LASERS IN PHYSICAL CHEMISTRY AND BIOPHYSICS

PROCEEDINGS OF THE 27th INTERNATIONAL MEETING OF THE SOCIETE DE CHIMIE PHYSIQUE

THIAIS, 17-20 June 1975

JACQUES JOUSSOT-DUBIEN
(editor)

ELSEVIER

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Edited by

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FOREWORD

International meetings are an inevitable blending of organisational nightmares and of scientific problems intermingled with difficult choices. The 27th Discussion of the Société de Chimie physique has been no exception in this respect.

Although our meetings are always fully published, including discussions, this was perhaps not made sufficiently clear to some authors, who were suddenly faced with my request for their manuscripts, a prospect they did not fully appreciate at short notice. But scientific people are definitely cooperative and everybody managed to comply with my preposterous request. Not only were the discussions lively but all participants accepted to set at once to the task of writing down extemporaneously their questions, answers and comments. So that, for the first time in the history of the Société de Chimie physique, the full manuscript of the book of Proceedings is ready for publication five weeks after the actual conference. It includes the contributions of the three colleagues whom illness or unexpected academic duties prevented from attending, as they handsomely offered to send in their texts. And so they did.

It was indeed a good meeting. We had a fairly complete review of the state of the art in lasers, as related to physicochemical or biophysical research, from solid-state to tunable dye lasers through organic lasing materials (including single crystals) and very promising new rare earths glasses. Nearly all the "picosecond community" was present and some papers even dealt with single subpicosecond pulses. After what there was a contribution in which everybody concerned was told why and how they had probably been wrong, this being done with the help of slides very fairly supplied by the very people under attack. This did not prevent some of the picosecond specialists of commenting aloud on the prospect of femto(10^{-15}) second pulses, a piece of futuristic equipement which raises but two problems:

-1-How to do it?

-2-What to do with it?

But it has to be admitted that picosecond work, now in full bloom, would not have sounded very realistic five years ago. When the

technical breakthrough is achieved applications follow fast enough.

For reasons one has to accept all research on isotope separation is not presented at meetings. But there is still some unclassified work being done in this field and part of its results are in this book.

As for the biophysical applications it is one of the fields where the Organising Committee was faced with very difficult choices. We had to decide quite arbitrarily that some good papers were outside the scope of the meeting. All we can say to justify our choice is that what we kept was of a fascinating variety, ranging from genetic surgery to a pretty piece of pure physics, where a check on the validity of the theory of convection found an application, through the knowledge of the velocity spectrum of spermatozoids, in the study of human fertility. Some more "conventional" work was devoted to chromosomes, retinal, photosynthesis, molecular motions in mitochondrial membranes, etc..

Although there was a contribution on the fundamental problems of ultrafast kinetics it is certain that the Organising Committee had to leave out all things related to the study of reaction mechanisms. But it did its best to keep most of the research that is at the same time significant and of general value.

It is our hope that this book will prove useful to many, as a review of what has been achieved and of the present trends, also as an illustration of the wonderful versatility of the laser, an irreplaceable instrument for physicists, chemists and biologists alike. The discussions will certainly shed some light on the main questions research teams ask themselves -or others-.

We received generous financial support from a number of organisations: their help is gratefully acknowledged. The Organising Committee performed a difficult task in a way which earned it the gratitude of the Société de Chimie physique and resulted in a timely, efficient and useful meeting. But our thanks are mainly due to the authors and participants, for providing us with their latest results for publication and for vigourous and good-natured discussions which enrich these Proceedings. This book is their work.

July 1975

C.Troyanowsky
General Secretary

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- Délégation générale à la recherche scientifique et technique
- European Research Office
- Direction des recherches et moyens d'essais
- Secrétariat aux Universités

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- Compagnie Française de Raffinage
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- Société Kodak Pathé
- Société Thomson-C.S.F.
- Union des Industries chimiques

We should like to renew our thanks to all the above.

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RECENT DEVELOPMENTS IN DYE LASERS

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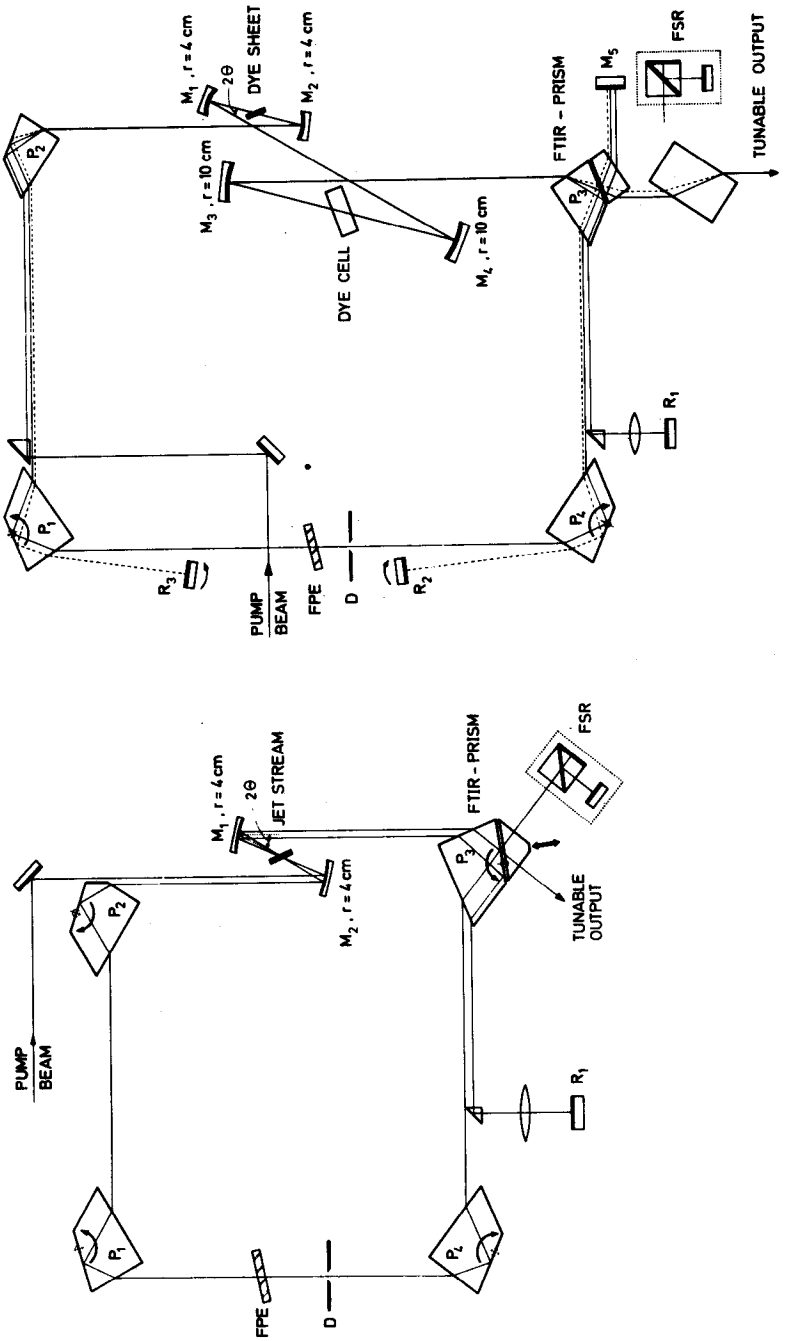
Progress in the dye laser field has been manifold recently :

1) New dyes have been found and new experimental findings allow a better understanding of the photodegradation process in dye lasers.
2) Vapor phase dye lasers have been operated.
3) New resonator configurations and tuning methods have been employed.
4) Higher average power and higher reliability have been achieved.
5) UV- and IR-wavelength regions that can be generated with dye lasers by nonlinear methods have been extended and the efficiencies of the conversion processes have increased.

While it is easy to find new dyes that are useful with nitrogen laser pumping (1), good dyes for flashlamp pumping are harder to find and only relatively few dyes will show satisfactory action in cw-dye lasers. Recently 16 new dyes of the cyanine class of dyes have been found by the Kodak group to operate satisfactory in flashlamp-pumped dye lasers in the near infrared (2). We have found a new class of dyes, namely the phenoxazones, which exhibit large solvatochromic shifts. One particular dye nileblue - A - phenoxazone, shows a shift of the center of the laser emission from 605 to 700 nm depending on the solvent (3). Molecular orbital calculations of dyes that have not yet been synthesized show, that it should be possible to obtain dye laser action with this class of dyes up to at least 1000 nm.

Another dye that shows a very large solvatochromic shift is the coumarin dye 3-phenyl-7(3) [1-phenyl-2-pyrazolinyl]) coumarin. The laser wavelength of this dye shifts from 530 nm in cyclohexane to 710 nm in DMSO because of a change of the dipolemoment with excitation which we determined to be 23 Debye. This dye also shows an interesting effect


M1, r = 4 cm
2θ
DYE SHEET
M2, r = 4 cm
M3, r = 10 cm
DYE CELL
M4, r = 10 cm
FTIR - PRISM
M5
FSR
TUNABLE OUTPUT
P1
P2
P3
P4
R1
R2
R3
PUMP BEAM
FPE
D
M1, r = 4 cm
2θ
JET STREAM
M2, r = 4 cm
FTIR - PRISM
FSR
TUNABLE OUTPUT
PUMP BEAM
P1
P2
P3
P4
R1
FPE
D

of an excited singlet state absorption between 550 and 680 nm (3).

A still disconcerting problem in dye lasers is the photodegradation of the dyes after about a million pump-fluorescence cycles even in the best dyes. There is no hope of improving this situation before the mechanisms of these photochemical processes have been elucidated. Recently a first step in this direction was taken by an American group who investigated the photochemical products formed in 7-diethylamino-4methylcoumarin (4) during dye laser operation. They found five different products, four of which were highly fluorescent and non-absorbing and thus without importance for the dye laser operation, while the fifth, a carboxylic acid that was produced by oxigenation of the methylgroup in 4-position, was strongly absorbing at the laser wavelength and thus quenched the laser action. Filtering the dye solution through an alumina filter that preferentically absorbed the carboxylic acid prolonged the useful life of the dye solution in the laser recirculation system by more than a factor of 3. This observation is consistent with an earlier observation, that the useful life time of a dye in which the methyl-group is replaced by a CF_3-group is much longer, since the CF_3-group cannot be oxidized so easily (5).

Another important recent development is the vapor phase dye laser which holds promise for the future of direct electrical excitation. We obtained dye laser emission in the vapor of the scintillator dye POPOP with nitrogen laser pumping (6) a result that was independently obtained by a group of the Bell Laboratories (7). Similarly, a Russian group reported dye laser operation of POPOP at 250° C with pentane "under high pressure" being present. Since the critical temperature of pentane is 190°C this means that here we have the dye in a supercritical solution and not a true vapor phase laser (8). Meanwhile we have used 3 additional dyes in a vapor phase laser and found a considerable number of dyes that will probably work in the near future. A vapor beam device that can be pumped by flashlamps or an electron beam is under construction in our laboratory.

Since a report on cw-dye lasers will be given in the afternoon session by Dr. Paugh, I will only report on some new developments in our lab. Two prism dye ring laser arrangements are shown in Fig. 1 The first one (Fig. 1 a) uses a Z-shaped arrangement for the leg containing the jet

stream. The four Abbe-Prisms are the predispersing elements while fine-tuning is possible with an uncoated etalon of 2 mm thickness and single-mode operation can be achieved with a Fox-Smith-reflector that couples back one of the beams exiting from the frustrated - total - internal - reflection prism. The second one (Fig. 1 b) uses two focussing regions in X-shaped configuration that not only reduces astigmatism but also coma (10). One focal region is for the active dye (e.g. rhodamine 6 G), while the second focal region can either contain a mode-locking dye (e.g. DODTC) or a second active dye (e.g. cresyl violet) that is pumped by the emission of the dye in the first focal region, so that simultaneous emission on two wevelengths can be obtained, if a resonator for the second dye is provided, as is done here using the two mirrors R_2 and R_3. The two laser beams can be made collinear by a compensating plate and then used for frequency-difference generation or similar applications. One important advantage of ring lasers as compared to linear lasers is the low threshold that is immediately apparent when closing the ring. In the case of the laser shown in Fig. 1 b threshold was as low as 10 m W after careful adjustment.

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