Excitation Energy Transfer among Chlorophyll a Molecules in Polystyrene: Concentration Dependence of Quantum Yield, Polarization and Lifetime of Fluorescence

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Electronic excitation energy transfer was studied for chlorophyll a in a solid solution of polystyrene by measuring the concentration quenching of quantum yield, polarization, and lifetime of fluorescence. The concentration quenching of the experimental fluorescence quantum yield is adequately described by Kelly and Porter's empirical formula (Proc. Roy. Soc., Lond. **A 315**, 149, 1970), and of polarization of fluorescence by the Jablonski theory (Acta Phys. Pol., **14**, 295, 1955). With increasing concentration of chlorophyll a, the fluorescence peak at 672 nm (mainly monomer) is red-shifted, the intensity of the emission peak at ∼730 nm (mainly aggregate) relative to that at the shorter wavelength is increased. The $\overline{R_0}$ values, calculated by using total concentrations, for the emission at 672 nm and 730 nm are 73 ± 2 Å and 45 ± 1 Å, respectively. This may suggest that the chlorophyll monomers have a greater efficiency of energy transfer than the aggregates, which fluoresce at ∼730 nm.

The photophysical primary process of photosynthesis involves the non-radiative transfer of electronic excitation energy from the light-harvesting (antenna) chlorophyll a molecules to the photochemical traps (reaction centers). The mechanism of this energy transfer in photosynthetic systems is not entirely understood (see Knox [1, 2]). To study this process, various model systems (see Seely [3]) have been used to simulate the state of chlorophyll *in vivo*: chlorophyll monolayers [4 — 6], chlorophyll incorporated in detergent micelles [7 — 9], lipid vesicles [10], bilayers [11], chlorophyll in solid solutions of cholesterol [12], lecithin [13, 14], and polystyrene [15 — 17], and chlorophyll covalently bound to polymers [18].

Since chlorophyll *in vivo* is in a relatively rigid matrix, the polystyrene systems is considered to be a good model for the antenna system. However, quantitative data for this system are scarce. Gorshkov [15] found that for ∼10^{-4} M chlorophyll a the degree of polarization of fluorescence greatly decreased when the temperature was lowered from 290 to 6 K. Vacek et al. [16] reported a flattening of the fluorescence polarization spectrum when the chlorophyll a concentration was increased. Vacek

et al. [17] have presented an absorption spectrum (peaks at 432 and 669 nm; [Chla], 0.23 mM; 297 K), an emission spectrum (peaks at 675 and 730 nm; [Chla], 0.77 mM; 77 K), and the degree of polarization (7.8%; λ-excitation, 633 nm; λ-observation, 730 nm; [Chla], 0.77 mM; 297 K) of Chla in polystyrene. Gorshkov and Vacek et al. invoked the presence of chlorophyll a aggregates to explain their results. Thus, to further understand and evaluate the use of this system — chlorophyll in polystyrene as a model for energy transfer in photosynthesis — we report here quantitative measurements on chlorophyll a fluorescence quantum yield, polarization, and lifetime.

Methods

Samples

Chlorophyll a was extracted and chromatographically purified as described by Skorkovská and Vavrinec [19]. The polystyrene foils containing chlorophyll were prepared as described by Vacek et al. [16, 17]: chlorophyll a and polystyrene were dissolved in acetone, spread on a glass plate and dried in an inert atmosphere. The average concentration of chlorophyll in the amorphous polystyrene matrix was estimated from the average thickness of the films and the measured optical density at 668 nm, using a molar extinction coefficient of $73.4\,\mathrm{mM^{-1} \cdot cm^{-1}}$ (estimated from the data of Seely and Jensen [20]).

Absorption and Emission Spectra

Absorption spectra were measured with a Cary 14 recording spectrophotometer. These spectra are plotted as $\varepsilon(\bar{\nu})/\bar{\nu}$ as a function of $\bar{\nu}$, where $\bar{\nu} =$ wavenumber at which absorbance was measured and $\varepsilon(\bar{\nu})$ is extinction coefficient at different $\bar{\nu}$; the peak was normalized to a value of 1.0. Plotting the absolute values did not seem meaningful as it is dependent on the sample thickness. Since chlorophyll concentration, the extinction coefficient, and the path length are known, optical densities (O.D.) can be easily obtained if necessary.

The emission spectra were measured with a spectrofluorometer described by Shimony *et al.* [21]. Front surface fluorescence was detected with an S-20 photomultiplier (EMI 9558B) through a Corning CS 2-58 glass filter and a Bausch and Lomb monochromator (model 33-86-45 − 0.5 meter; 600 grooves/mm; blazed at 750 nm; linear dispersion, 3.3 nm/mm). Excitation was through an interference filter with peak transmission at 635 nm (half-maximum bandwidth, 8 nm). The emission spectra were corrected for photomultiplier sensitivity and monochromator (model 33-86-45 − 0.5 meter; 600 grooas $F(\bar{\nu})/\bar{\nu}^3$ as a function of $\bar{\nu}$, where F is the fluorescence intensity, after normalization to 1.0 at the emission peak. Deconvolution of the spectra was done as described by Vacek *et al.* [22]. The absorption and the emission spectra, plotted in the form they have been plotted here, could be used to later check the mirror symmetry principle and to calculate the overlap integral between the donor fluorescence and acceptor absorbance. This, however, was not done in this paper.

Fluorescence Polarization

Light from a quartz-iodine lamp (200 W, GE Q 6.6 AT4/CL) was focused on the entrance slit of a Bausch and Lomb grating monochromator (model 33-86-45 − 0.5 meter; 600 grooves/mm; linear dispersion, 3.3 nm/mm); the monochromatic beam was then collimated with a lens system and passed through a Glan-Thompson polarizer ($12 \times 12 \times 24$ mm, Karl Lambrecht Corp., Chicago, IL.) (Fig. 1). The fluorescence from the sample was passed through a second Glan-Thompson polarizer and appropriate optical filters (see later), and detected by an S-20 photomultiplier (EMI 9558 B). The anode current (I) of the photomultiplier was converted into voltage (V), using an operational
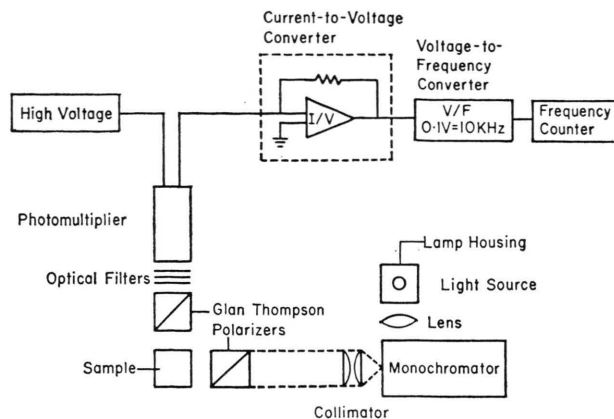

Fig. 1. Schematic of laboratory-assembled fluorescence polarization spectrometer (see text).

amplifier, then to frequency (F), using a voltage to frequency converter (Anadex Instruments Inc., Model DF-110R), and the digital signal was displayed on a frequency counter (Hewlett Packard, Model 5382A) with a gating time of 10 s.

The results reported here are for the following combinations of excitation wavelength and optical filters: (a) excitation at 655 ± 0.9 nm for fluorescence detected through a Schott cut-off filter RG 655 (3 mm, thickness) and a 673 nm interference filter (half-maximum bandwidth, 13.2 nm); and (b) excitation at 655 ± 1.7 nm for fluorescence observed through Schott filter RG 10 and 730 nm interference filter (half-maximum bandwidth, 8.4 nm).

The optical detection system showed systematic unequal sensitivities to vertically and horizontally polarized light. A multiplicative correction factor, which empirically normalizes the response of the system to equal sensitivities for the two orientations of polarization, was applied to the measurement of horizontally polarized light. This factor was obtained from the quotient (G) of the vertically (F_{HV}) to the horizontally (F_{HH}) polarized components of fluorescence ($F_{HV}/F_{HH} \equiv G$), from a 10^{-7} M solution of rhodamine B in glycerol excited with horizontally polarized light at 546 ± 1.7 nm; the fluorescence was observed through a Corning CS 3-66 glass filter. The degree of polarization of fluorescence is given by the relation:

$$p = \frac{F_{VV} - G(F_{VF})}{F_{VV} + G(F_{VH})} \tag{1}$$

where, F_{VV} and F_{VH} are the vertically and horizontally polarized components of fluorescence from the sample using vertically polarized excitation.

Fluorescence Lifetime (τ)

τ was measured by the phase-delay method using a mode-locked He-Ne laser ($\lambda = 632.8$ nm) modulated at a frequency of 75 MHz (incidence irradiance, 40 mW·cm^{-2}) as described by Merkelo *et al.* [23]. The detector was an S-1 photomultiplier (RCA 7102) tube. The fluorescence lifetime was calculated from:

$$\tau = \frac{\tan \Delta\Phi}{2\pi f} \qquad (2)$$

where, $\Delta\Phi$ = phase delay between the incident light and fluorescence, and f = modulation frequency. The $\Delta\Phi$ was measured as the phase delay between the laser radiation scattered from the sample and the chlorophyll fluorescence through a Schott RG 5 glass cut-off filter (3 mm thickness) after adjusting the photon flux of the scattered radiation by neutral density filters to give the same photomultiplier anode current as the fluorescence.

Results and Discussion

Absorption and emission spectra at 293 K

To characterize our system, we measured the absorption spectra (in the red region) and the emission spectra for four concentrations of chloro-

phyll a in amorphous solid solutions of polystyrene. The main features of our results are illustrated in Fig. 2 by the absorption spectrum for a sample containing ~ 400 μM chlorophyll a and the emission spectra of those with ~ 100 μM and ~ 2.7 mM chlorophyll a.

At low chlorophyll a concentrations (up to 430 μM) the absorption peak corresponding to the Qy transition is at 668 ± 0.5 nm (14970 ± 11 cm^{-1}). At high concentrations (~ 10 mM) this band is slightly red shifted and broadened with all the increase on the long wavelength side (also see Monchor and Vacek [24]). However, the band at ~ 617 nm appears relatively unaffected even at a chlorophyll concentration of 2.7 mM. The ratio of the absorbance at 700 nm to that at 617 nm changed from 0.11 at a chlorophyll concentration of 430 μM to 0.14 at 2.7 mM — an increase of $\sim 25\%$. These observations suggest the appearance of a long wavelength absorption species when the average chlorophyll concentration in polystyrene is increased. In addition, it is known that increasing the average chlorophyll a concentration leads to the appearance of a new band at 454 nm in the linear dichroism spectra which can be attributed with high probability to the appearance of chlorophyll dimers [24].

The emission peak at low concentrations ($\lesssim 100$ μM) of chlorophyll a is at 672 ± 0.5 nm ($14,881 \pm 11$ cm^{-1}) having a Stokes' shift of 3.5 ± 1 nm (77 ± 22 cm^{-1}); a band at 727 ± 0.5 nm ($13,755 \pm 9$ cm^{-1}) is also observed. Upon increas-
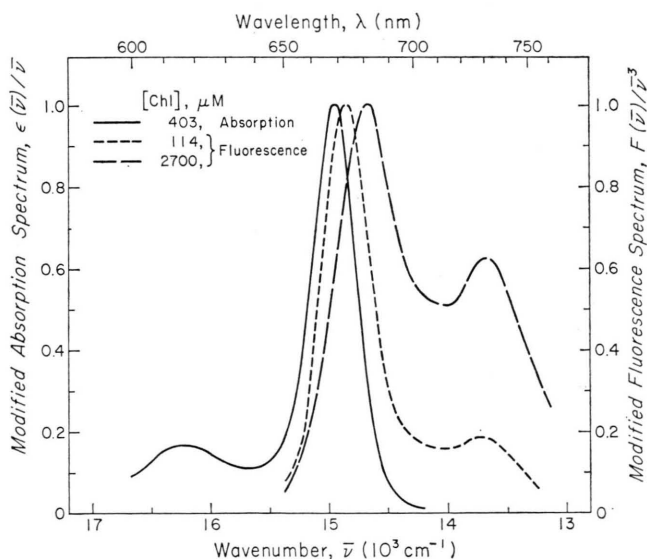

Fig. 2. Absorption and emission spectra of chlorophyll a in polystyrene at room temperature. The absorption spectrum was plotted as $\varepsilon(\bar{\nu})/\bar{\nu}$ as a function of wavenumber ($\bar{\nu}$) and normalized to 1.0 at the absorption peak, whereas the emission spectra were plotted as $F(\bar{\nu})/\bar{\nu}^3$ as a function of $\bar{\nu}$ also normalized to 1.0 at their emission peaks; $\varepsilon(\bar{\nu})$ = extinction coefficient at $\bar{\nu}$; $F(\bar{\nu})$ = fluorescence intensity at $\bar{\nu}$. (———), absorption spectrum, [chlorophyll] = 403 μM; (— — —) and (— —), emission spectra for 114 μM and 2.7 mM respectively.

ing the average chlorophyll concentration, the peak at 672 nm is red shifted, and the long wavelength peak is increased. A mathematical deconvolution of the emission spectrum at a chlorophyll concentration of 770 μM gives the bands with the following peak locations and bandwidths at half height: 675.7 ± 0.1 nm $(14799 \pm 2$ cm$^{-1})$, 308 ± 8 cm^{-1}; 685 ± 3 nm $(14599 \pm 64$ cm$^{-1})$, 810 ± 60 cm^{-1}; and 730 ± 1 nm $(13699 \pm 19$ cm$^{-1})$, 809 ± 49 cm^{-1}. The above results can best be explained as follows: with increasing average chlorophyll concentration a greater chlorophyll aggregation results, with the long wavelength aggregates fluorescing at ~ 730 nm.

To understand the role of monomeric and aggregated forms of chlorophyll a in energy transfer, we then measured the relative quantum yield, the degree of polarization, and the lifetime of fluorescence for different concentrations of chlorophyll a in polystyrene.

Relative quantum yield

The mechanism(s) for the concentration quenching of the relative fluorescence quantum yield (Φ) must be established from quantitative studies (Table I, Fig. 3). Our results for the concentration dependence of the relative quantum yield of fluorescence at 673 nm (open squares, Fig. 3; vertical column 4 in Table I) are in qualitative agreement with those of Kelly and Porter [13] for chlorophyll

in lecithin. A satisfactory fit of our experimental data was obtained with their equation:

$$\frac{\Phi}{\Phi_0} = \frac{1}{1 + (C/C_{1/2})^2} \tag{3}$$

where, Φ is the measured quantum yield, Φ_0 is the quantum yield in the absence of quenching, C is the pigment concentration, and $C_{1/2}$ is the half-quenching concentration. The theoretical curve (long and short dashes) in Fig. 3 was obtained with $C_{1/2} = 151.5$ μM corresponding to an average nearest neighbor distance of 123 Å in a random distribution of chlorophyll molecules. (We note that for chlorophyll a in lecithin Kelly and Porter [13] reported $C_{1/2}$ to be $\sim 10^{-3}$ M. We believe that the difference may be due to the different system used.) The explanation of the second power dependence on concentration of quenching is the direct excitation of non-fluorescing pairs of molecules, as in ref. [14]. The "pairs" of molecules or "spatial dimers" need not be chemically bonded complexes, but could be two molecules which, in a random distribution, happen to be sufficiently close to cause quenching when one or the other is excited. (For other details, the reader is referred to refs. [13] and [14].) The above mechanism suggests de-excitation rates one or two orders of magnitude higher than fluorescence decay, and would facilitate the non-radiative dissipation of the energy of certain absorbed quanta with virtually no competition from fluorescence. In addition,

Table I. Fluorescence parameters for chlorophyll a in amorphous solid solution of polystyrene at 293 °K.

Sample	Chlorophyll [μM]	673 nm				730 nm		
		D[Å][a]	Φ[b]	P[c]	$\tau/\tau_{(0)}$[d]	Φ	P	$\tau/\tau_{(0)}$
I	11.1	294	1.000	0.886	1.10	1.000	1.015	0.80
II	114	135	0.643	0.937	1.1	0.460	–	0.96
III	403	89	0.123	0.779	1.0	0.185	0.926	0.98
IV	430	87	–	–	–	–	0.913	–
V	2 700	47	0.030	0.283	0.66	0.093	0.668	0.64
VI	11 610	29	–	0.298	0.38	–	0.215	0.41

[a] Random distribution nearest neighbor distance (see, Chandrasekhar [29]) $D = 0.55396\, n^{-1/3}$ where n = concentration expressed in molecules per cm^3.

[b] Relative experimental fluorescence quantum yield, $\Phi = F/I_0(1 - 10^{-\mathrm{OD}})$, where F = measured fluorescence intensity in arbitrary units, I_0 = exciting intensity, and OD = optical density at 635 nm. Excitation was at 635 (half-maximum bandwidth, 8 nm) and fluorescence was detected with a resolution of 3.3 nm.

[c] Probability of fluorescence being emitted by initially excited molecule, $P = (1/p_0 - 1/3)/(1/p - 1/3) = r/r_0$, where p = degree of polarization of fluorescence, p_0 = intrinsic polarization (p when $c \to 0$), r = anisotropy of fluorescence, related to p by the relation $r = 2p/3 - p$, and r_0 = intrinsic anisotropy related to p_0.

[d] Relative lifetime, $\tau/\tau_{(0)}$, where τ = measured lifetime and $\tau_{(0)}$ = lifetime in the absence of concentration quenching ($c \to 0$). $\tau_{(0)}$ is assumed to be equal to 5.6 ns (see also ref. [30]).
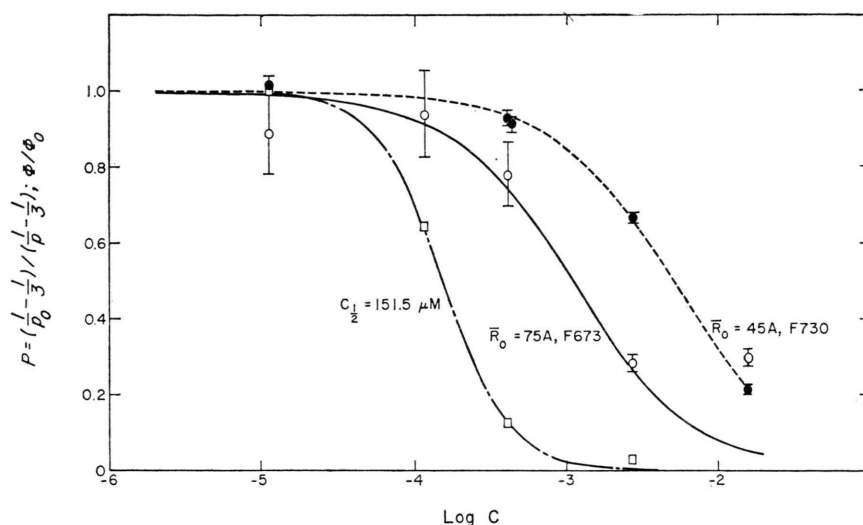
Fig. 3. Plot of P or Φ/Φ_0 against log C. $P = (1/p_0 - 1/3) / (1/p - 1/3)$, where P = probability that the fluorescence is emitted by an initially excited molecule, p_0 = intrinsic degree of polarization as concentration (C) approaches zero, and p = measured degree of polarization at various chlorophyll concentrations; Φ = measured quantum yield of fluorescence and Φ_0 = quantum yield of fluorescence in the absence of quenching; $C_{1/2}$ = half-quenching concentration; $F\lambda$ = fluorescence at the wavelength indicated in nm; and $\overline{R}_0$ = distance between donor and acceptor, in Å, at which the probability of energy transfer is equal to the probability of de-excitation by all other processes — greater $\overline{R}_0$ means more efficient energy transfer. ($\bigcirc$), experimental P(673 nm); (————), theoretical curve from the modified Jablonski equation [28] with $\overline{R}_0 = 75$ Å. ($\bullet$), experimental P(730 nm); (— — —), theoretical curve with $\overline{R}_0 = 45$ Å. The error bars represent the standard deviations for the least squares straight line in the p^{-1} against C used to obtain P_0. ($\square$), experimental Φ/Φ_0; (— · —), theoretical curve using the Kelly-Porter equation [13] with $C_{1/2} = 151.5$ μM.

quenching of fluorescence quantum yield may also result from energy transfer from excited monomers to non- or weakly fluorescent aggregates. The latter may also be implied by the concentration dependence of the relative fluorescence quantum yield at 730 nm (vertical column 7, Table I), which does not obey Eqn. (3) — the relative emission yield at high concentrations (*e. g.* 2.7 mM) is much higher than that predicted by Eqn. (3) using the data obtained for lower chlorophyll concentrations. Clearly, the fluorescence quantum yield has a complex dependence on the chlorophyll concentration, and is not a suitable parameter for quantifying energy transfer between like molecular species.

Degree of polarization of fluorescence

For a randomly oriented set of molecules in a rigid suspension, the degree of polarization of fluorescence is one of the most important sources of information on energy transfer between like molecules. The fluorescence from the initially excited molecules contribute to almost all of the polarization. We, thus, investegated the concentration dependence of the degree of polarization of fluorescence at 673 nm (open circles, Fig. 3) and 730 nm (closed

circles, Fig. 3). The intrinsic polarization (p_0) is obtained from the least-squares straight line in a plot of p^{-1} against C by extrapolating the line to $C = 0$. Our experimental values were $p_0 = 0.300$ and 0.193 for fluorescence at 673 nm and 730 nm, respectively. The probability (P) that the fluorescence is emitted by an initially excited molecule is given by:

$$P = \frac{1/p_0 - 1/3}{1/p - 1/3}. \qquad (4)$$

This is equivalent to r/r_0 where r = anisotropy of fluorescence related to p as $r = 2p/3 - p$ and r_0 = intrinsic anisotropy related to p_0. According to the suggestion of Knox [25] and Craver and Knox [26], we have displayed our results as a plot of P against log C (Fig. 3). The theoretical curves (solid and dashed lines) in Fig. 3 were generated from Jablonski's equation [27] rewritten in the form:

$$p = \frac{2(\nu + 1 - 3^{-\nu})}{\nu^2} \qquad (5)$$

where, $\nu = (1.3)^3 C/C_0$ (see Knox [25] and Bojarski [28]) and $C_0 = ((4/3)\pi \overline{R}_0^3)^{-1}$, where R_0 is the intermolecular separation when the probability of energy transfer equals the probability of de-excita-

tion by all other processes. The most striking feature of the fluorescence polarization results is that the calculated $\overline{R}_0$ for the fluorescence at 673 nm ($\overline{R}_0 = 73 \pm 2$ A) is much larger than that at 730 nm ($\overline{R}_0 = 45 \pm 1$ A). If the 673 nm and 730 nm emissions at high chlorophyll concentrations originate mainly from monomeric and aggregate forms, the above suggests that $\overline{R}_0$ for the monomeric chlorophyll a is 73 ± 2 Å and that for the chlorophyll aggregates is < 45 Å, since part of the fluorescence at 730 nm originates from monomers. In other words, chlorophyll a monomers have a greater efficiency of energy transfer than the aggregates. This conclusion may have to be modified if recalculation of $\overline{R}_0$, based on actual concentrations of aggregates, becomes available. An important points to note is that the data point for P for F 673 at the highest concentration used does not match the theoretical curve; it is much higher than the theoretical curve. We explain this to be due to the competition of energy transfer to the aggregate form (at his high chlorophyll concentration) that leads to a decrease in energy transfer among monomer forms and to contribution of aggregate fluorescence at 673 nm at this concentration.

The half quenching concentration for fluorescence depolarization is higher than $C_{1/2}$ for fluorescence yield for chlorophyll a in polystyrene. On the other hand, for chlorophyll b in lecithin Kelly and Patterson [14] observed $C_{1/2}$ for fluorescence yield to be higher than the half quenching concentration for fluorescence depolarization. We do not know the reasons for this difference.

Lifetime of excited states

To test whether the monomeric and aggregated forms of chlorophyll a in polystyrene have different fluorescence lifetimes we measured the latter at 673 and 730 nm as a function of the average chlorophyll concentration (or random distribution nearest neighbor distance [29]) in the films. The two lifetimes are found to differ by $< 20\%$ (lower at 730 nm); at low chlorophyll concentrations ($\sim 10-100$ μM, samples I and II, Table I), a lifetime of ~ 5.5 ns is obtained (*cf.* ref. [30]). Increasing the chlorophyll concentration to 400 μM does not change the lifetime, but at 2.7 mM and above the fluorescence lifetime is indeed lowered, presumably, as a result of the greater competition between non-radiative energy transfer and fluorescence. If energy transfer between these randomly oriented chlorophylls in polystyrene

is by the Förster mechanism, then the pairwise transfer rate is inversely proportional to the sixth power of distance. Increasing the average concentration of chlorophyll, thus, decreases the average distance (see Table I) and increases the energy transfer rate. If all other rates are unaffected, the lifetime of fluorescence decreases.

Table I (*cf.* vertical columns 4 with 6 and 7 with 9) shows that decrease in τ with increasing concentration of chlorophyll a in polystyrene is much less than the decrease in Φ. This clearly indicates the formation of non-fluorescent aggregates. Similar experimental results were obtained by Kelly and Patterson [14] for chlorophyll b in lecithin. Furthermore, the $\sim 20\%$ lower τ for F 730 indicates that there are also weakly fluorescent aggregates. These aggregates also increase with increasing concentration of chlorophyll a. At the highest concentrations it appears that even τ of F 673 is drastically reduced, due to competition of energy transfer to aggregates with those among monomers and/or to increased contribution of fluorescence from the aggregates. [It may be coincidental that τ (F 730) is the same as τ (F 673) at the highest concentrations used.]

Concluding remarks

In conclusion, the concentration dependence of the fluorescence yield at 673 and 730 nm suggest the existence of at least three molecular species: monomers (fluorescing mainly at ~ 672 nm), weakly fluorescing aggregates (fluorescing at 730 nm), and non-fluorescent "spatial dimers". The relative increase in the longwave emission peak (~ 730 nm) with concentration, and the possible different values of $\overline{R}_0$ at 673 nm ($\overline{R}_0 = 73 \pm 2$ Å) and at 730 nm ($\overline{R} = 45 \pm 1$ Å) support the idea that chlorophyll a aggregates exist and fluoresce at 730 nm at room temperature in a solid solution of polystyrene. The monomers may have a greater efficiency of transfer than the aggregates. Our results show that the electronic properties of chlorophyll a in polystyrene are very similar to those of chlorophyll in other solid solutions [12, 13] and in detergent micelles [7−9]. We conclude that chlorophyll a in a solid solution of polystyrene is as effective as other model systems in the study of electronic excitation energy transfer in photosynthesis, but with the advantage that the electronic properties are stable over a longer period of time and over a wider range of temperatures.

G. R. Seely (personal communication) has pointed to us that further calculations, based on the knowledge of fractional absorption by monomers, dimers and aggregates, may charge some of the interpretations of data presented here. In particular, caution should be excercised in the interpretation of the different $\bar{R}_0$s calculated here for F 672 and F 730.

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