## Letter

Satellite picosecond pulses investigated with an optical Kerr gate\*

Since picosecond laser pulses are currently being used to study ultrafast phenomena in liquids and solids [1], it is extremely important to beware that the picosecond laser pulse train emerging from a Nd<sup>3+</sup>: glass laser mode-locked and Q switched by a saturable dye does not usually consist of only pulses separated by the round trip time of the laser cavity. Extraneous satellite pulses, whose temporal positions depend on the relative position of the dye cell in the laser cavity, appear persistently in the pulse train interlacing with the main pulse [2]. Limited by the finite response time of the combination of fast photodiode and fast oscilloscope, it is difficult to discern satellite pulses separated from the main pulse by 0.5 ns in such a detecting system. An apparently clean pulse train when the dye cell is positioned near the end mirror of the laser cavity still includes these satellites. The selection of what is believed [3] to be a 'single' pulse by the isolation of a 5 ns portion of the pulse train by a Pockel shutter still may contain satellite pulses. Two photon fluorescence techniques [4] in principle, can measure the pulse width and relative temporal positions of the picosecond satellite pulses; however, systematic study of the pulse train is inconvenienced by large spatial dimensions required for the fluorescent dye cell and photographic plates. We have employed the ultrafast light gate technique, first developed by Duguay and Hansen [5], to systematically investigate the temporal development of the laser pulse train emitted from a linear modelocked Nd<sup>3+</sup>: glass laser cavity. As long as the dye cell is separated from the mirror, satellite pulses are always observed.

The laser oscillator consists of a Nd<sup>3+</sup>; glass rod  $(\frac{1}{2}in \times 7\frac{1}{2}in)$ , whose ends are cut at Brewster's angle and placed between two wedged reflectors separated by an optical path of 0.9 m. A Kodak dye cell with Kodak 9860 dye in dichloro-1.06 µm) is placed near Brewster's angle between the rod and a 100% reflector (1 m curvature and ½° wedge). The laser pulse is coupled out through the 40% transmitting front mirror (flat,  $\frac{1}{2}$ °

wedge) of the oscillator cavity. The frequency doubled laser output is aligned collinearly with the probing 1.06 µm beam at the optical gate, a 1 cm CS<sub>2</sub> cell situated between crossed polaroids. The gate has a signal-to-noise ratio of  $\sim$ 500, and transmits  $\sim$ 1.5% of the 0.53  $\mu$ m light when the gate is opened by a 0.5 GW, 0.5 cm diameter probing beam. The prompt response curve of the gate has a full width at half height of about 10 ps, which is a measure of the convoluted width of the 1.06 µm, 0.53 µm pulses, and the response time of CS<sub>2</sub> molecules, which is about

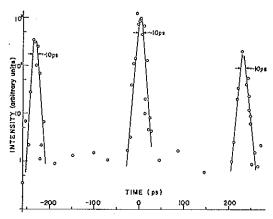


Figure 1 The normalized transmitted light intensity (arbitrary units) at 0.53 µm versus the time delay of the probing 1.06 µm beam. Each data point is the result of one

1.8 ps. As the probing beam is gradually delayed, transmitted signals would appear at positions  $\tau = \pm n\tau_0 = \pm n(2\gamma/c)$ , where  $\gamma$  is the distance between the dye cell and the near mirror, c is the velocity of the light, and  $n = 0, 1, 2, \ldots$  In Fig. 1, the transmission of 0.53 um light, normalized by the product of incident intensity of 0.53 µm light and the square of the intensity of probing 1.06 µm beam, is plotted against the delay of the probing beam for  $\chi = 3.5$  cm, and  $\tau_0 = 233$  ps. Satellite signals at delay position ethane (1.5 mm thick, and 70% transmission at  $\tau = \pm \tau_0$ , as well as the main pulse at  $\tau = 0$  are observed. This observation implies the existence of one satellite, since two groups of m pulses can overlap at (2m-1) positions at  $\tau = \pm n\tau_0$ , and  $n=0,1,\ldots m-1$ . The observed full width at

243

© 1974 Chapman and Hall Ltd.

half height of each pulse is about 10 ps. Occasionally satellite signals at  $\tau = \pm 2\tau_0$  are observed, but never at  $\tau = \pm 3\tau_0$ . As the position of the dye cell is varied, the positions of satellite signals are observed to shift according to the relation  $\tau = \pm 2n\chi/c$ . Assuming a constant ratio, k, between the intensity of the satellite pulse and the main pulse over the whole pulse train, the ratio of the transmitted light signal at  $\tau = \pm \tau_0$  and that at  $\tau = 0$  can be approximated by  $k^2/(1 + k^4)$ . Our data yields  $k = (54 \pm 8)\%$ . A 15% variation in the intensity of the satellite signal is also observed for a given laser main pulse.

Our observations support the model that the picosecond laser pulses develop from noise pulses generated in the initial stage of the laser action process. Each time the dye is bleached by an intense fluctuation pulse a satellite pulse may simultaneously pass through the dye cell in the opposite direction, thus leading or lagging the main pulse by  $2\gamma/c$  [6]. Because the main and the satellite pulses return to the dye cell once more during the round trip within a linear laser cavity, depending on the power content in the pulses, a sequence of secondary or higher order satellites can be generated at position  $\tau = \pm 2n\chi/c$ , where  $n = 1, 2, 3, \dots$  It should be noted here, in a ring cavity [7], a pulse only passes through the dye cell once in a complete trip; therefore, the pulse train from such a cavity can be relatively cleaner.

It is obvious that satellite pulses can become a nuisance in the application of picosecond pulses in time spectroscopy. In conclusion, in order to eliminate the satellite pulses, dye in contact with rear mirror [8], i.e.  $\chi \sim 0$ , should be employed. Or the dye cell should be placed in position such that no satellite pulse would appear in the temporal interval of interest.

## References

- 1. For instances, see
  - R. R. ALFANO and S. L. SHAPIRO, Scientific Amer. 228 (1973) 43-60.
  - Idem, Phys. Rev. Lett. 26 (1971) 1247-1251.
  - A. J. DEMARIA, D. A. STETSER and W. H. GLENN, Jun., Science 156 (1967) 1557-1568.
  - M. A. DUGUAY and J. W. HANSEN, Opt. Comm. 1 (1969) 254-256,
  - K. B. EISENTHAL, Chem. Phys. Lett. 6 (1970) 155-157.
    A. LAUBEREAU, D. VON DER LINDE and W. KAISER,
    Phys. Rev. Lett. 28 (1971) 1162-1165.
  - P. M. RENTZEPIS, *Chem. Phys. Lett.* 2 (1968) 117-120. R. I. SCARLET, J. F. FIGUEIRA and H. MAHR, *Appl. Phys. Lett.* 13 (1968) 71-72.
- 2. A. A. MALYUTIN and M. YA. SHCHELEV, *JETP Lett.* 9 (1969) 266-268.
- R. HARRACH and G. KACHEN, J. Appl. Phys. 39 (1969) 2482-2483.
- A. SCHMACHPEFFER and H. WEBBER, *Phys. Lett.* **24A** (1967) 190-191.
- 3. T. L. NETZEL, P. M. RENTZEPIS and J. LEIGH, Science 182 (1973) 238-241.
- 4. J. A. GIORDMAINE, P. M. RENTZEPIS, S. L. SHAPIRO and K. W. WECHT, *Appl. Phys. Lett.* 11 (1967) 216-218.
- 5. M. A. DUGUAY and J. W. HANSEN, *ibid.* 15 (1969) 192-194.
- 6. E. M. GARMIRE and A. YARIV, IEEE J. Quant. Elect. 3 (1967) 222-226.
- A. J. DeMARIA, D. A. STETSER and W. H. GLENN, Jun., Science 156 (1967) 1557-1568.
- 7. R. C. ECKARDT, C. H. LEE and J. N. BRADFORD, *Appl. Phys. Lett.* 19 (1971) 420-423.
- 8. D. J. BRADLEY, G. H. C. NEW and S. J. CAUGHEY, Opt. Comm. 2 (1970) 41-44.

## 3 December 1973

W. YU and R. R. ALFANO
Physics Department,
City College of the City University
of New York,
New York, New York 10031, USA

<sup>\*</sup>Supported in part by Cottrell Research Grant and by the City University Faculty Award.