*Photochemistry and Photobiology* Vol. 33, pp. 103 to 108

0031-8655/81/0101-0103$02.00/0

# ACTION SPECTRA OF CATION EFFECTS ON THE FLUORESCENCE POLARIZATION AND INTENSITY IN THYLAKOIDS AT ROOM TEMPERATURE

DANIEL WONG* and GOVINDJEE†

Departments of Physiology, Biophysics and Botany, University of Illinois, Urbana, IL 61801, USA

(*Received* 11 *June* 1980; *accepted* 22 *July* 1980)

**Abstract**—The degree of polarization of chlorophyll-*a*(Chl-*a*) fluorescence is known to monitor the extent of excitation migration and/or the orientation of the photosynthetic pigment molecules. We report here the effects of cations, at room temperature, on the degree of polarization of Chl-*a* fluorescence, and fluorescence intensity in thylakoids as a function of excitation wavelength. Observations of maxima at 650 and 675 nm in the cation-induced changes in the excitation spectrum for fluorescence at 730 and 762 nm, and, in the action spectra for the depolarization of fluorescence lead us to suggest that the regulation of the initial distribution of excitation to photosystem II involves the better coupling of Chl-*b* and -*a* in the light harvesting complex with Chl-*a* in the reaction center II complex.

## INTRODUCTION

Mono- and divalent cations concurrently affect several primary photoprocesses in thylakoids with the largest effect on the excitation energy distribution from photosystem (PS)II to PSI (Wong *et al.*, 1978a; Wong, 1979). It is generally accepted that the excitation energy transfer in the photosynthetic system is by Förster's inductive resonance mechanism (Duysens, 1964; Förster, 1965, 1967; Knox, 1975, 1977). The most direct method for measuring changes in the excitation energy migration among the same type of molecules is that of the degree of depolarization of fluorescence (see Michel-Villaz, 1976). This method for the study of cation-induced changes in energy migration was used by Wong and Govindjee (1979) [see also Govindjee and Wong (1979)]. These authors measured cation effects on the polarization of Chl-*a* fluorescence at wavelengths selected to monitor preferentially PSII or PSI emission; it was shown that $Na^+$ added to salt-depleted thylakoids increases the degree of polarization of Chl-*a* fluorescence at 685 nm (mainly PSII) and decreases that at 712 nm (PSI); this effect was fully reversed by the subsequent addition of $Mg^{2+}$. This is taken as evidence for the cation-induced regulation of excitation distribution at room temperature. In this paper, we report the effects of cations on the degree of polarization of Chl-*a* fluorescence and fluorescence intensity at 730 or 762 nm (to avoid artifacts due to scattering of excitation light) as a function of the excitation wavelength. The ratio excitation spectrum for fluorescence at 730 and 762 nm, at 296 K, shows relative maxima at ~650 and ~675 nm, when the sample containing $Mg^{2+}$ is compared with that without $Mg^{2+}$. Simultaneously, the relative depolarization of fluorescence between the samples shows maxima at ~650 and ~676 nm. On the basis of results presented here, and, preliminary results included in Wong and Govindjee (1979), we suggest that the regulation of initial distribution of excitation to PSII involves the better coupling of light harvesting complex with Chl-*a*II.

## MATERIALS AND METHODS

Thylakoid membranes were prepared from pea leaves as described elsewhere (Wong *et al.*, 1978a). These were suspended in 100 m*M* sucrose and 0.4 m*M* Tris–HCl (pH 7.2). The $Na^{2+}$ samples contained 10 m*M* $Na^+$, whereas, $Mg^{2+}$ samples contained in addition 10 m*M* $Mg^{2+}$. Chlorophyll concentration was determined by the method of Mackinney (1943), the action spectra of Chl-*a* fluorescence was measured as described by Gasanov *et al.* (1979), and the degree of polarization of Chl-*a* fluorescence as described by Wong *et al.* (1978b). The degree of polarization of fluorescence $P$ is obtained as follows:

$$P = \frac{F_{vv} - G \cdot F_{vh}}{F_{vv} + G \cdot F_{vh}}$$

where, $F_{vv}$ and $F_{vh}$ are the vertically and horizontally polarized components of fluorescence from the sample using vertically polarized excitation, and G is the instrumental correction factor, equal to $F_{hv}/F_{hh}$, where, $F_{hv}$ and $F_{hh}$ are the vertical and horizontal components of fluorescence when a 0.1 $\mu M$ solution of rhodamine B is excited with horizontally polarized light at 547 ± 1.7 nm, the fluorescence being detected through a Corning CS3-66 glass filter.

Certain methodological questions had to be answered before we could interpret our present results. These included reproducibility, concentration, temperature, and fluorescence level dependences.

*Reproducibility.* The degree of polarization ($P$) of fluorescence showed excellent reproducibility ($P$ uncertainty, ±0.0015), for different aliquots of the same thylakoid preparation. For thylakoids from different chloroplast isolations, however, slight variations ($P$ uncertainty, ±0.003) in the degree of polarization of fluorescence were observed.

*Present Address: c/o Miles Labs., P.O. Box 70, Elkhart, IN 46515, USA.

†To whom correspondence should be sent: 289 Morrill Hall, Department of Botany, University of Illinois, Urbana, IL 61801, USA.

Table 1. Concentration dependence of the cation effects on the polarization of fluorescence

| $[Na^+]$ (m$M$) | $[Mg^{2+}]$ (m$M$) | $P$(F > 694 nm) (%) |
|---|---|---|
| 0 | 0 | 3.0 |
| 2 | 0 | 3.7 |
| 4 | 0 | 3.4 |
| 6 | 0 | 3.5 |
| 8 | 0 | 3.4 |
| 10 | 0 | 3.4 |
| 10 | 5 | 2.7 |
| 10 | 10 | 2.4 |
| 0 | 0 | 3.0 |
| 0 | 2 | 2.2 |
| 0 | 4 | 2.5 |
| 0 | 6 | 2.3 |
| 0 | 8 | 2.3 |
| 0 | 10 | 2.2 |
| 5 | 10 | 1.9 |
| 10 | 10 | 2.0 |

Excitation was at 600 ± 8.3 nm, and fluorescence was detected through a Schott RG 10 glass filter. Thylakoids from pea chloroplasts were suspended at pH 7.6; [Chl] = 5 μg/mℓ; [DCMU] = 5 μ$M$; temperature = 23°C; medium, 100 m$M$ sucrose + 0.2 m$M$ Tris–HCl.

*Concentration dependence.* The $Na^+$-induced increase and the $Mg^{2+}$-induced decrease in the degree of polarization of the total Chl-*a* fluorescence [Schott RG 665 filter; Fluorescence (F) > 650 nm] saturates when the cation concentrations reach 4–6 m$M$ (Table 1). Note also that when $Mg^{2+}$ is added to thylakoids containing 10 m$M$ $Na^+$, the fluorescence polarization assumes the value of the sample with $Mg^{2+}$. The converse, however, does not hold, demonstrating that when equal concentrations of $Na^+$ and $Mg^{2+}$ are added the $Mg^{2+}$ effect dominates.

*Temperature dependence.* The degree of polarization of fluorescence from thylakoids under a particular cationic condition (salt-depleted, $Na^+$, or $Na^+ + Mg^{2+}$) is relatively constant between 0 and 25°C (see also Weber, 1960) showing that the mono- and divalent cation effects are temperature independent over the range 0–25°C (Table 2).

*O and P level dependence.* The cation effects on the fluorescence polarization occur at both the 0 (initial) and the P (maximum) levels of Chl-*a* fluorescence transient measured at 686 nm (Table 3).

*Emission wavelength dependence.* Wong and Govindjee (1979) have already shown the effects of $Na^+$ and $Mg^{2+}$ on the polarization of Chl-*a* fluorescence at 686, 712, 730 and 762 nm. For the present study, we have chosen 730 and 762 nm as the emission wavelength of observation to facilitate the measurements of the action spectra over both the Chl-*b* and Chl-*a* absorption bands. The degree of polarization of F762, $P$(F762), was found to decrease with $Mg^{2+}$ addition to the $Na^+$ samples at all excitation wavelength ($\lambda_{ex}$) in the range $600 \leqslant \lambda_{ex} < 700$ nm.

The rationale for the choice, at room temperature, of using the measuring wavelengths at 685 (F685), 712 (F712), 730 (F730) and 760 nm (F760) are as follows. Based on experiments on fluorescence spectra of physically separated pigment systems PSI and II (Briantais, 1969; Gasanov *et al.*, 1979), of unseparated chloroplast preparations excited preferentially with PSI or PSII light (Govindjee and Yang, 1966) and of algae during fluorescence transient (Lavorel, 1963), the following assignments have been made (see also, Wong, 1979): F685 is the major band of PSII; F712 has the highest value for the ratio of PSI/PSII emissions; and F730 and F760 are both from the vibrational satellite band of the main F685 band of PSII. The observation that the cation effects on $P$(F762) were similar to that on $P$(F686) and $P$(F730) (Wong and Govindjee, 1979) suggested that F762 reflects the fluorescence characteristics of Chl-$a_{II}$.

All experiments reported here were in the pH range 7.0–7.5, and the temperature range 21–26°C. All other details are presented in the legends of the tables and the figures. However, for a complete description of Materials and Methods, and, instrumental tests, see Wong (1979).

## RESULTS

A decrease in the degree of polarization of fluorescence ($P$) is suggested to mean an increase in excitation energy migration as was done to explain the 3(3,4 dichlorophenyl 1,1,dimethylurea (DCMU-)-induced decreases in $P$ of Chl-*a* fluorescence (Mar and Govindjee, 1972. Becker *et al.*, 1976; Van Rensen *et al.*, 1978; Wong, 1979); DCMU, simultaneously, causes an increase in the fluorescence yield. On the other hand, an increase in $P$ as a result of the addition of dinitrobenzene (DNB) has been explained to be due to decreased energy migration in thylakoids (Lavorel and Joliot, 1972). Figure 1 shows a relationship between $F^0/F$ (where $F^0$ is fluorescence intensity in the absence of DNB, and F in the presence) and the degree of polarization of Chl-*a* fluorescence in thylakoids. Based on the above, we assume that increases

Table 2. Temperature dependence of the cation effects on the intensity and polarization of fluorescence at 686 nm

| Temperature (°C) | Salt-depleted F686 | Salt-depleted $P$ | 10 m$M$ NaCl F686 | 10 m$M$ NaCl $P$ | 10 m$M$ NaCl + 10 m$M$ $MgCl_2$ F686 | 10 m$M$ NaCl + 10 m$M$ $MgCl_2$ $P$ |
|---|---|---|---|---|---|---|
| 1.3 | 57 | 2.0 | 39 | 3.2 | 100 | 2.2 |
| 4.3 | 57 | 2.0 | 39 | 3.2 | 98 | 2.2 |
| 9.5 | 56 | 2.0 | 38 | 3.2 | 95 | 2.2 |
| 14.5 | 53 | 2.1 | 37 | 3.2 | 91 | 2.2 |
| 19.4 | 49 | 2.2 | 35 | 3.4 | 87 | 2.3 |
| 25.0 | 45 | 2.2 | 33 | 3.5 | 82 | 2.3 |

Fluorescence excited with light at 633 ± 6.6 nm, and detected through interference filter at 686 nm (full width at half maximum, 6.4 nm). Samples were pea thylakoids suspended in the usual medium (see the legend of Table 1), pH 7.6. [Chl] = 5 μg/mℓ; [DCMU] = 5 μ$M$. Temperature was regulated to better than ±0.5°C. $P$ denotes the degree of polarization, and F the intensity of the fluorescence.

Table 3. Cation effects on the degree of polarization at the initial (0) and maximum (P) levels of fluorescence transient at 686 nm

| Sample | P(F686) (%) 0 Level | P Level |
|---|---|---|
| Salt-depleted | 3.9 | 3.7 |
| + 5 m*M* NaCl | 4.8 | 4.1 |
| + 5 m*M* NaCl + 5 m*M* $MgCl_2$ | 3.6 | 3.1 |

Fluorescence excited at 600 ± 6.6 nm, and detected with 686 nm interference filter (full width at half maximum, 6.4 nm) plus Schott RG 665 filter. Samples consisted of pea thylakoids suspended in the usual medium (see the legend of Table 1) at pH 7.2; temperature = 23°C.

and decreases in *P* reflect decreases and increases in excitation energy migration; we report here the cation-induced effects on *P* and on the intensity of Chl-*a* fluorescence in thylakoids as a function of excitation wavelength.

*Excitation wavelength dependence of the cation effects on the degree of polarization of chl-a fluorescence*

The degree of polarization of Chl-*a* fluorescence, measured at 762 nm (F762), as a function of excitation wavelength for the $Na^+$ and $Na^+ + Mg^{2+}$ samples are shown in Fig. 2. It is clear that the two curves are different (the errors are indicated by the size of the points shown). A clear decrease in the degree of polarization of Chl-*a* fluorescence at ~675 nm, upon the addition of 10 m*M* $Mg^{2+}$, is observed in this spectrum; a decrease at 650 nm is also noticeable, but, it becomes more obvious when the ratio of *P* $(Na^+ + Mg^{2+})/P$ $(Na^+)$ is plotted. Figure 3 shows the relative difference in the degree of polarization of F762 between the $Na^+$ and $Mg^{2+}$ sample and the $Na^+$ sample expressed as $P$ $(Na^+ + Mg^{2+}) - P(Na^+)/P(Na^+)$. Here, negative bands at 650, 675 and 685 nm are obvious suggesting that $Mg^{2+}$ induced an increase in deploraization of fluorescence, i.e. an increase in energy migration to Chl-*a* species of PSII; possible changes in the orientation of Chl-*a* species absorbing around 685 nm will be discussed later.

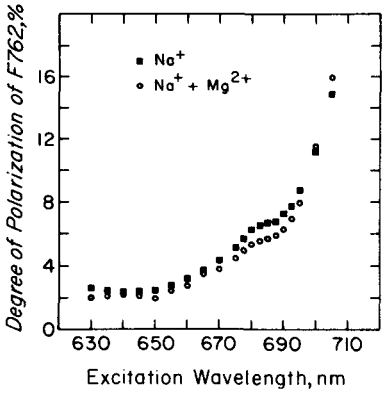


Figure 2. Degree of polarization of fluorescence at 762 nm (full width at half maximum, 11.3 nm) vs excitation wavelength (full width at half maximum, 5 nm). Sample details as given in the legend of Table 1. [$Na^+$], 10 m*M*; [$Mg^{2+}$], 10 m*M*.

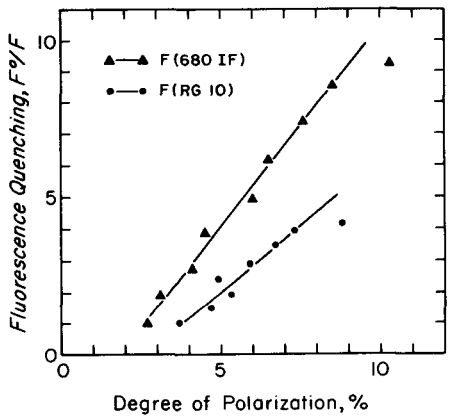


Figure 1. Linear relation between the extent of quenching and the degree of polarization of fluorescence in pea thylakoids. Excitation at 600 nm (full width at half maximum, 8.8 nm); fluorescence detected through Schott RG 10 glass filter or interference filter (IF) at 680 nm (full width at half maximum, 14 nm); Chl concentration, 5 μg/m*l*; medium, 100 m*M* sucrose, 0.4 m*M* Tris–Hcl, pH, 7.0; temperature, 23°C. Different points were obtained by treating thylakoids with different concentrations (0–3.2 m*M*) of dinitrobenzene (DNB).

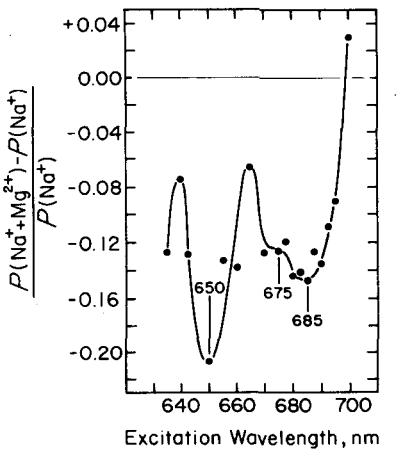


Figure 3. Excitation spectrum of the relative difference in the degree of polarization of F762 between the $Na^+ + Mg^{2+}$ sample and the $Na^+$ sample expressed as $[P(Na^+ + Mg^{2+}) - P(Na^+)]/P(Na^+)$. The excitation bandpass (full width at half maximum) was 5 nm. The fluorescence was detected through a combination of a Schott RG 10 glass filter and an interference filter at 762 nm (full width at half maximum, 11.3 nm). Sample details are as given in the legend of Tables. (Also see Wong and Govindjee, 1979.)

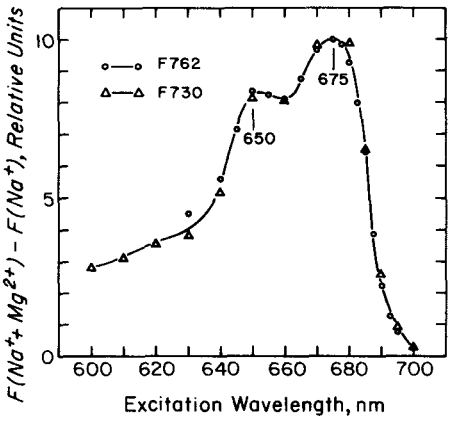


Figure 4. Difference excitation spectrum, between $Na^+$ + $Mg^{2+}$ and $Na^+$ samples for fluorescence at 730 and 762 nm, normalized at 675 nm. Experimental details as given in the legend of Fig. 3.

*Excitation wavelength dependence of the cation effects on the fluorescence intensity*

The ratio action spectra for $F(Na^+ + Mg^{2+})/F(Na^+)$ as a function of excitation wavelength measured at 730 nm (F730) and at 762 nm (F762) for the samples used in Fig. 2 show a general increase at all wavelengths, a prominent peak at 650 nm and a shoulder at 675 nm (not shown). The amplitude of $Mg^{2+}$-induced changes are, however, best observed (see Fig. 4) in difference spectra $(F(Na^+ + Mg^{2+}) - F(Na^+))$ measured as a function of excitation wavelengths at 730 nm (F370) and at 762 nm (F762). These difference spectra, in contrast to the ratio spectra for both F730 and F762, are identical and show the two bands at 650 and 675 nm.

The above results shown in Figs 2–4 are best explained in terms of (a) an increase in excitation energy transfer from the light harvesting Chl-*a*/Chl-*b* complex (LHC) to Chl-*a* II, and (b) a decrease in excitation energy transfer from PSII to PSI (see Discussion).

## DISCUSSION

*General*

The DCMU-induced increase in yield and depolarization of Chl-*a* fluorescence and the decline of the depolarization effect with increasing wavelengths of excitation (finally disappearing at ~680 nm) (Mar and Govindjee, 1972; Whitmarsh and Levine, 1974; Becker *et al.*, 1976; Wong, 1979) strongly support the concept that by keeping the PSII reaction centers closed, DCMU enhances the extent of energy migration among the less mutually aligned Chl molecules absorbing at wavelengths <680 nm. DNB, on the other hand, is an excellent quencher of Chl fluorescence *in vitro* (Livingston and Ke, 1950) and *in vivo* (Teale, 1960; Amesz and Fork, 1967; Lavorel and Joliot, 1972; Etienne *et al.*, 1974). Its influence in decreasing the extent of energy transfer is based on the kinetics of the photochemical 0 to P phase of the fluorescence transient (Lavorel and Joliot, 1972). The direct relation between fluorescence quenching and depolarization (Fig. 1) and their linear dependence on the concentration of DNB (Teale, 1960; Wong, 1979) demonstrate the inverse relation between the fluorescence yield and degree of polarization in the case of fluorescence quenching. The above known results with DCMU and DNB empirically support the occurrence in thylakoids of the inverse relation between the extent of energy transfer and the degree of polarization of the fluorescence. This concept forms the basis for the interpretation of the results presented here. However, a change in orientation may also cause a change in polarization of fluorescence (see below).

*A possible change in orientation of chl-*a *685 may cause quanta redistribution*

The concept that a change in the orientation of a few key Chl-*a* molecules can significantly alter the rate of energy transfer from PSII to PSI was suggested by Seely (1973). Biggins and Svejkovsky (1978) observed $Mg^{2+}$-induced changes in linear dichroism spectra of magnetically-oriented chloroplast membranes. They suggested that $Mg^{2+}$ causes a species of chromophores (Chl-*a* 690) to reorient closer into the plane of the thylakoid membrane. Since we have observed (Figs 3 and 4) a decrease in the polarization of fluorescence with 685 nm excitation upon $Mg^{2+}$ addition, this could be due to a decrease in the mutual order in the Chl-*a* 685 chromophore group(s). We shall not discuss Biggins' work any further because of the possibility of scattering and selective scattering artifacts (Latimer, 1959) in dichroism measurements (see also Schooley and Govindjee, 1976). Becker *et al.* (1976) have suggested that Chl-*a* molecules absorbing at > ~680 nm have a high mutual order, so that increased energy transfer between these molecules has little consequence on the fluorescence polarization. Thus, changes in polarization of fluorescence with excitation at 685 nm may be taken as a reduction of mutual orientation of Chl-*a* 685. Finally, since linear dichroism and fluorescence polarization spectral studies of (*cf.* Garab and Breton, 1976) suggest that the shorter wavelength forms of Chl-*a* (absorbing < ~670 nm) are less aligned with respect to the membrane plane, i.e. sustaining a greater angle with the plane, a reorientation of Chl-*a* 685 closer to the membrane plane would decrease the orientation factor (Förster, 1965) between Chl-*a* 670 (in PSII) and Chl-*a* 685 (in PSI) decreasing energy transfer from PSII to PSI.

*Possible changes in coupling of Chl-*a/b *complex (LHC) with Chl-*$a_{II}$

On the other hand, changes in polarization of fluorescence excited at shorter wavelengths (in Chl-*a* 670, Chl-*b* 650, etc.) have been suggested to be due to

changes in excitation energy transfer between Chl species with low mutual order (Michel-Villaz, 1976; Becker *et al.*, 1976). Thus, the negative peaks at ~650 nm (due to Chl-*b*) and at 675 nm (due to Chl-*a* 670) in the $Mg^{2+}$-induced changes in polarization of Chl-$a_{II}$ fluorescence (Figs 2 and 3) and positive peaks at 650 nm and 657 nm in the $Mg^{2+}$-induced increase in relative fluorescence yield (Fig. 4) may be interpreted to be due to an enhancement of energy transfer from these complexes (present in LHC) to fluorescent Chl-$a_{II}$. It is proposed that this is the process which increases the initial distribution of quanta from LHC to PSII. The observation that the relative enhancement of fluorescence of Chl-*b* is small (~6%) for F730 is consistent with previous findings (Butler and Kitajima, 1975a; Moya *et al.*, 1977; Wong *et al.*, 1978a, 1979) that the $Mg^{2+}$-induced variations in the sensitization of PSII fluorescence is $\leqslant 20\%$. These changes are, perhaps, manifestations of the proposed $Mg^{2+}$-induced increase in the coupling of LHC with Chl-$a_{II}$ (Arntzen and Ditto, 1976; Butler and Strasser, 1978; Paillotin, 1978).

The observation of peaks at ~650 and ~675 nm in Figs 3 and 4 leads to the first unambiguous suggestion of a $Mg^{2+}$-induced increase in LHC to Chl-$a_{II}$ transfer in normal thylakoids at physiological temperatures. A brief discussion of the two earlier studies on this subject is given below. Vernotte *et al.* (1973) showed that $Mg^{2+}$ causes no change in the action spectrum of PSII reactions while it inhibits PSI reactions with a spectrum having peaks at ~650 and ~670 nm. These results could have been interpreted equally well by a $Mg^{2+}$-induced inhibition of initial quanta distribution from LHC to Chl-$a_I$, as by a $Mg^{2+}$-induced inhibition of redistribution of quanta from PSII (initially transferred there from LHC) to PSI. Loos (1976), on the other hand, showed changes in the action spectra of the variable yield fluorescence and oxygen evolution (PSII) and $O_2$ uptake (Mehler reaction, PSI) at 480 nm, but this study could not have unequivocally distinguished between the involvement of Chl-*b* and carotenoids. Interpretations of the results in Figs 3 and 4 are, however, more definitive for two reasons: (1) By limiting the actinic light to wavelengths longer than 600 nm (exciting only the chlorophylls), complications of interpretations from the possible involvement of carotenoids in excitation energy transfer was avoided, and (2) by the simultaneous evaluation of the fluorescence intensity and polarization results, the possibility for ambiguities of interpretation of the type encountered with the data of Vernotte *et al.* (1973) was reduced. The importance of simultaneous evaluation of the fluorescence data, presented here, must not be overlooked. For instance, with only the fluorescence depolarization results, even if it could be independently established that $Mg^{2+}$ induces an increase in the extent of energy transfer, it would not be possible to conclude whether the change results from an increase in the initial partitioning of quanta to PSII, or an inhibition of their subsequent redistribution from PSII to PSI. However, if the inhibition of quanta redistribution were the only cause of the fluorescence polarization changes, the same relative enhancement of fluorescence intensity by $Mg^{2+}$ would be observed whether Chl-$a_{II}$ or LHC were excited. The observation in Fig. 4 that the action spectrum for the $Mg^{2+}$ enhancement of Chl-$a_{II}$ fluorescence intensity contains relative maxima at ~650 and ~675 nm implies more definitely that variations in the initial partitioning of energy are the result of changes in the energy coupling between LHC and Chl-$a_{II}$.

*Acknowledgement*—We thank the Biomedical Grant (1980–1981), provided through the School of Life Sciences, University of Illinois at Urbana, for financial support.

## REFERENCES

Amesz, J. and D. C. Fork (1967) *Biochim. Biophys. Acta* **143,** 97–107.
Arntzen, C. J. and C. L. Ditto (1976) *Biochim. Biophys. Acta* **449,** 259–274.
Becker, J. F., J. Breton, N. E. Geacintov and F. Trentacosti (1976) *Biochim. Biophys. Acta* **440,** 531–544.
Biggins, J. and J. Svejkovsky (1978) *FEBS Lett.* **89,** 201–204.
Briantais, J.-M. (1969) Doctoral Thesis, University of Paris-Sud (Orsay), France.
Butler, W. L. and M. Kitajima (1975) *Biochim. Biophys. Acta* **396,** 72–85.
Butler, W. L. and R. J. Strasser (1977) *Proc. Natl Acad. Sci. USA* **74,** 3382–3385.
Duysens, L. N. M. (1964) *Prog. Biophys. Mol. Biol.* **17,** 1–104.
Etienne, A. L., C. Lemasson and J. Lavorel (1974) *Biochim. Biophys. Acta* **333,** 288–300.
Förster, Th. (1965) In *Modern Quantum Chemistry*, Part III, *Action of Light and Organic Crystals* (Edited by O. Sinanoglu), pp. 93–137. Academic Press, New York.
Förster, Th. (1967) In *Comprehensive Biochemistry*: *Bioenergetics* (Edited by M. Florkin and E. H. Stotz), Vol. 22, pp. 61–80. Elsevier, Amsterdam.
Garab, G. I. and J. Breton (1976) *Biochem. Biophys. Res. Commun.* **71,** 1095–1102.
Gasanov, R., Z. K. Abilov, R. M. Gazanchyan, V. M. Kurbonova, R. Khanna and Govindjee (1979) *Z. Pflanzenphysiol.* **95,** 149–169.
Govindjee and D. Wong (1978) In *Proceedings of the Third International Seminar on Excitation Energy Transfer in Condensed Matter, Prague* (Edited by J. Fiala), pp. 19–28. Univerzita Karlova, Praha, Czechoslovakia.
Govindjee and L. Yang (1966) *J. Gen. Physiol.* **49,** 763–780.
Knox, R. S. (1975) In *Bioenergetics of Photosynthesis* (Edited by Govindjee), pp. 183–221. Academic Press, New York.

Knox, R. S. (1977) In *Primary Processes of Photosynthesis* (Edited by J. Barber), pp. 55–97. Elsevier, Amsterdam.
Latimer, P. (1959) *Plant Physiol.* **34,** 193–199.
Lavorel, J. (1963) *Colloq. Int. C.N.R.S.* **119,** 161–176.
Lavorel, J. and P. Joliot (1972) *Biophys. J.* **12,** 815–831.
Livingston, R. and C. L.-Ke (1950) *J. Am. Chem. Soc.* **72,** 909–915.
Loos, E. (1976) *Biochim. Biophys. Acta* **440,** 314–321.
MacKinney, G. (1943) *J. Biol. Chem.* **140,** 315–330.
Mar, T. and Govindjee (1972) In *Proceedings of the 2nd International Congress on Photosynthesis Research* (Edited by G. Forti, M. Avron and A. Melandri), Vol. I, pp. 271–281. Junk, The Hague.
Michel-Villaz, M. (1976) *J. Theoret. Biol.* **58,** 113–129.
Moya, I., Govindjee, C. Vernotte and J.-M. Briantais (1977) *FEBS Lett.* **75,** 13–18.
Paillotin, G. (1978) In *Proceedings of the 4th International Congress on Photosynthesis* (Edited by D. O. Hall, J. Coombs and T. W. Goodwin), pp. 33–44. The Biochemical Society, London.
Schooley, R. and Govindjee (1976) *FEBS Lett.* **65,** 123–125.
Seely, G.R. (1973) *J. Theoret. Biol.* **40,** 189–199.
Teale, F. W. J. (1960) *Biochim. Biophys. Acta* **42,** 69–75.
Van Rensen, J. J. S., D. Wong and Govindjee (1978) *Z. Naturforschung.* **33c,** 413–420.
Vernotte, C., J.-M. Briantais and P. Bennoun (1973) *C.R. Acad. Sci. Paris,* **277D,** 1695–1698.
Weber, G. (1960) In *Comparative Biochemistry of Photoreactive Systems* (Edited by M. B. Allen), pp. 395–411. Academic Press, New York.
Whitmarsh, J. and R. P. Levine (1974) *Biochim. Biophys. Acta* **368,** 199–213.
Wong, D. (1979) Ph.D. Thesis, University of Illinois, Urbana, IL.
Wong. D. and Govindjee (1979) *FEBS Lett.* **97,** 373–377.
Wong, D., Govindjee and P. Jursinic (1978a) *Photochem. Photobiol.* **28,** 963–974.
Wong, D., K. Vacek, H. Merkelo and Govindjee (1978b) *Z. Naturforschung.* **33c,** 863–869.
Wong, D., H. Merkelo and Govindjee (1979) *FEBS Lett.* **104,** 223–226.