Short Communication

Development of picosecond time-resolved techniques by continuous-wave laser amplitude modulation
V: Elimination of r.f. interference problems

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In previous papers we have described a dual-beam phase fluorometer with picosecond time resolution [1] and we have recently listed the systematic errors in this type of measurement and methods of controlling or eliminating them [2]. The wavelength-dependent photomultiplier transit time was found to be a measurable quantity provided that measurements at various modulation frequencies are available. This also applies to the resolution of multiexponential decays. However, at most frequencies the signal-to-noise ratio is too poor for precise measurements to be made. The noise originates from the power amplifier of the electro-optic modulator and is of exactly the same frequency as the bandpass of the detection electronics. Efforts to shield the noise sources did not produce the desired result.

The problem can be elegantly overcome by applying a double-modulation scheme of the type described by Bado *et al.* [3]. Successive modulation of a laser beam with r.f. and audiofrequency (a.f.) signals produces sum and difference frequencies with the advantage that the sideband signals are free from coherent noise. The main features of the experimental arrangement are shown in Fig. 1.

A sinusoidal a.f. signal of 1800 Hz is imposed on the beam by means of an acousto-optic modulator while the r.f. modulation is achieved using an electro-optic modulator. The anode current of the photomultiplier is detected at one of the sidebands using a spectrum analyser in the fixed frequency mode with a resolution of 300 Hz. The frequency of the tracking generator, which is coupled to the local oscillators of the spectrum analyser, can be shifted by ±2.1 kHz so that either sideband can be chosen for investigation.

The temporal stability of the acquired signals expressed by their r.m.s. is typically ±0.01 dB for the reference or the sample and ±0.04 dB for the sum signal, resulting in error estimates of less than ±0.2 cm for distance measurements, which corresponds to a time resolution of less than ±7 ps.

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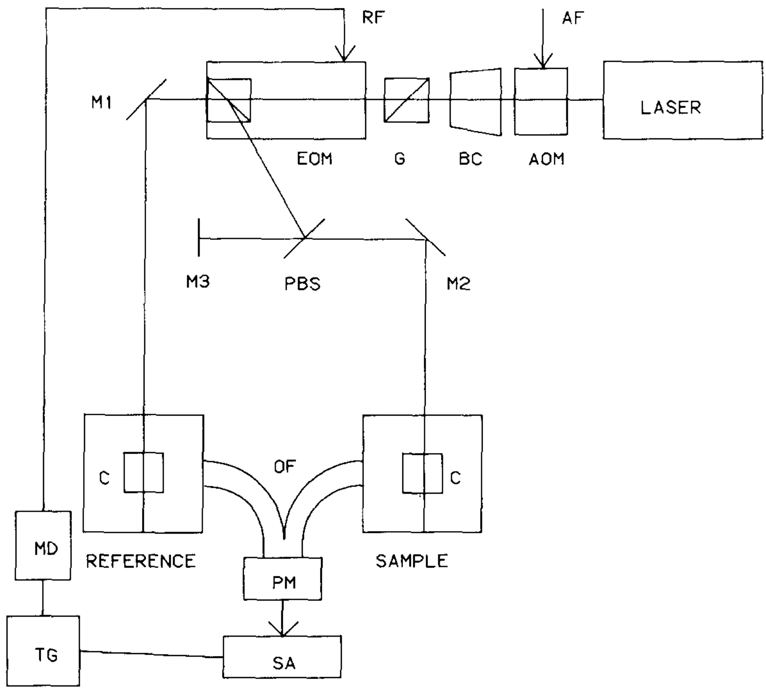

Fig. 1. Main features of the experimental arrangement: AOM, acousto-optic modulator; AF, a.f. input; BC, beam collimator; G, Glan prism polarizer; EOM, electro-optic modulator; RF, r.f. input; M1 - M3, mirrors; PBS, pellicle beam splitter; C, cuvette; OF, optical fibre bundle; PM, photomultiplier; SA, r.f. spectrum analyser; TG, tracking generator; MD, modulator driver.

The remaining noise measured in the absence of any light is of the same magnitude as the noise of the spectrum analyser itself when all the modulation equipment is switched off. The fluctuations of the signals can therefore be regarded as statistical and the contribution of non-random noise need no longer be included in the error estimation [1].

The advantage of sideband detection is offset by a poorer a.c.-to-d.c. ratio v_ω of the light flux. v_ω is defined by

$$v_\omega = \frac{i_\omega{}^0}{\langle i \rangle}$$

where $\langle i \rangle$ denotes the mean current delivered by the photomultiplier. The signal component $i(t)_\omega$ at frequency ω, whose mean power $\langle P_\omega \rangle$ is measured by the spectrum analyser, is

$$i(t)_\omega = i_\omega{}^0 \cos \omega t$$

When the impedance is 50 Ω the current amplitude $i_\omega{}^0$ is given by

$$i_\omega{}^0 = \frac{\langle P_\omega \rangle^{1/2}}{5}$$

For 100% distortion-free modulation, the values of v_ω at the sideband and at the carrier frequency are expected to be 0.5 and 1 respectively. The ratios v_ω at the sideband cover the range 0.4 - 0.5 for frequencies in the range 5 - 50 MHz.

Two tests of geometric performance using 0.3% solutions of Ludox HS40 [2] as the light scatterer were performed to check systematic errors in phase determination.

(1) The path length in a fixed geometric configuration was measured by detecting at the lower and upper sidebands alternately for a period of 3 h. The results are given in Table 1. It is evident that the information obtained from both sidebands is identical within the estimated error and that long time stability is guaranteed. In a previous attempt to achieve double modulation we used a mechanical chopper instead of the acousto-optic modulator and we observed that the measured path lengths at the two sidebands differed from one another in an uncontrollable way. This effect arises from the finite time which the chopper blade takes to turn the beam on or off. This problem is eliminated when the acousto-optic modulator is used because the modulation is homogeneous over the whole beam area.

TABLE 1

Measured path lengths L, estimated errors dL and residuals R at the lower and upper sideband

L (cm)	dL (cm)	R (cm)
Lower sideband		
43.02	0.15	−0.13
43.08	0.17	−0.07
43.28	0.14	0.14
43.14	0.17	0
Upper sideband		
43.15	0.17	0.01
43.15	0.20	0.01
43.12	0.17	−0.02
43.22	0.20	0.07
Mean		
43.14		

The residuals refer to the mean value of all determinations: r.f., 40 MHz; a.f., 1800 Hz.

(2) The path length was altered stepwise by moving the mirror M3. A plot of the measured displacements *versus* the effective displacements is shown in Fig. 2. This procedure can be considered as an absolute check of the apparatus against a well-defined physical parameter. It follows from

Fig. 2 that distance and hence phase measurements are reliable in the absolute sense within the estimated error of less than ±0.2 cm.

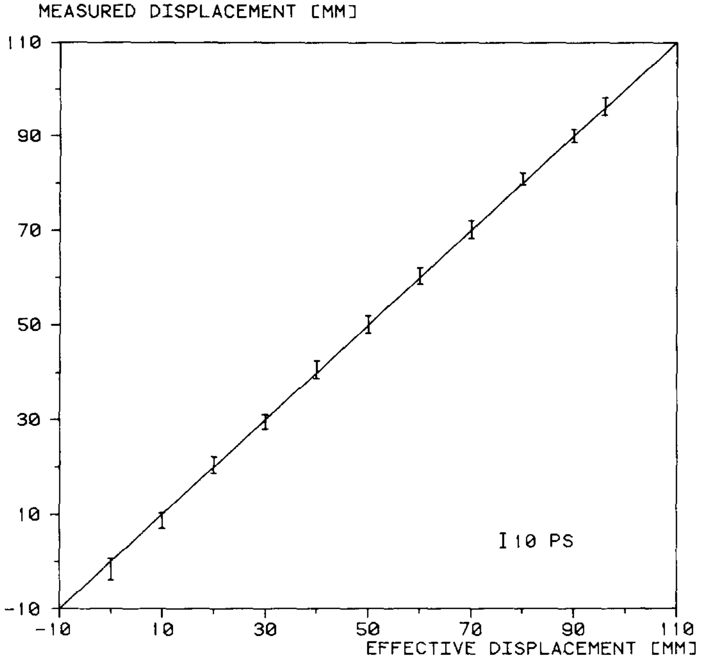

Fig. 2. Accuracy check by controlled variation in the path length (see text for explanation): I, $L \pm dL$; ——, best fitting straight line with slope unity (r.f., 35 MHz; a.f., 1800 Hz).

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3 Ph. Bado, Sc. B. Wilson and K. R. Wilson, *Rev. Sci. Instrum.*, *53* (1982) 706.