



Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

ENERGY & ENVIRONMENT DIVISION

RECEIVED
LAWRENCE
BERKELEY LABORATORY

JUL 15 1982

LIBRARY AND
DOCUMENTS SECTION

To be published in Optics Communications

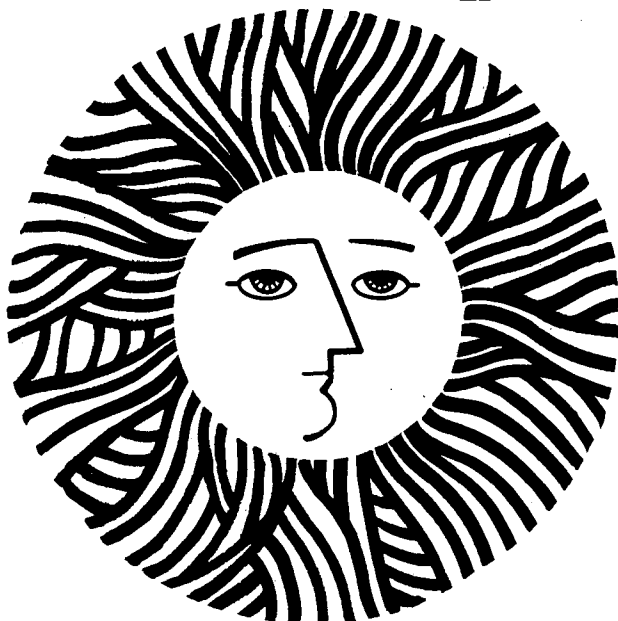
HIGHLY EFFICIENT WIDELY TUNABLE SUBPICOSECOND
DOUBLE MODE-LOCKED LASER

Zafer A. Yasa and Nabil M. Amer

March 1982

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 6782.*



LBL-14407
c.2

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

HIGHLY EFFICIENT WIDELY TUNABLE SUBPICOSECOND
DOUBLE MODE-LOCKED LASER

Zafer A. Yasa and Nabil M. Amer

Applied Physics and Laser Spectroscopy Group
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720
U.S.A.

ABSTRACT

A high average power (~ 150 mW) subpicosecond (0.5 psec) passively double mode-locked dye laser is described. Two synchronized cw pulse trains are generated which are independently tunable over broad wavelength ranges (~ 600 Å).

This work was supported by the Office of Energy Research, Characterization and Measurement Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, and by the Lawrence Berkeley Laboratory Director's Program Development Fund.

Highly Efficient, and Widely Tunable Double Mode-Locked Dye Laser

I. Introduction

With the advent of synchronously pumped dye lasers,⁽¹⁻⁴⁾ picosecond spectroscopy is becoming widely investigated. Despite the difficulty in achieving subpicosecond operation, these dye lasers have the favorable characteristics of high average output power (~ 100 mW) and wide tuning range ($\sim 600 \text{ \AA}$) when operated in the picosecond regime. Conventional* passively mode-locked dye lasers,⁽⁵⁻⁸⁾ on the other hand, while generating shorter pulses, yield low output power (~ 10 mW) and have a relatively restricted tuning range ($\sim 200 \text{ \AA}$).

In this letter, we report on a passively double mode-locked dye laser which produces subpicosecond pulses (0.5 psec) with high output power (~ 150 mW) and high conversion efficiency (5-10%) over a wide tuning range ($\sim 600 \text{ \AA}$). In addition, through double mode-locking^(9,10), a highly synchronous secondary picosecond beam, tunable over a similar range, is generated at longer wavelengths.

Furthermore, we point out that, in the subpicosecond regime, passively mode-locked dye lasers are ultimately more efficient than their synchronously pumped counterparts, as demonstrated below.

II. Experiment

A cw Ar^+ laser (5145 \AA) pumped rhodamine 6G(R6G) laser is double mode-locked by a mixture of rhodamine 101 (R101) and cresyl violet (CV)

* Throughout this paper, the word "conventional" implies a non-ring type cavity for which there is no overlap of counter propagating pulses in a dye medium. We consider only conventional mode-locked dye lasers.

in an independently tunable cavity configuration as shown in Fig. (1).

The following factors optimize the operation of this laser:

- (1) Output coupler M_3 has a transmission value T of 3% to 10%, thus yielding high output coupling efficiency.
- (2) The double mode-locking mixture R101/CV has an absorption spectrum which closely matches the gain spectrum of R6G, thus enabling easy tunability over a wide wavelength range.
- (3) Cavity length L is kept at about 75 cm to avoid multiple pulse operation.
- (4) The R6G jet is placed near the center of the cavity to maximize the gain for single pulse operation.⁽¹¹⁾
- (5) The ratio of the radii of curvature R_4/R_1 of mirror M_4 and M_1 , respectively, is kept high resulting in a wide wavelength range of stable mode-locking.
- (6) Folding angles are kept to the minimum possible to obtain good astigmatic compensation. Mode structure of the R6G beam is maintained a good Gaussian.
- (7) The angle between the two beams in the R101/CV jet is minimized to obtain maximum overlap.
- (8) The thickness of the birefringent tuning filter (LF1) is sufficiently thin (0.375 mm) to suppress a red lasing tendency in the R6G cavity. This also results in less dispersion.

With these conditions satisfied and the cavity lengths matched, this double mode-locked dye laser provides 0.5 psec pulses (yellow) with

a wide tuning range (570-630 nm), and high average power (~ 150 mW for 2W pump power). Typical efficiencies of 5-10% for pump laser powers of 1-2.5 W are readily obtained, with a weak dependence on pulse width. To our knowledge, this is the highest output power and efficiency for a cw mode-locked subpicosecond dye laser ever reported. Excellent pulse stability and compact autocorrelation traces are maintained over many hours of operation. Multiple and satellite pulsing problems are readily eliminated. Subpicosecond pulses are obtained near and also significantly above threshold. It was observed that stable picosecond operation persists while the birefringent tuning filter is continuously rotated over nearly the entire tuning range without the need for adjusting the pump power or the cavity alignment.*

In Fig. (2) we show two typical (yellow) pulse autocorrelation traces, at 575 nm and 625 nm, as displayed by a rapid scan autocorrelator.⁽¹²⁾ Assuming a sech^2 pulse shape, we measure a pulsewidth Δt of 500 fsec. Pulsewidth-bandwidth products $\Delta\nu\Delta t$ are typically in the range of 0.5 - 1.0. Peak powers correspond to ~ 1.5 KW for the repetition rate of 200 MHz.

By means of the double mode-locking process⁽⁹⁾, independently tunable picosecond pulses are synchronously^(13,14) generated at a longer wavelength (red), tunable over 630 - 660 nm. With the output coupler of $T = 1\%$, typical average power for these red pulses was a few mW. The long wavelength limit, at present, was set by the coating characteristics on M5. In principle, the tuning range of the red beam should

* Pump power and cavity adjustments are necessary to obtain subpicosecond pulses at a given wavelength.

encompass most of the lasing spectrum of the R 101/CV mixture (620 nm - 700 nm). In the present work, no attempt was made for the optimization of the red pulses. It was also observed that as the shorter wavelength (yellow) was tuned towards 630 nm, the mode-locker ceases to lase with the yellow beam remaining conventional passively mode-locked.

Double mode-locked operation of this laser depends on the close matching of the lengths of the two cavities. For subpicosecond pulses, cavity lengths needed to be matched to within a few μm . At present, thickness fluctuations of the R6G jet is likely to be the main pulsewidth limiting factor. This separate cavity independently tunable configuration provides for the correction of the inherent cavity mismatch (due to different group delays of the pulses in various optical elements) present in the collinear cavity configuration previously described⁽¹⁰⁾. Similar results of pulsewidth (yellow), output power, and tuning range were obtained also in the collinear configuration, which indicates a dynamic compensation of the mismatch. The average power in the red beam was higher (~ 20 mW) in the collinear configuration, as contributed by the R6G gain. However, independent tunability is difficult to obtain. The present configuration also has the advantage over the prism tuned configuration⁽¹⁵⁾ that the thickness of the tuning elements can be kept smaller (less dispersion) while the cavity length is kept shorter to avoid multiple pulsing.

The absorption spectrum of the double mode-locking mixture is given in Fig. (3). The peak absorptance corresponds to a single pass small signal absorption of 8% (0.15 mm thick jet) and a threshold pump power of ≈ 2 W for $T=3\%$. The corresponding dye concentrations are 1.4×10^{-5}

molar for R101 and 1.3×10^{-5} molar for CV. Also shown in Fig. (3) is the emission spectrum of R6G. The excellent matching of the two spectra is important for the demonstrated wide tunability.

III. Discussion

In Fig. (4) we plot the output pulse energy and ultimate pulsewidth for an ideal (stable pump pulses, no cavity length perturbation, no dispersion) conventionally synchronously pumped dye laser ($T=50\%$, pump pulsewidth = 100 psec) as a function of cavity mismatch⁽¹⁶⁾. The pulsewidth and energy of the conventional passively mode-locked lasers depend upon parameters such as $S = F\sigma_a/\sigma_e$ (where F is the ratio of the focal spot areas in the two dye jets, σ_a is the absorption cross-section of the mode-locking dye, and σ_e is the emission cross-section of the laser dye), cavity length, threshold pump power, and the output coupler transmission. A typical energy/pulse for $T=3\%$ under ideal conditions (no triplet losses, etc.) is $\sigma_e E_{out} \cong 0.1$ photons⁽¹¹⁾. The corresponding pulse width depends on the cavity dispersion but can be in the subpicosecond range. This value is plotted in Fig. (4) as a straight line. It can be readily seen that the pulse energy of synchronously pumped dye lasers decreases significantly as the pulsewidth decreases, to below the pulse energy value for passively mode locked lasers for the subpicosecond regime.

Hence, Fig. (4) depicts the generally unrecognized fact that, despite its apparent higher losses, the pulse compression dynamics in the passively mode-locked dye laser cavity is such that subpicosecond pulses of higher energy can be stably sustained, as compared to a conventional synchronously pumped dye laser. In the latter case, only a

small fraction of the available gain is usable for sustaining a subpicosecond pulse⁽¹⁶⁾, with the remainder lost to fluorescence or to the generation of spurious satellite pulses. Pulses of higher energy become stable in a synchronously pumped dye laser cavity only at the expense of a broadened pulsewidth.⁺

The extent of the role of the double mode locking process (lasing of the mode locking dye) in the demonstrated high efficiency of our system remains to be fully understood. However, there is a strong indication that this phenomenon enables stable subpicosecond operation at high values of S , which also correspond to higher pulse energies.⁽¹¹⁾ On the other hand, for conventional passively mode locked dye lasers, there exists theoretical⁽¹¹⁾ and experimental⁽¹⁸⁾ evidence that S must not reach very high values, particularly for long cavity lengths; this may result in lower pulse energies.* The observed wide tuning range of our double mode locked laser provides further related evidence in support of this argument.

V. Conclusions

In summary, a highly efficient cw double mode-locked dye laser is described. In addition to generating widely tunable, compact subpicosecond pulses with high power, a highly synchronous and independently tunable pulse is simultaneously generated at a longer wavelength. This two-color cw ultra-short pulsed laser source should prove to be a

⁺ The energy of stable pulses generated by a combination of synchronous pumping and passive mode-locking⁽¹⁷⁾ is most likely⁽⁴⁾ more characteristic of passive mode-locking than of synchronous pumping.

* Additional pulse sharpening mechanism, as in Fork et.al.⁽¹⁹⁾ can improve the pulse energy and the pulse width of passively mode-locked lasers.

versatile tool for excite-and-probe type experiments in picosecond spectroscopy.

VI. Acknowledgements

This work was supported, in part, by the Office of Energy Research, Pollutant Characterization and Safety Research Division of the U.S. Department of Energy, Contract No. DE-AC03-76SF00098, and by the Lawrence Berkeley Laboratory Director's Program Development Fund.

VII. References

- (1) J.P. Heritage and R.K. Jain, Appl. Phys. Lett. 32, 727 (1978).
- (2) A.I. Ferguson, J.N. Eckstein and T.W. Hansch, J. Appl. Phys. 49, 5389 (1978).
- (3) J. Kuhl, H. Klingenberg, and D. von der Linde, Appl. Phys. 18, 279 (1979).
- (4) J.P. Ryan, L.S. Goldberg and D.J. Bradley, Opt. Commun. 27, 127 (1978).
- (5) E.P. Ippen, C-V. Shank, and A. Dienes, Appl. Phys. Lett. 21, 348 (1972).
- (6) I.S. Ruddock and D.J. Bradley, Appl. Phys. Lett. 29, 296 (1976).
- (7) J.C. Diels, E. Van Stryland, and G. Benedict, Opt. Commun. 25, 93 (1978).
- (8) D. Rosen, A-G. Donkas, Y. Budansky, A. Katz and R.R. Alfano, IEEE J. Quantum Electron. QE-17, 2264 (1981).
- (9) Z.A. Yasa and D. Teschke, Appl. Phys. Lett., 27, 446 (1975), and Z.A. Yasa, J. Appl. Phys. 46, 4895 (1975).
- (10) Z.A. Yasa, A. Dienes, and J.R. Whinnery, Appl. Phys. Lett. 30, 24 (1977).
- (11) Z.A. Yasa, Ph.D. Thesis (University of California, Berkeley, 1976) (unpublished).
- (12) Z.A. Yasa and N.M. Amer, Opt. Commun. 36, 406 (1981).

- (13) E. Bourkoff, A. Dienes and J.R. Whinnery, Appl. Phys. Lett. 34, 455 (1979).
- (14) G. Arjavalingham, A. Dienes and J.R. Whinnery, to be published in Optics Letters, May 1982.
- (15) E. Bourkoff and J.R. Whinnery, Opt. Lett. 4, 179 (1979).
- (16) Z.A. Yasa and N.M. Amer, to be published.
- (17) G.A. Mourou and T. Sizer II, Commun. 41, 47 (1982).
- (18) The difficulty of obtaining stable mode-locking at the peak absorption of DODCI is not fully accounted for by its photoisomeration.
- (19) R.L. Fork, B.I. Greene and C.V. Shank, Appl. Phys. Lett. 38, 671 (1981).

VIII. Figure Captions

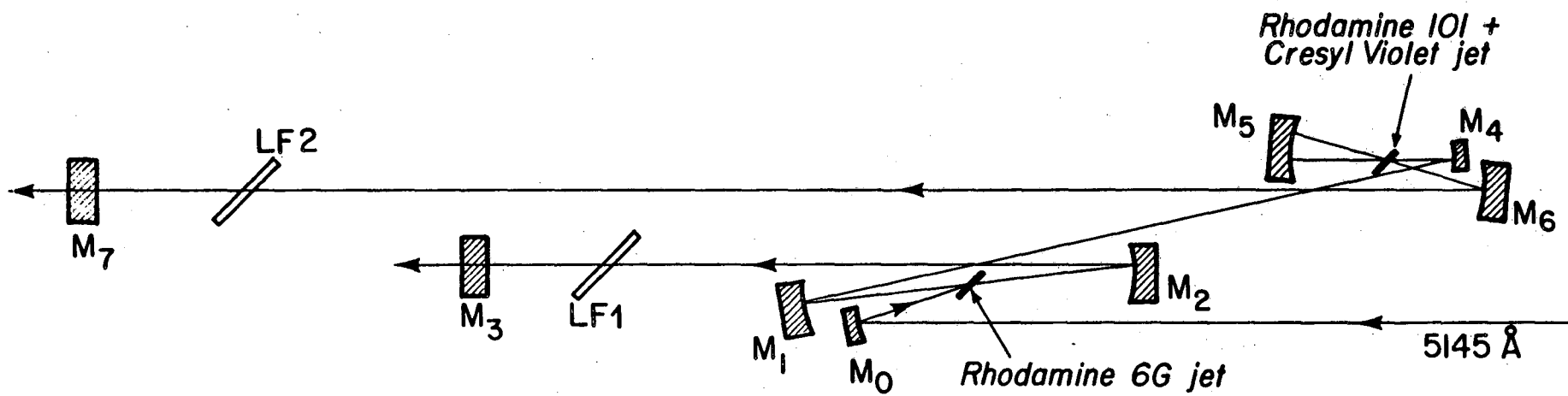
Fig. 1 Cavity configuration of the independently tunable double mode-locked dye laser. M_1 , M_2 and M_4 are R6G high reflectors (560 nm - 650 nm) with radii of curvature $R_1=R_2=10$ cm and $R_4=3.5$ cm (or 5 cm). M_5 is a broadband high reflector with $R_5=2.5$ cm. M_6 is a red high reflecting mirror (600 nm - 700 nm) with $R_6=5$ cm. Output couplers M_3 has $T=3\%$ or 10% and M_7 has $T=1\%$. LF1 and LF2 are single plate birefringent filters.

Fig. 2 Autocorrelation traces of two R6G pulses. (a) $\lambda=575$ nm (b) $\lambda=625$ nm. Horizontal scale corresponds to 1.5 psec/division for a sech^2 pulseshape.

Fig. 3 Absorption spectrum of the mode-locking mixture of R101/CV and the emission spectrum of R6G. Peak absorptance of R101/CV is \cong

0.4 for a 1 mm pathlength.

Fig. 4 Pulsewidth and energy/pulse for synchronously pumped dye lasers ($T=50\%$, pump pulsewidth = 100 psec) vs. cavity mismatch. Also shown is a typical output energy value (0.1 photons) for a sub-picosecond passively mode-locked dye laser pulse.



XBL 823-296

Fig. 1

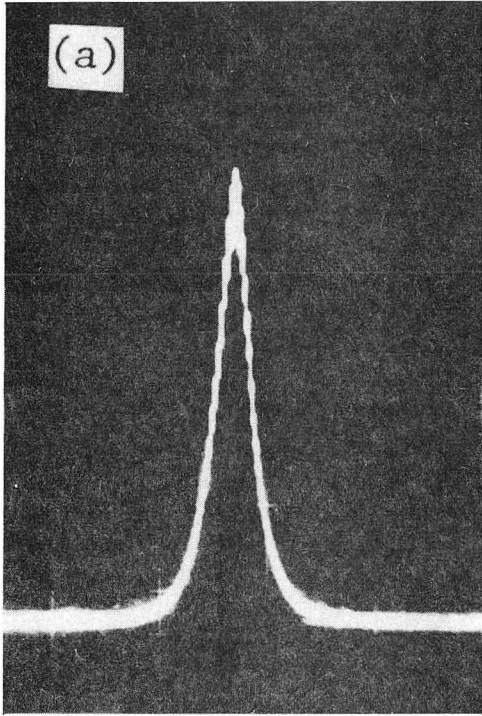
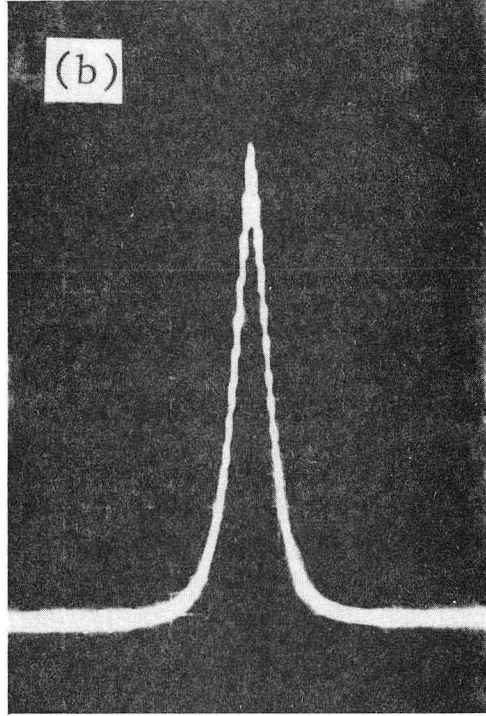
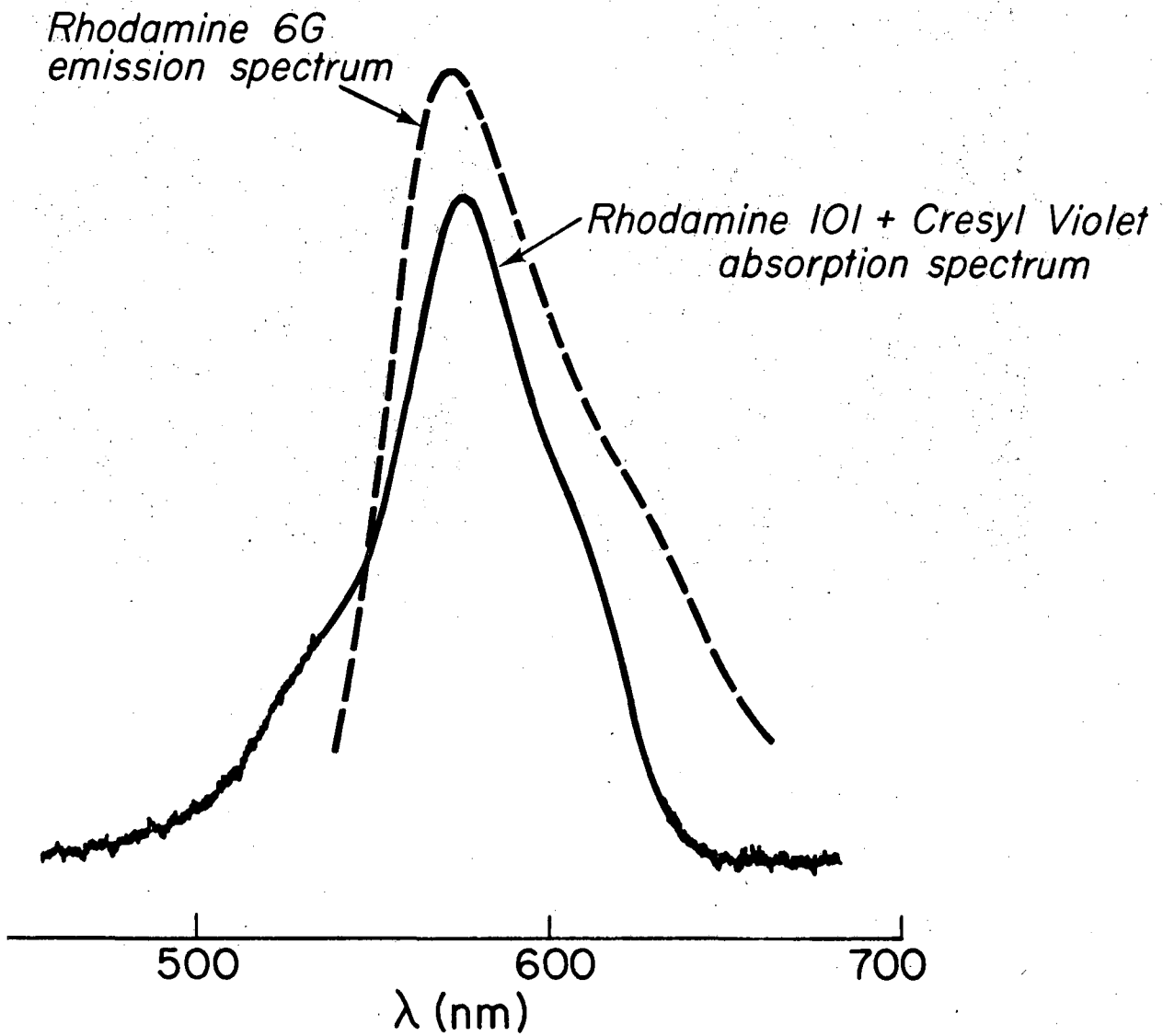


Fig. 2

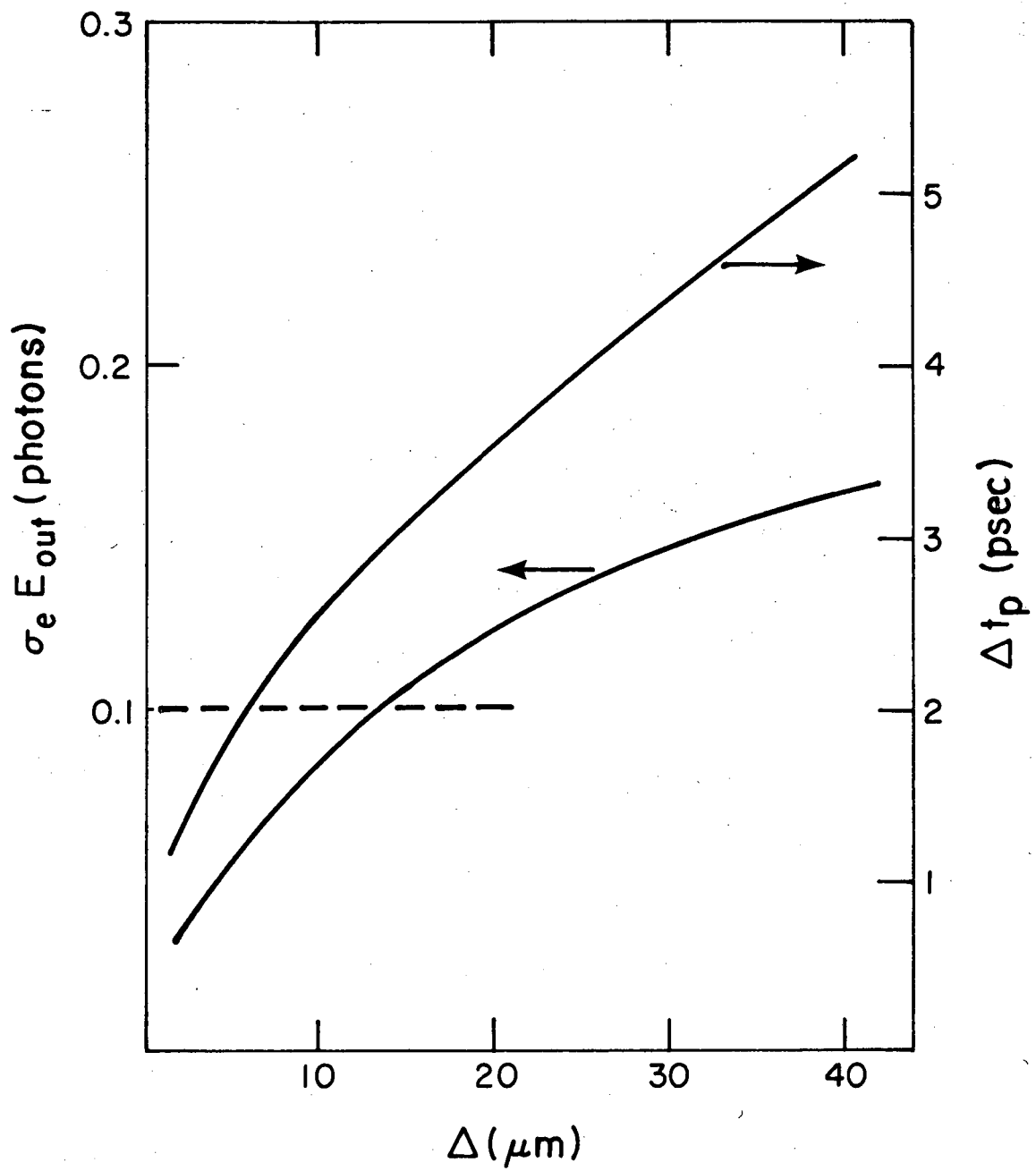


XBB 826-4935



XBL 823-295

Fig. 3



XBL 823-286

Fig. 4

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720