NONLINEAR ABSORPTION OF CHLOROPHYLL-a

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Prospects and preliminary results of nonlinear absorption spectroscopy of chlorophyll are reported.

The nonlinear absorption is a field of laser spectroscopy. Like in the linear absorption spectroscopy the intensity change of a probing light beam travelling through a sample is measured. But whereas in the linear case the probing beam has a low intensity per definition (demand for validity of Beer's law), the range of nonlinearity begins at such intensities where measurable populations of excited states occur in this range the absorption (transmission) is a very informative function of intensity, reflecting populations, cross sections, and relaxations of excited states.

Our application ot the method of nonlinear absorption to the field of chlorophyll action in photosynthesis is directed on two problems:

I. Examination of the reality of subbands of the red absorption band of in-vivo-chlorophyll.

These subbands (up to now only result of the mathematical deconvolution of the overall bandshape) [1] are attached to different absorbing forms of antenna chlorophylls and play an important role in the discussion an construction of the photosynthetic unit.

If the conception is valid, in a nonlinear absorption measurement with tunable lasers the several subbands should have a transmission behavior independent of each other (inhomogeneous broadening behaviour).

II. Study of the primary processes of reaction centre chlorophylls. In this case the measurement and mathematical analysis of the nonlinear absorption over a wide range of intensity of in-vivo-chlorophyll at fixed wavelengths with favour the PS-centres can give information on excited state absorptions and relaxations in the reaction centre molecules.

To solve these problems, numerous preliminary investigations are necessary, including for example the stability behaviour of chlorophyll or intact plants under high intensity irradiation, including the overlap of spectral changes from antenna molecules and reaction centre molecules as well as the search for several causes of real or virtual inhomogeneous broadening.

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As a first step in this field, we investigated the nonlinear absorption of chlorophyll solutions, chloroplastsuspensions, and intact plants at the wavelength of the ruby laser (694 nm). The experimental device is shown in Fig. 1, parts of the results are shown in Fig. 2 and Fig. 3.

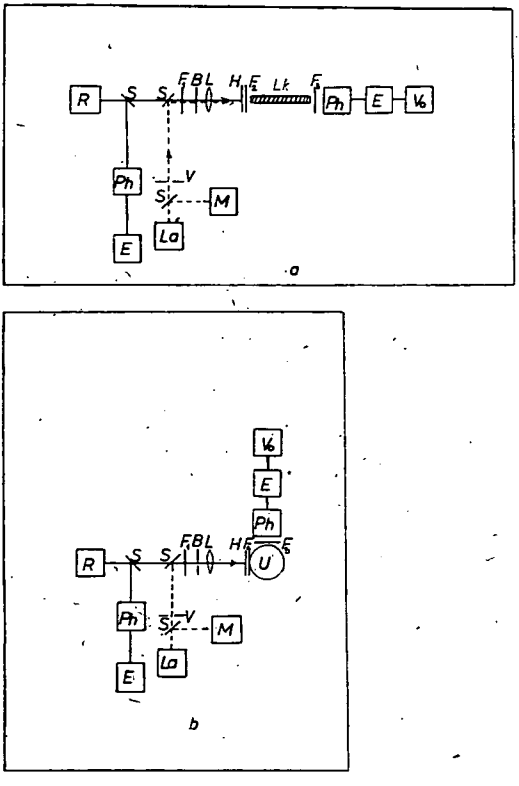

Fig. 1. Device for nonlinear absorption measurement
a with guide cable (Lk)
b with integrating sphere (U)
B: diaphragm, *E:* electrometer, *F:* filters, *H*: sample holder, *L:* lens, *La:* He-Ne laser, *M:* power meter, *Ph:* photocell, *R:* Q-switch ruby laser, *S:* mirrors, *V:* shutter, *Vo:* digital voltmeter

The curves give clear hints to excited state absorptions for in-vivo-chlorophyll-a as well as in-vivo-chlorophyll-a with a cross section greater than the corresponding cross section of the ground state absorption $(\sigma_2(694\text{ nm}) > \sigma_1(694\text{ nm}))$ [2]. It is not yet decided wheter the excited state absorption of in-vivo-chlorophyll belongs to the antenna molecules or to the reaction centre molecules.

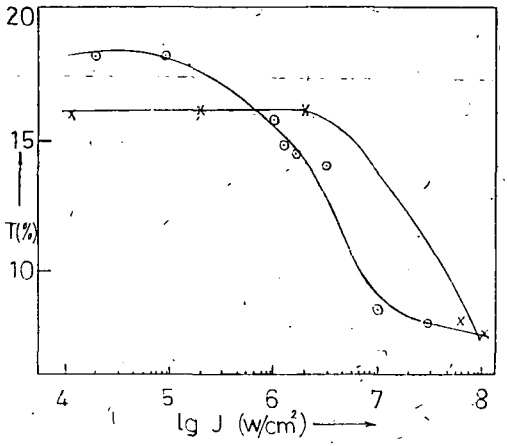

Fig 2. Intensity dependent transmission of chlorophyll-a in ethanol (x) and in dioxane (•) at 694 nm

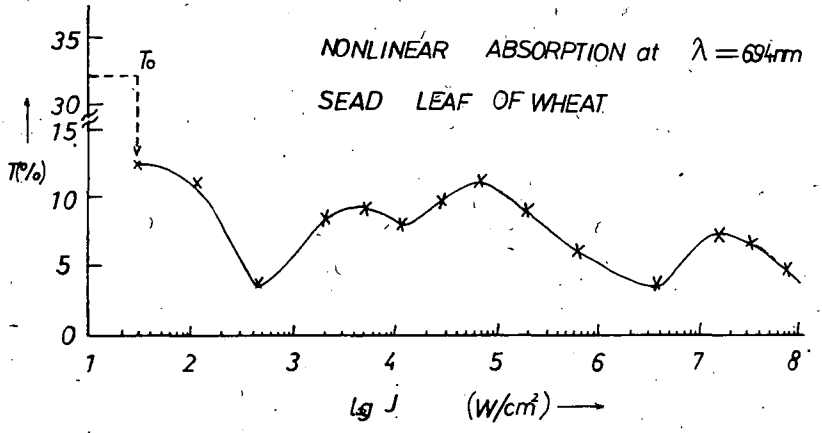

Fig. 3. Intensity dependent transmission of an intact plant (leaf of wheat) at 694 nm. (With reference to the signal transmission T_0 the curve as a whole is located at smaller transmission values)

References

[1] *Szalay, L., J. Hevesi, E. Lehoczki:* Acta Phys. et Chem. Szeged **19**, 403 (1973).
[2] *Leupold, D., S. Mory, P. Hoffmann, B. Hike, R. König:* Opt. Commun. (to be published).

НЕЛИНЕЙНОЕ ПОГЛОЩЕНИЕ ХЛОРОФИЛЛА *a*

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Представлены перспективы и предварительные результаты нелинейной абсорбционной спектроскопии хлорофилла.