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Analysis of time-resolved emission spectra of oriented phycobilisomes

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Abstract

The polarized time-resolved (in ps range) fluorescence spectra of phycobilisomes obtained from cyanobacteria *Tolypothrix tenuis* embedded in poly(vinyl alcohol) films and oriented by film stretching have been analysed. Fluorescence spectra were deconvoluted on Gaussian components supposing the same positions of components maxima in three sets of time-resolved spectra taken in natural and polarized light. A good fit of the experimental and calculated spectra was obtained when using the following maxima: 580 and 595 nm in the phycoerythrin region, 634 and 650 nm in the phycocyanin region, 660 and 680 nm in the allophycocyanin region. The area under curve of the Gaussian component vs. time gives the shape of rise and decay of emission of chromophores contributing to the given component. These kinetics were analysed using several model functions. The experimental excitation profile was convoluted with a multiexponential model individually or "globally" e.g. assuming the same lifetime values for the given species in all sets of spectra. The Foerster–Hauser types of two- and three-dimensional models we also convoluted with excitation profile and fitted to the decay of primarily excited species. The first acceptor decay can be described well by the Foerster–Hauser models or by a monoexponential function. The accuracy of fit in either case of three- and two-dimensional Foerster–Hauser function is similar. The fluorescence rise and decay of the next species in a donor–acceptor chain can be analysed in terms of two or three exponential functions. Obtained lifetimes of fluorescence are similar to those reported in literature. The results suggest that there are more than one chain of excitation donors and acceptors in the phycobilisomes of cyanobacteria *Tolypothrix tenuis*.

Keywords: Time-resolved fluorescence; Excitation energy transfer; Phycobilisomes

1. Introduction

Phycobilisomes (PBS), antenna macrocomplexes occurring in cyanobacteria and red algae, are efficient transducers of excitation energy [1,2]. The polarized time-resolved (in the ps time range) fluorescence spectra of PBS from *Tolypothrix tenuis* embedded in poly(vinyl alcohol) (PVA) film and oriented by film stretching have been presented and discussed previously [3]. It is known [1–3] that in the investigated PBS the excitation energy migrates from excited phycoerythrin (PE), through phycocyanin (PC) to allophycocyanin

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(APC). Excitation was done at 546 nm (PE absorption region) and the time evolution of peaks belonging to various PBS biliproteins was observed. The kinetics of the rise and decay of the same peak obtained from two polarized spectra of partially oriented PBS are different. For example, an excitation energy transfer from excited PE to the final acceptor allophycocyanin (APC) is more efficient in the parallel polarized component of emission than in the perpendicular one.

The discussion of all sets of spectra shows that light absorbed by differently oriented PE molecules reaches different groups of final emitters. This finding suggests that there is more than one independent pathway of excitation energy transfer (ET) in PBS. This, consequently, is in agreement with suggestions from previous papers [4–6]. The discussion of the results presented in previous paper [3] was grounded on whole PBS emission spectrum in which the fluorescence bands of various biliproteins overlap strongly. More trustworthy results, allowing to draw conclusions concerning the mechanism responsible for energy attachment to the protein part of biliprotein [8]. As it follows from X-ray diffraction on crystalized biliproteins [11,12] their orientation in respect to PBS rods axis is also different. In oriented PBS different types of chromophores can give different contributions to each polarized component of the fluorescence spectrum.

We have tried several model fits of experimental spectra, starting from those known from literature [1,2,13,14] positions of emission maxima of various types of chromophores of every biliprotein. However, a good fit can be obtained by using less components than suggested in the literature. Because of a high number of analysed spectra the fitting to components of the same position of maxima in all sets of spectra is not quite arbitrary and therefore the result of such analysis can be treated as a piece of new information about the emission spectra. It is always much easier to fit a sum of Gaussians to an experimental curve using a larger number of components. Such a case can be analysed in several ways; thus the results are not univocal. Therefore, we decided to decompose the PBS spectrum only into six components common to the whole set of spectra recorded at various times after excitation of isotropic and oriented samples. The meaning of our components differs from that of s, m, f chromophores. Every component which belongs to a group of chromophores has similar positions of absorption band. Various types of chromophores (s, m, f) attached to a given type of biliprotein (PE, PC or APC) can contribute to such a component to a different degree. Recently, the time resolved spectra of PBS have been fitted by using various numbers of components [5] but the molecular origin of the proposed components was not clear. We obtained a good fit of the migration, can be obtained from the analysis of the time course of fluorescence components belonging to various biliproteins.

2. Methods of calculations

The time resolved spectra were measured at room temperature at excitation by 6 ps (FWHM) pulse and intensity at 546 nm of 10^8–10^9 photons/cm^2 per pulse, which is low enough to avoid non-linear effects. Due to the response of the detection system the observed excitation pulse was broadened to 70–80 ps (FWHM). The observed shape of the excitation pulse was taken as an apparatus curve and used later in the decay analysis. Details of the experimental methods have been given previously [3]. The time resolved spectra were registered starting from the time −39 ps (i.e. before the maximum of the laser peak was reached) till +1071 ps after the laser peak in three different (19, 40 and 79 ps) intervals of time (depending on the time elapsed after the laser peak; first when spectra were changing quickly, the used intervals were shorter, afterwards longer).

2.1 Spectral decomposition

In the result 24 spectra were recorded for each polarized component of a stretched and an unstretched sample, respectively. Then the spectra were analysed in order to separate contributions from individual chromophores. The shape of biliprotein components was assumed Gaussian. It

is generally accepted [7–12] that chromophores attached to each type of biliprotein can be divided into three groups (sensitizing-s, medium-m and fluorescent-f). Different features of chromophores are related to their various calculated and experimental spectra using the following positions of the component maxima: in the PE region 580 and 595 nm, in the PC region 634 and 650 nm, and in the APC region 660 and 680 nm. These spectral components will be denoted by the abbreviation of biliprotein name followed by the position of maximum in nm. Figure 1 shows a typical example of a Gaussian decomposition. The surface under the Gaussian component as a function of time gives the shape of the rise and decay of emission of chromophores contributing to the given component.

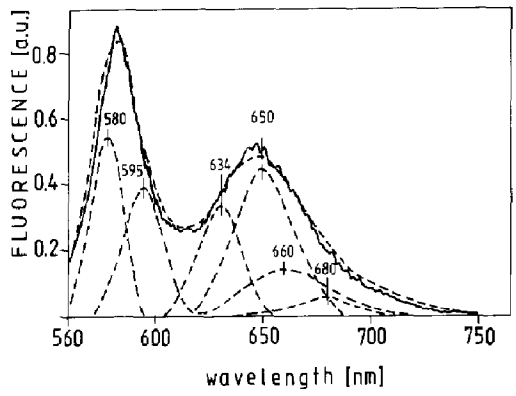

Fig. 1. Gaussian deconvolution of fluorescence spectrum of PBS from *Tolypothrix tenuis* taken 39 ps after the laser peak maximum (unstretched sample). Solid line denotes the experimental result, the dashed line the sum of Gaussian components.

2.2 Rise and decay analysis

Sets of such curves are presented in Figs. 2(a) and (b). Each chromophore in a donor–acceptor chain (except for the first one excited directly by laser light) is excited by energy transfer from the predecessor and can be deactivated by spontaneous radiative or nonradiative transitions and by excitation energy transfer to an acceptor molecule. The excited state decay by radiative and nonradiative spontaneous transitions is, for

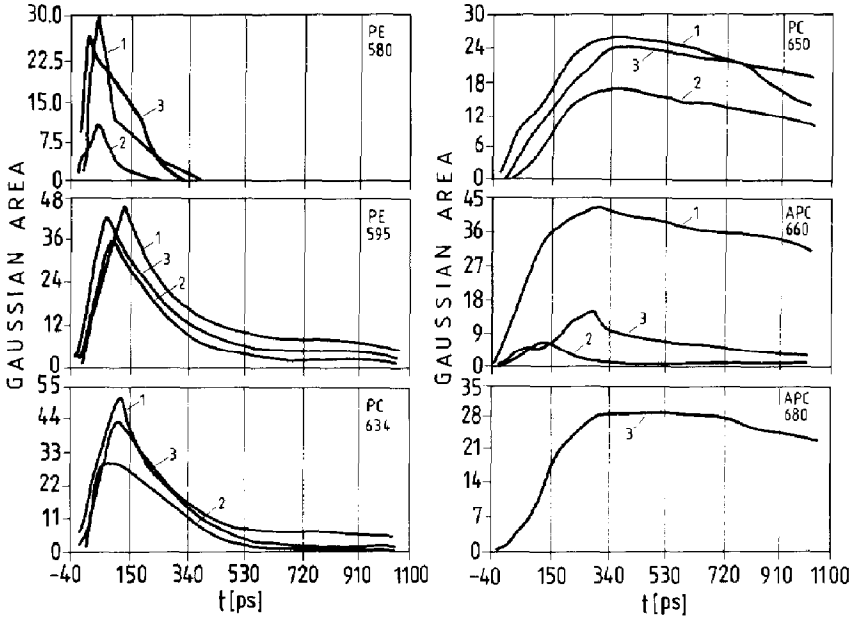

Fig. 2. (a) Time vs. change of the areas under the Gaussian components. Kinetics of rise and decay of fluorescence obtained for stretched samples: (1)–parallel component, (2)–perpendicular component; and unstretched samples (3)–unpolarized light. (b) Time vs. change of the areas under the Gaussian components. Kinetics of rise and decay of fluorescence obtained for stretched samples: ((1)–parallel component, (2)–perpendicular component) and unstretched (3)–natural light) samples.

one type of the isolated chromophore, monoexponential, having the shape $\alpha \exp(-t/\tau)$ where α is amplitude, t elapsed time, and τ lifetime of the excited state. In a case of superposition of emission from several (i) pools of chromophores (which can be chemically identical, but spectroscopically different because of various environments) the decay is the sum of exponential functions with various τ_i and α_i. In the case of Foerster-type energy transfer the rate coefficients depend on time and such simple kinetics does not hold [15]. In the cases of homogeneous three- and two-dimensional random distribution of acceptor molecules the donor decays (at δ function excitation) have the forms:

$$I_{D\delta} = I_{0\delta} \exp\Big[-t/\tau_D - c_A(4\pi^{3/2}/3) R_0^3 (t/\tau_D)^{1/2} \Big] \quad (1)$$

$$I_{D\delta} = I_{0\delta} \exp\Big[-t/\tau_D - c_A(4\pi/3) R_0^2 (t/\tau_D)^{1/3} \Big] \quad (2)$$

where c_A denotes the acceptor concentration, R_0 is a critical radius, and τ_D is the donor fluorescence lifetime. For the cases of three- and two-dimensional isotropic (chaotic) distributions of acceptor molecules, the decay of the first acceptor in the chain should be [16]:

$$I_A = \exp(-t/\tau_A) \int^x e^{u^2} \exp\Big[-4/3\pi^{3/2} c_A R_0^3 \times \{(1/\tau_D)/(1/\tau_A - 1/\tau_D)\}^{1/2} u \Big] du$$

where

$$x = \{(1/\tau_A - 1/\tau_D)t\}^{1/2} \quad (3)$$

$$I_A = \exp(-t/\tau_A) \int^y e^{u^3} \exp\Big[-4/3\pi c_A R_0^2 \times \{(1/\tau_D)/(1/\tau_A - 1/\tau_D)\}^{1/3} u \Big] du \quad (4)$$

where $y = \{(1/\tau_A - 1/\tau_D)t\}^{1/3}$, and τ_A is the acceptor fluorescence lifetime.

For the next acceptors in the chain the decay function is more complex. Several authors [17–20] fit the decays by an exponential of the form $\exp(-2kt^{1/2})$ which is in contradiction with the theoretical discussion given by Hauser and Wagenblast [15] and Hauser et al. [16] but can be derived from Foerster kinetics under the condition that the energy transfer occurs with very high efficiency [17,19,20]. Such a simple function is valid only when the donor fluorescence is quenched by energy transfer to a random neighborhood of acceptors under the additional assumption that in the kinetic equations the time-dependent rate is proportional to $t^{-1/2}$ [21]. In PBS the donors and acceptors have specific mutual orientations and distances. Therefore, the supposition of a random distribution of acceptors seems to be inappropriate. This belief is supported by the fact that analysis of the time course of PBS fluorescence in terms of multiexponential decay gives, in many cases, also an excellent fit [5,21–23]. Taking into account that it is not yet univocally established that only one chain of acceptor and donor molecules is responsible for the excitation energy transfer in PBS [1,3,4] it is not easy to predict which approximation, the one-, two- or three-dimensional system of acceptors, is closer to the real situation. Therefore we tried several model functions using exponential functions and the Foerster–Hauser type functions (eqs. 1 and 2). The Foerster–Hauser type model function was convoluted with the experimental excitation profile and fitted to the observed decay of primarily excited species (PE-580). This decay was treated as excitation to the next type of chromophore in a donor–acceptor chain (PE-595) and than the procedure was repeated.

The observed excitation profile (apparatus curve) was also convoluted with the proposed mono- and multiexponential models with free parameters α and τ. These parameters were adjusted by a non-linear least squares method until the best fit was achieved. The fit was judged both visually and by the value of the sum of squares of deviations (SSD). Such a procedure was applied to all individual decay curves. In a "global" approach we were simultaneously fitting fluorescence decays of the same species from three different spectral sets (unstretched sample in natural light and two polarized components of stretched samples) assuming that the lifetimes in all three sets are the same. The "global" approach seems to be reasonable because the chro-

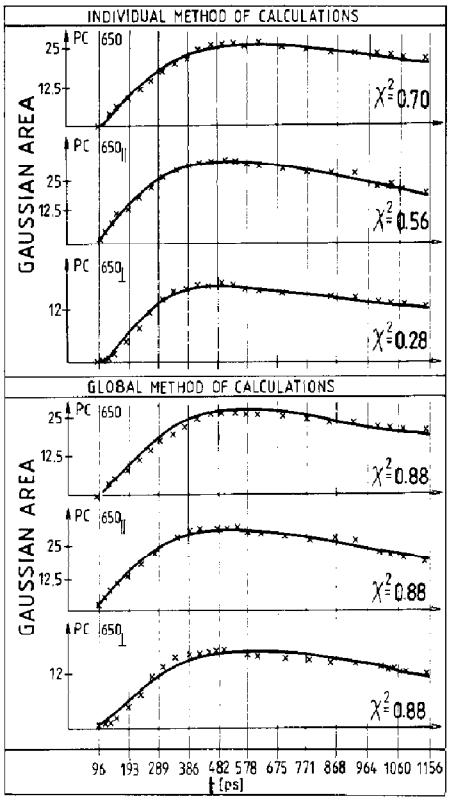

Fig. 3. The individual and global analysis on exponential functions of the fluorescence kinetics of PC-650. Polarization of light marked on the graphs.

mophore surroundings in all cases are the same. It has been previously shown [3] that the PBS in stretched PVA are oriented without the shape deformation. Therefore, the fluorescence decay rates of each type of chromophore have to be identical in unstretched and stretched samples.

As it follows from Fig. 3, representing the PC-650 decay analysis by individual and global methods, it is easier to obtain a good fit using the former of these methods.

3. Results of calculations and discussion

The results of calculations are gathered in Table 1 (individual analysis), Table 2 (global analysis) and Table 3 (Foerster–Hauser model functions). Figure 4 presents the fit of the time course of emission of PE-595 to three-exponential functions, and to three- and two-dimensional systems of acceptors of excitation energy (eqs. 1 and 2). A similar situation was obtained for PE-580 (Table 3). The quality of fit resulting from the sum of exponential functions is better than that obtained from using eqs. (1) or (2). In the Foerster–Hauser type functions, however, a lower number of adjustable parameters is applied than in the case of the sum of two- or three-exponential functions. The decay of directly excited pigment (PE-580) can be described only by one exponential function (Tables 1 and 2) or by using the Foerster–Hauser type function (eqs. (1) or (2), Table 3). This is very interesting because the formulas ap-

Table 1

The individual method calculations data

Gaussian component	Unstretched films			Stretched films					
	τ(ps)	α_u	SSD	τ(ps)	$\alpha_\parallel$	SSD	τ(ps)	$\alpha_\perp$	SSD
PE-580	73.8	0.011	29.46	74.7	0.014	12.11	45.5	0.040	1.47
PE-595	378.4	0.009	5.88	315.7	0.014	17.8	394.6	0.008	11.42
	239.6	0.020	6.67	115.8	0.059	16.3	463.3	0.070	2.97
	22.5	−0.036	6.67	65.0	−0.060	16.3	–	–	–
PC-634	70.9	0.197	1.10	67.6	0.430	2.8	–	–	–
	58.5	−0.214	1.10	61.6	−0.433	2.8	–	–	–
	646.3	0.05	1.10	3785.1	0.020	2.8	–	–	–
PC-650	1136.3	0.01	0.53	602.4	0.023	0.56	1346.3	0.05	0.28
	227.6	−0.009	0.53	293.0	−0.021	0.56	112.5	−0.006	0.28
APC-660	582.9	0.007	0.61	2319.9	0.012	3.59	597.6	0.002	0.53
	131.2	−0.008	0.61	97.8	−0.013	3.59	49.4	−0.004	0.53
APC-680	1717.4	0.010	0.94	–	–	–	351.0	0.011	0.65
	135.9	−0.012	0.94	–	–	–	226.6	−0.012	0.65

Table 2

The global method calculations data

Gaussian component	τ(ps)	α_u	$\alpha_\parallel$	$\alpha_\perp$	SSD
PE-580	72.92	0.011	0.015	0.003	12.8
PE-595	38.97	−0.026	−0.042	−0.032	3.91
	90.97	0.021	0.038	0.027	3.91
	728.34	0.004	0.005	0.003	3.91
PC-634	232.30	0.020	0.018	0.011	11.41
	20.53	−0.040	−0.016	−0.013	11.41
	64.70	0.685	0.617	0.165	2.04
	60.91	−0.703	−0.621	−0.166	2.04
	801.38	0.050	0.030	0.040	2.04
PC-650	854.60	0.012	0.013	0.008	0.88
	228.33	−0.012	−0.011	−0.008	0.88
	151.56	0.836	0.207	−0.048	0.57
	152.48	−0.846	−0.216	0.041	0.57
	997.86	0.010	0.111	0.006	0.57
APC-660	55.91	−0.004	−0.01	0.0001	2.46
	2447.94	0.003	0.011	0.0001	2.46
	167.68	−1.085	−1.452	−0.409	0.99
	168.97	1.082	1.447	0.409	0.99
	−18436.00	0.001	0.008	0.0001	0.99
APC-680	1627.72	0.010	–	0.002	1.27
	135.97	−0.012	–	−0.002	1.27
	243.82	0.410	–	0.183	0.83
	237.98	−0.417	–	−0.184	0.83
	6277.99	0.006	–	0.0001	0.83

ply to the first donor decay. The decays of further chromophores in the donor–acceptor chain are theoretically described by very complex formulas (for example eqs. 3 and 4 for the first acceptor) which cannot be easily applied directly in a fitting procedure. In both two- and three-dimensional Hauser–Foerster model functions the goodness of fit is similar. Therefore it is not possible to decide which configuration is more suitable.

Table 3

Data obtained by using Foerster–Hauser model functions

	Fractal dimension	τ(ps)	b	SSD	w
PE-580	2	90.91	0.150	10.02	0.33
	3	100.00	0.084	10.03	0.50
	"t^w"	90.91	0.120	10.03	0.39
PE-595	2	200.00	−0.046	10.63	0.33
	3	200.00	−0.142	10.22	0.50

The yields of nonradiative and radiative spontaneous deactivations of excited biliproteins in immobilized PBS are higher than those of PBS in organisms or in fluid medium [3]. Therefore, the yields of excitation energy transfer between biliproteins in PBS in polymer film are less efficient than those in organisms. For one type of chromophores to deactivate spontaneously, the decay curve has to be monoexponential. The energy transfer is a bimolecular process which leads to decay curves which have been in several cases well approximated by functions of the form: $\exp(-T^{1/2}/\tau)$ [5,17–20]. The theoretical formula (1) contains both factors. In native PBS the yield of ET is very high. Consequently, for biliproteins included in donor–acceptor chains the main way of deactivation of excitation is ET. But even in

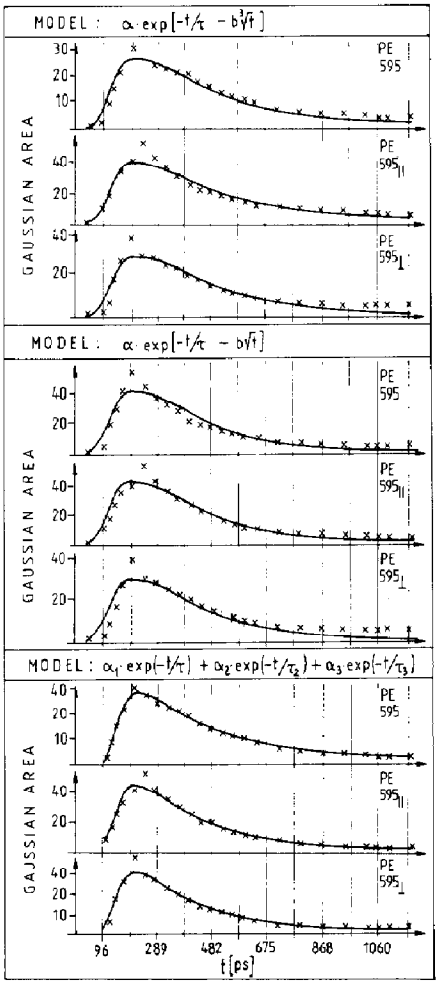

Fig. 4. The comparison of the analysis of PE-595 fluorescence decay using Foerster–Hauser models for three- and two-dimensional systems and three exponential functions.

organisms the deactivation by radiative and non-radiative transitions cannot be neglected. Thus, formulas (1) and (2) have to be applied rather than other approximations.

The decay of PE-580, which is directly excited by laser light, can be well described by this formula (Fig. 1 and Table 3). Assuming that PE-580 is a source of its excitation the PE-595 can also be described by the Foerster–Hauser formula (Table 3). We also assumed that the power of time "w" in formula:

$$I_a = \alpha \exp(-t/\tau - bt^w) \qquad (5)$$

is unknown. The best fit of experimental data to such an approximation was obtained at $w = 0.39$. However, the quality of the fit was similar to that in the case of two- and three-dimensional models (Table 3). By using exponential functions, the decay of PE-580 can always be (in both individual and global analysis) described by a monoexponential function (Tables 1 and 2). In most cases $\tau = 74$ ps is observed, only the decay of the perpendicular component in individual analysis is shorter ($\tau = 45$ ps). Porter et al. [17] have reported a τ of about 70 ps for PE in PBS. Wendler et al. [24] have obtained a similar value ($\tau = 60$ ps). Therefore, the value obtained by us is in good agreement with literature data. The emission related with PE-595 rises with 39 ps time and decays with 91 ps and 728 ps components according to global analysis (Table 2).

In individual analysis in terms of a sum of two exponential functions, the faster rise time and longer decay have been found to differ in stretched and unstretched films with a rather low fitting accuracy (Table 1) (SSD values can be compared only in the same set of data). Many authors [24,25] have reported long-time decays of biliproteins in PBS and in biliproteins complexes. Much different values of τ (from about 1 ns to almost 3 ns) were reported for long-time components. These slow decaying emissions are related to chromophores not involved in the energy transfer (ET) process or having low efficiency of ET. The long-living fluorescence with τ lower than 1 ns has been obtained not only for PE-595 but also for PC-634 and PC-650 as a result of global analysis of three components (Table 2). In all these components the intensity is low showing that the number of chain chromophores isolated from ET, is rather low.

Previously [3] a shift in PE maximum from 582 nm to 585 nm occurring in 70 ps time has been found. The laser light (546 nm) excites predominantly shortwave length chromophores of PE. The observed shift of emission maximum was interpreted as a redistribution of excitation energy between various PE chromophores. Now (from Table 2) it seems that the time of redistribution of energy between short- and long-wavelength PE chromophores is shorter about 40 ps. Sandstrom et al. [26,27] have found that a rapid energy transfer channel exists between PC chro-

mophores, with transition times from 27 ps to 57 ps depending on the type of chromophores. There are several similarities between PC and PE structure. Thus the rise time of PE-595 fluorescence of $\tau = 39$ ps that we have obtained seems to be reasonable. Also decay times of PE-595 fluorescence obtained by global analysis (91 ps and 728 ps) are similar to those earlier reported for biliproteins [2627]. The process of energy transfer from PE to PC was investigated by Yamazaki et al. [20] and Mimuro et al. [28] by comparing the time-resolved emission of cyanobacteria cultured in green light (PE-rich) and in red light (PE-less). The presence of PE influences the kinetics of ET not only between PE and PC but also between PC and APC [28]. These effects are explained by PC chromophores heterogeneity. In PE-rich *Tolypothrix tenuis*, the PC emission maximum is reached in 50 ps time. For isolated PBS embedded in PVA [3] the delay time of the PC peak (i.e. the time in which this peak reaches its maximal value) was 238 ps. From both Table 1 and Table 2 it follows that after separation of contributions from various biliproteins the rise time of PC-634 emission (which is related with ET between PE and PC) is about 60 ps. There is much more literature data concerning emission decay of PC than PE decay [9,21,25–27]. The ET between various PC chromophores seems to be rather slow (the rise time of PC-650 emission in individual and global approach by two components analysis is about 230 ps, at three components about 150 ps (Tables 1 and 2)). Sandstrom et al. [26,27] have found the intermediate lifetime of PC aggregates to be about 300 ps in monomers and about 100–120 ps in trimers, fast ET ranging from 27 ps to 57 ps and a long-living component with τ about 1 ns. Wendler et al. [9] have reported only two components: one about 40 ps and the second weaker about 1 ns for R-PC. Values of reported τ of separated biliproteins and biliproteins in PBS [8] depend on the source of PBS, methods of preparation and mathematical analysis. Contributions from various chromophores (s, m, f) are different in various spectral regions and therefore the results strongly depend on wavelengths of excitation and fluorescence [22,23]. In our analysis also τ obtained for short- and long-wavelengths regions are markedly different. Hefferle et al. [22,23] have shown that different aggregated forms of biliproteins exhibit decays in three time ranges: short 30–120 ps, intermediate 140–370 ps and long 1500–2200 ps. Table 1 shows that the emission of stretched samples is strongly anisotropic in most spectra regions. As a result of the normalization procedure applied in global calculations, the polarized component amplitudes α are more closely related to experimental fluorescence intensities in individual than in global analysis. It is possible to compare $\alpha_{\parallel}$ and $\alpha_{\perp}$ in a case of similar $\tau_{\parallel}$ and $\tau_{\perp}$. Therefore, information about emission anisotropy values can be obtained on the basis of Table 1 data.

Emission anisotropy changes over time. PE-580 and some PC components exhibit negative emission anisotropy. As it follows from previously measured linear dichroism (LD), the absorption of PBS in 580 nm region is strongly negative [3]. It remains still, though less, negative in PE-595 and PC regions. In these regions the observed LD can be a superposition of negatively and positively polarised components. This is also supported by the results of our analysis for PC-634 and PC-650 emissions presented in this paper. For APC the parallel component of polarized emission is much higher than the perpendicular one, whereas the LD is negative. This is possible when differently oriented forms of APC exhibit various yields of fluorescence. Again we have suggested [3] that these forms are pools of differently oriented final emitters.

4. Conclusions

We have shown that the first acceptor (PE-580) can be fairly well described by monoexponential decay. The first acceptor decay can be equally well described by the Foerster–Hauser formula (1). The accuracy of fit in the three-dimensional case (eq. 1) and two-dimensional case (eq. 2) is similar. Therefore it is not possible to decide which model is better suitable for describing energy transfer in phycobilisomes. For other than first acceptor (PE-580) biliproteins, at least a sum of two-exponential functions for decay analysis is

needed. The Gaussian analysis and the analysis of time-resolved spectra show that for each type of biliprotein (PE, PC, APC) at least two pools of chromophores exist characterised by different kinetics of risc and decay of fluorescence. This suggests that there is more than one chains of excitation donors and acceptors. Mimuro et al. [28] supposed there are different longer or shorter paths of energy migration. The length of these paths depends on the type of chromophores involved in the energy transfer process. In some cases the energy is rather randomly distributed between the chromophores belonging to the same biliprotein, whereas in other cases the energy is more likely transferred to other biliproteins than distributed between chromophores belonging to the initially excited biliprotein. Our results are in agreement with Mimuro et al.'s supposition.

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