectroscopy using nitroxide spin labels [4—6]. It was found that membrane vesicles could be dried on planar surfaces to produce a stacking of the membrane bilayer parallel to the plane of the surface. By observing the angular dependence of the spectrum of spin labels incorporated into these multilayers, the orientation of the nitroxide relative to the membrane can be discerned. Similar experiments can be performed with innate membrane-bound paramagnets. Research has commenced not only in our laboratory [1] but also in others using either vesicle drying [7—9] or large magnetic fields [10,11] to orient biological membranes in order to determine the orientation of the various membrane-bound proteins.

In this paper we describe the determination of the orientation of the triplet of the special primary electron donor pair of bacteriochlorophyll molecules and the primary ubiquinone acceptor in iron-depleted phototrap complexes of *Rhodospirillum rubrum*. A computer simulation of the angular dependence of the oriented ESR spectra of these two species suggests that their molecular planes are nearly perpendicular to one another. In this system, the plane of the dimer makes an angle of 10—20° with the normal to the membrane surface while the quinonoid plane of the primary ubiquinone lies approximately coplanar with the membrane.

Methods

Wild type *Rhodospirillum rubrum* was grown according to conventional procedures [12]. Chromatophores were prepared by ultrasonication with a Branson sonifier (Model 200) in glycylglycine buffer (pH 7.5). These chromatophores were concentrated by centrifugation and resuspended in Tris buffer (0.1 M, pH 7.5) such that the absorbance at 870 nm was approx. 150. Only freshly prepared chromatophores were used.

Prior to forming membrane multilayers for observance of the triplet of the primary bacteriochlorophyll dimer, chromatophores were reduced under nitro-

gen gas with dithionite to a potential $E^{\circ\prime}$ of approx. −150 mV. 200 μl of the reduced chromatophores were layered on a 0.5 × 5 cm quartz slide fused to a quartz rod. This suspension was allowed to dry under nitrogen until chromatophore multilayers were formed. Properly dried multilayers appeared dark red and glossy while overdrying produced a nearly black layer which tended to flake off the slide. This slide was positioned in the insert dewar of a Heli-tran liquid helium transfer system (Air Products, Model LTD-3-110) and held in place by an aluminum goniometer. All spectra were recorded at 6 K in a cavity of a Varian E-109 ES ESR spectrometer resonating in a TM_{110} mode at X-band frequencies. To improve the signal-to-noise ratio of the triplet spectrum, the spectrum of the dark adapted sample was first recorded and stored on a computer of average transients (Varian CAT-1024) and subtracted from the spectrum of the irradiated sample. In most experiments, ten spectral scans were stored of both irradiated and dark samples. All samples were irradiated in situ with a 100 W tungsten-iodine lamp.

To prepare iron-depleted phototrap complexes, the AUT-e method was used as formulated by Loach and coworkers [13—15]. In this method, chromatophores are solubilized in a solution of 3% Triton X-100 detergent and 36% urea in phosphate buffer (pH 12.0) held at 4°C for 45 min. This solution is then neutralized, dialysed and electrophoresed as previously described [13—15]. The resultant pigmented protein contained active iron-depleted phototraps with much of the original carotenoids and antenna bacteriochlorophyll. This protein will be referred to as an AUT-e particle. This method was chosen over the conventional preparation of reaction centers since the AUT-e particle still possesses much of the antenna pigment-protein, and therefore, it was felt it would reconstitute in membranes in an orientation closer to that of the in vivo complex than might the antenna-free reaction centers. Furthermore, the presence of carotenoids in the AUT-e particle enhances its longevity by decreasing its lability to oxygen.

After their separation and isolation on a liquit electrophoresis column [15], the AUT-e particles were detritonized by extensive dialysis against 0.1 M phosphate buffer (pH 7.8) and subsequently concentrated by centrifugation at 150 000 × *g* for 45 min. Although it does not remove completely all of the Triton, this procedure has been shown [13,14] to decrease the detergent concentration to the point of producing AUT-e aggregates. Phosphatidylcholine-AUT-e vesicles were prepared by evaporating an ethanol solution (approx. 0.6 ml) of 60 mg phosphatidylcholine in the bottom of a 5-ml beaker. 0.2 ml phosphate buffer was added to the beaker and sonicated for 1 min to form a suspension of phosphatidylcholine vesicles. A second 1-min sonication was performed after the addition of 0.4 ml detritonized AUT-e (absorbance at 865 = 80). This suspension was centrifuged at 20 000 × *g* for 10 min to percipitate the membrane bound from any remaining soluble AUT-e. The pellet was resuspended in phosphate buffer and allowed to slowly dry on quartz slides under nitrogen gas as described above for chromatophores.

Computer simulations of oriented spectra were conducted on an IBM 370 Model 3033 using a version of the basic program described by Libertini et al. [16] expanded by us to simulate oriented radicals with three different principal *g* factors and as well as anisotrpic proton hyperfine coupling.

Results

Bacteriochlorophyll dimer triplet

In order to be able to investigate the orientation of a membrane-bound radical in multilayers, the radical must possess detectable anisotropy in its powder spectrum. Due to the high degree of delocalization of its unpaired electron as well as the small amount of spin-orbit coupling, the principal g and a parameters of the bacteriochlorophyll cation and the cation of its dimer ($[BChl]_2^+$) are virtually isotropic. For this reason, the highly anisotropic triplet spectrum [17] was used to investigate the orientation of the special donor pair of bacteriochlorophyll molecules.

The triplet signal of the special pair of bacteriochlorophyll molecules in the donor unit was first observed by Dutton et al. [17]. The spectrum contains six lines due to anisotrpic spin-spin interaction and centered about $g = 2.00$. This spectrum is unique in that it is spin-polarized such that three lines represent absorption and three emission. It is only observed at very low temperatures when the primary acceptor is blocked by reduction.

Fig. 1 shows the light-minus-dark difference spectra recorded at 6 K of the bacteriochlorophyll dimer triplet ($[BChl]_2^T$) in oriented chromatophore multilayers with the membrane multilayer plane aligned parallel (90°, top) and perpendicular (0°, bottom) to the direction of the external magnetic field. The six derivative lines of the triplet spectrum are grouped into axis-specific pairs and labeled according to an arbitrary set of molecule-based principal axes. The

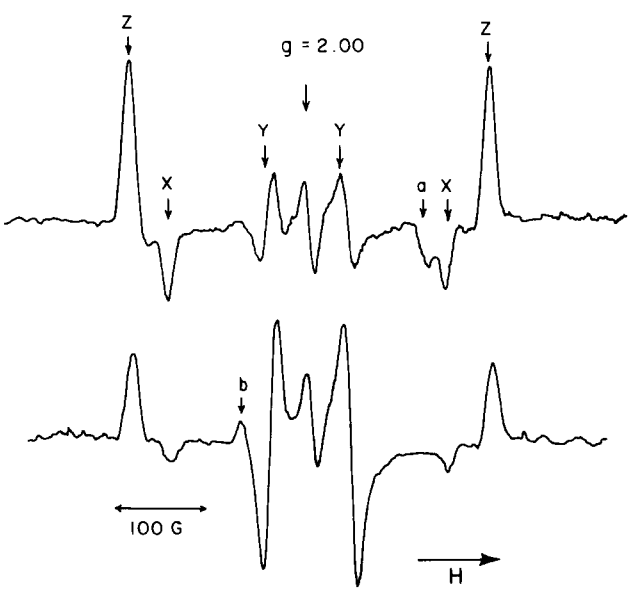


Fig. 1. Light-minus dark difference ESR spectra of *R. rubrum* in oriented chromatophore multilayers at 6 K. Top figure corresponds to spectrum taken with multilayers oriented parallel to the external magnetic field (membrane normal at 90°) while the bottom spectrum corresponds to the perpendicular direction (normal at 0°). The six triplet transitions are labeled according to an arbitrary molecule-based axis system. Signal a and b are light-induced but of unknown origin. Spectrometer settings: power 1.0 mW, frequency 9.1 GHz, modulation amplitude 6G, time constant 0.1 s.

axis labeling of the spectra uses the normal convention of choosing the z-axis to correspond to the lowest and highest field transitions. The identities of inflections a and b in Fig. 1 are not known at this time, but each is light-induced and the intensity of each is angle dependent. The signal at $g = 2.00$ is assigned to $[BChl]_2^+$ associated with a small concentration of unreduced reaction center. At the spectrometer settings used to record the spectrum in Fig. 1, the residual iron-quinone acceptor signal is unobservable.

In Fig. 2 the angular dependencies of the relative amplitudes of each axial pair of transitions is plotted as a function of the angle subtended by the external magnetic field and the normal to the plane of the membrane. These amplitudes are all standardized relative to the amplitude of $[BChl]_2^+$ in the spectrum. Since membrane multilayers produce only a two- and not a three-dimensional orientation of $[BChl]_2^T$, the variation can be plotted only for angles from 0 to 90°C. The variation from 90 to 180° would, of course, be just the mirror image of this figure. In fact, the two-dimensional nature of multilayers requires that all angle-dependent plots of spectral amplitudes (such as Fig. 2) have zero slopes at both 0 and 90°. The amplitude variation shown in Fig. 2 can best be explained in terms of the principal axes of $[BChl]^T$ oriented such that the spectral labeled x-axis is nearly parallel to the plane of the membrane while the y- and z-axes are tilted 10—20° from the normal to the plane and the parallel to the plane of the membrane, respectively. Computer simulations of these variation suggest an angular standard distribution about each axis of approx. 30°.

Ubiquinone acceptor

Chromatopore multilayers obviously cannot be used to determine the orientation of ubiquinone in the quinone-iron complex. The orientation of this complex can be easily determined by the above technique but tells us nothing about the orientation of the quinone in the complex; it only tells us the principal axis system of the complex whose structure is unknown. The orientation of the quinone acceptor can therefore, only be determined after the iron is removed. This is why AUT-e particles were used for this determination.

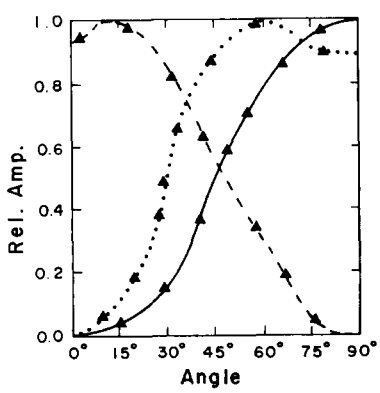
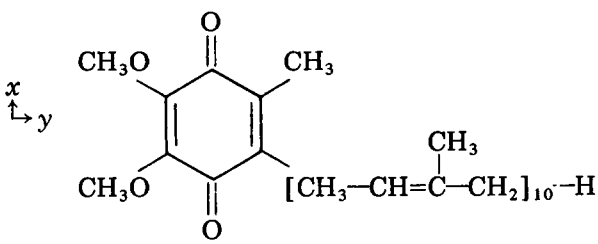


Fig. 2. Angular dependencies of relative intensities of each axial transition in triplet spectrum (———, x-axis; - - - - - -, y-axis; and · · · · · ·, z-axis; see Fig. 1 for axial assignment). The angular dependency of oriented $[BChl]_2^T$ in AUT-e-phosphatidylcholine multilayers is also shown by a small number of representative data (▲).

As explained in the Methods section above, AUT-e particles were isolated from *R. rubrum* chromatophores, detritonized and reconstituted into phosphatidylcholine vesicles by sonication. Evaporation of the buffered vesicle suspension layered on quartz slides produced phosphatidylcholine membrane multilayers containing the membrane-bound AUT-e. The ability of these vesicles to produce well-oriented multilayers was tested in earlier (unpublished) experiments in which the spin label 5-doxylstearic acid was incorporated into the membrane vesicles prior to multilayer formation. The orientation dependence of the label's ESR spectrum was similar to that observed by others [4—6] on phosphatidylcholine multilayers and clearly implies good multilayer formation.

The two carbonyl groups on quinone give the molecule enough spin-orbit coupling to impart to the semiquinone radical an anisotropic g factor. On the other hand, the absence of protons directly bonded to the quinonoid ring, i.e.,



Ubiquinone—10

means that most of the radical's anisotropy will occur in its g factor with only a minor contribution from the various proton coupling constants. To date, no measurements have been performed to determine the magnitudes of the various principal g factors of the ubisemiquinone anion ($Q^{\cdot-}$). Recently, we determined [18] the principal g factors for the benzosemiquinone anion radical in methanol to be $g_{xx} = 2.0065$, $g_{yy} = 2.0053$ and $g_{zz} = 2.0023$ based on the axis system where the molecular x-axis is along the carbonyl bonds and the z-axis is perpendicular to the plane of the molecule. Since hydrocarbon substitution onto the quinonoid ring produces only a minor increase of the isotropic g factor over that for benzosemiquinone (e.g., $g_{\text{benzosemiquinone}} = 2.0047$ while $g_{\text{durosemiquinone}} = 2.0049$ [19], the principal g factors can be assumed to be approximately the same for all aliphatic substituted benzoquinones including methoxy substitution. Therefore, since we can assume that the splitting constants are isotropic and the principal g factors are approximately those determined for benzosemiquinone, in order to simulate the spectrum of the immobilized $Q^{\cdot-}$, only a line-width broadening factor is needed. This term is obtained by determining the line width which yields the computer simulation closest to the experimental spectrum.

Since it was desired to observe $Q^{\cdot-}$ in its least perturbed environment, it was felt that the natural photochemical reduction of ubiquinone by the donor bacteriochlorophyll special dimer would be better than chemical reduction using an exogenous reduction. The use of chemical reduction is further complicated by the fact [20] that the quinone in the AUT-e particle is difficult to chemically reduce requiring a very low potential (approx. —350 mV). On the

other hand, photochemical generation of Q^- also produces the bacteriochlorophyll dimer radical. The resultant spectrum is therefore, the overlap of the spectra of these two species. As it turns out, our computer simulations show that for approximately equal concentrations of $[BChl]_2^+$ and Q^-, the spectrum of the oriented phototrap shows a greater angular variation than can be observed in the spectrum of the anion alone. Apparently the different g factor and line-width of the bacteriochlorophyll dimer cation [21] compared to the ubisemiquinone anion yields a resultant spectrum with greater angle-dependent line-shape variation than that of the anion alone.

Fig. 3 shows that angular dependence (circles) of the peak-to-peak line-width of the light-induced signal in phosphatidylcholine multilayers containing AUT-e particles. This set of data represents the statistical average of the angular dependence of seven different samples. The calculated standard deviation is shown as error bars for each point. As implied by the small magnitude of these error bars, each of the seven determinations showed essentially the same angular variation.

In order to computer simulate the data in Fig. 3, four parameters must be specified. As mentioned above, since the principal g factors and line-widths have already been determined for both $[BChl]_2^+$ and Q^-, the only parameters needed for simulation are: (1) the ratio of $[BChl]_2^+$ to Q^- in the composite signal, (2) the tilt angles that two of the Q^- principal axes makes with the normal to the membrane, and (3) the angular distribution (σ) about these specified angles. It was determined that the best computer simulation of the empirical data corresponds to a $[BChl]_2^+/Q^-$ concentration ratio of approx. 1.1, a tilt angle of the Q^- x-, y and z-axes with the normal to the membrane of 0, 90 and 90°, respectively, and an angular standard distribution about these angles of 40°. This best-fit simulation is shown as the solid curve in Fig. 3.

It is important to understand how changes in these parameters effect the

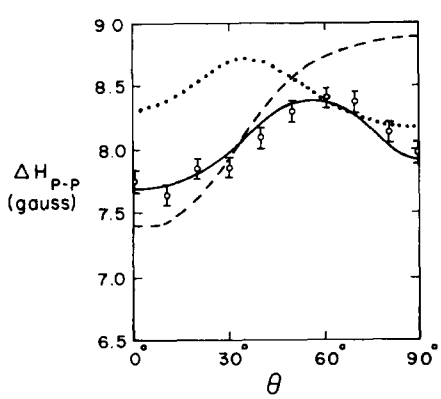


Fig. 3. Angular dependence (○) of composite line-width of light-induced bacteriochlorophyll dimer-ubiquinone radical signals from AUT-e particles in oriented phosphatidylcholine multilayers. Data represent average of seven different experiments on seven different preparations. Error bars shown represent calculated standard deviation. All spectra were recorded at room temperature. Solid line represents calculated variation with molecular-based z-axis (normal to quinonoid plate) parallel to the normal to the membrane with an angular standard distribution (σ) of 40° and a bacteriochlorophyll dimer/ubiquinone radical concentration ratio of 1.1. Dotted and dashed lines represent similar calculation with the molecular x- and y-axes, respectively, positioned normal to the membrane.

simulated curve. Variations in the $[BChl]_2^+/Q^-$ ratio have their most dramatic effects on the spectrum for these tilt angles when the magnetic field is 90° to the membrane normal. Fig. 4 shows this effect for ratios close to 1.1 while the tilt angles and standard distribution are maintained at the values used in Fig. 3.

The experimentally determined ratio of 1.1 is not unexpected. Iron-depleted AUT-e particles [15] and reaction centers [22] are both known to retain residual amounts of iron in active iron-quinone acceptor complexes. Irradiation of these reaction centers at room temperature will yield only the spectrum at $[BChl]_2^+$. Therefore, the total observed concentration of $[BChl]_2^+$ will always be greater than that of Q^-. A ratio of 1.1 corresponds to a retention of approx. 6% iron in the particles.

Therefore, changing the concentration ratio shifts the simulation up and down the line-width coordinate in Fig. 4. On the other hand, changing the tilt angles shifts the maximum of the simulation along the angular coordinate. Fig. 3 shows plots obtained from simulations where the Q^- principal x-, y- and z-axes were alternately positioned normal to the membrane. Again, for these simulations the simulation parameters of concentration ratio and angular distribution were held constant.

Finally, varying the magnitude of the standard angular distribution (as can be seen in Fig. 5) changes the extent of variation of the line-width with angle, i.e. the larger the distribution, the smaller the variation. This is understandable since the larger the distribution, the more the system looks like one which is randomly oriented. Furthermore, the larger the value of σ the more difficult it is to determine the exact orientation of the radical.

At this point two questions naturally arise concerning whether this is the primary quinone acceptor and whether it is oriented in the phosphatidylcholine multilayer the same as its parent structure in the chromatophore membrane. It is known [23] that *o*-phenanthroline blocks the transfer of an electron from

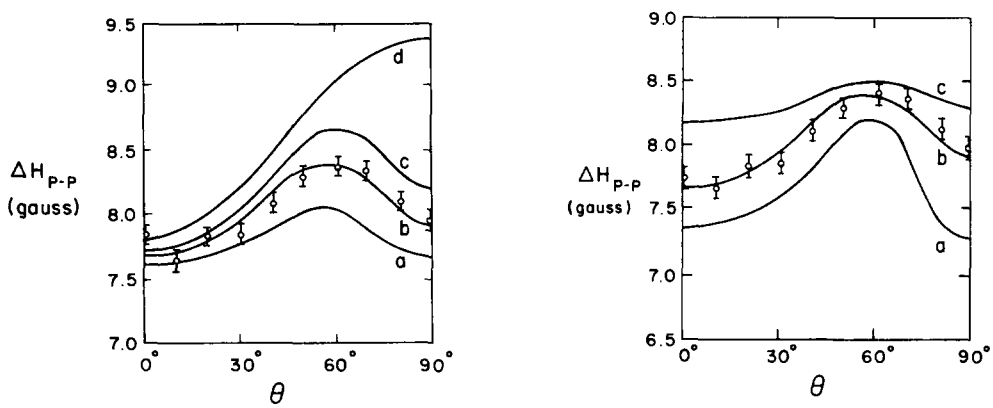


Fig. 4. Simulations of angle-dependent line-width variations of $[BChl]_2^+$-Q^- pair with tilt angle of x equal to 0° and $\sigma = 40°$. $[BChl]_2^+/Q^-$ concentration ratios equal to (a) 1.0, (b) 1.1, (c) 1.2 and (d) 1.3. Circles represent empirical data.

Fig. 5. Simulation of angle-dependent line-width variations of $[BChl]_2^+$-Q^- pair with tilt angle of x equal to 0° and the $[BChl]_2^+/Q^-$ concentration ratio equal to 1.1. Standard deviations (σ) about each tilt angle equal to (a) 30°, (b) 40° and (c) 50°.

the primary to the secondary quinone acceptor. The addition of *o*-phenanthroline (10^{-3} M) to our lecithin-bound-AUT-e prior to multilayer formation yielded angle-dependent line-width data identical to that shown in Fig. 3. Because we do not know the action of *o*-phenanthroline on AUT-e particles, it should be cautioned that these experiments merely suggest and do not definitely show that the observed quinone signal is due to reduction of the primary quinone.

Photolysis of samples in which Q had been partially chemically reduced with dithionite produced $[BChl]_2^T$ whose relative signal intensities mimicked those shown for oriented chromatophores. Part of this data is represented as triangles in Fig. 2. Therefore, the oriented primary quinone acceptor is in a phototrap complex whose relative orientation in the lecithin membrane is the same as its parent complex in the chromatophore. A final question concerning the orientation of Q^- which, unfortunately, cannot be answered at this time is whether extraction of Fe in the AUT-e preparation disturbed the orientation of Q. All that can be stated at this time is that the observed retention [13—15] of the structural integrity of the system implies that its structure has not been dramatically altered.

Discussion

Knowledge of the principal g factors of Q^- has allowed us to interpret the angular dependence of Q^- in phosphatidylcholine multilayers in terms of a well defined orientation. On the other hand, before the orientation of the bacteriochlorophyll dimer triplet can be unequivocally assigned, two properties of this system must be known. First of all, what is the structure of the dimer? Included in this question are the questions of whether or not the structures of the dimer triplet, the dimer cation and the normal ground state dimer are the same and how dependent is our interpretation of the orientation of this system on the assignment of this species as a dimer as opposed to a larger oligomer. Secondly, accepting a structure, what is the correlation between the spectral axis system labeled in Fig. 1 and the principal axis system of the triplet?

Unfortunately, our present knowledge of this system does not enable us to unambiguously answer any of these questions. We can, however, propose a model based on our present knowledge and interpretation of this system with the usual realization that our interpretation may change with time as does our knowledge. A popular structure of the dimer donor unit [24] is one in which the two porphyrins of the bacteriochlorophyll molecules are nearly parallel to one another. Such a structure would have an obviously principal axis system dictated by symmetry where one of the axes is perpendicular to the two porphyrin planes while the other two axes lie in a plane parallel to the two planes and midway between them. Which axis is perpendicular to the porphyrin planes? Unfortunately, only further experimentation on model systems will answer this question. On the other hand, there is some evidence [25] that suggests that our z-axis in Fig. 1 corresponds to this unique axis. For one thing, this is the preferred axis system of the chlorophyll monomer [26]. This should also be true for the dimer as long as the two porphyrin planes are nearly parallel. This axial assignment would also be in agreement with those described

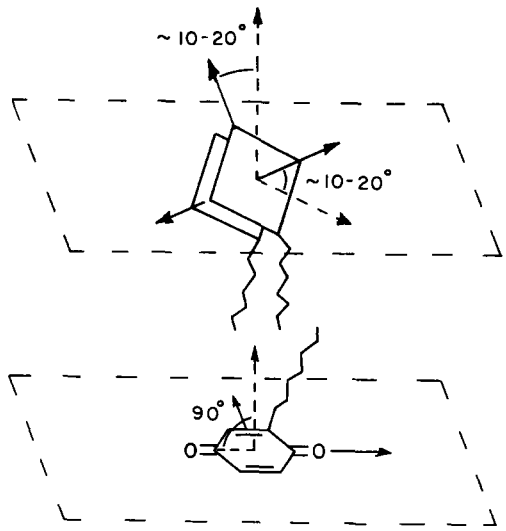


Fig. 6. Schematic representation of relative tilt angles of $[BChl]_2^T$ and Q^- in phototrap complex oriented in membrane bilayer as described in text.

by Vermeglio and Clayton [27] in their interpretation of the polarization of bacteriochlorophyll fluorescence in oriented multilayer systems. Accepting this assignment, we can draw a picture of the membrane-bound reaction center showing the orientation of the chlorophyll triplet and the ubisemiquinone anion relative to the plane of the membrane. This is done in Fig. 6.

Finally, it is interesting to examine our present knowledge of the orientation of various electron transport components in membrane systems. Virtually all biological electron transport components (e.g., chlorophylls, quinones, metaloporphyrins, flavins, ferrodoxins) have approximate planar symmetry. It is therefore the normal to the plane whose orientation is unequivocallly assigned. Is it not significant that many of the systems investigated so far [7—11] have assigned the direction of the normal to the radical plane either coincident with or perpendicular to the membrane plane? It may be that the orientation of each electron transport component is important in the basis mechanism of electron transport among them.

Acknowledgements

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