TIME RESOLVED SPECTROSCOPY OF PICOSECOND LASER PULSES WITH THE AID OF OPTICAL KERR EFFECT PHOTOGRAPHY

A.N. RUBINOV*, M.C. RICHARDSON, K. SALA and A.J. ALCOCK
Division of Physics, National Research Council of Canada, Ottawa, Canada

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An extension of the transverse gating, focal plane, optical Kerr effect photographic technique has permitted the direct observation of a frequency sweep within single ~10 psec duration 0.53 μ wavelength pulses derived from a mode-locked Nd:glass laser.

Direct time resolved studies of the output of mode-locked Nd:glass lasers have been reported by a number of workers [1–10]. Both the time duration [4, 5, 7–10], and the integrated spectrum [1–3, 5] of each individual pulse in the mode-locked laser output have been directly resolved. In addition, the application of non-linear optical diagnostic techniques, and picosecond streak photography, to the investigation of mode-locked pulses dispersively delayed by a grating pair have indicated the occurrence of frequency sweeping processes in the generation of these high power pulses [11, 12, 9]. However, although the existence of a frequency chirp on mode-locked pulses has been inferred from two-photon fluorescence [11] and four-photon fluorescence studies [12] of the entire pulsetrain and from picosecond streak photography of single dispersively delayed pulses [9], its direct observation on individual pulses has not yet been made. In this paper, we wish to report some initial results of a picosecond, time-resolved, study of the second harmonic spectrum of single Nd:glass mode-locked laser pulses. These have been obtained by using an extension of the technique of transverse gating, focal plane, optical Kerr effect (OKE) photography, which has previously been used in the picosecond framing photography of laser produced plasmas [13], and of individual second harmonic pulses [14, 15].

The general experimental arrangement is shown schematically in fig. 1. A single pulse, having an energy of ~1 mJ was isolated from the output pulsetrain of a mode-locked Nd:glass laser by means of an electro-optical shutter switched with a laser triggered spark gap. This pulse was then amplified to an energy of ~0.5 J by double passage through a 60 cm long, 25 mm diameter amplifier. It was then passed through a phase-matched KDP crystal, which converted a fraction of the pulse to the second harmonic frequency (λ = 5300 Å). This green light wad then separated from the infrared pulse by means of a mirror, M, which fully reflected the 1.06 μ radiation, but was transparent to its second harmonic. The reflected fundamental pulse was slightly focused into a 2 cm long, 1 cm wide cell, containing CS_2 and situated between two crossed (HN 22) polarisers, forming the central part of the transverse gating OKE photography arrangement [13]. The second harmonic pulse was focused by a 20 cm focal length lens (L_1) into a small scattering cell containing powder milk in suspension in alcohol. Radiation scattered from the narrow focal region within the acceptance cone of a $f/0.8$, 20 mm lens (L_2) was imaged in the OKE cell. Thus, providing that there are equal optical path lengths for the fundamental and second harmonic radiation to travel from mirror M to the OKE cell,

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an image of the 0.53 μ pulse passing through the scattering cell is gated by the 1.06 μ pulse in the cell of CS_2. As has already been pointed out [14, 15], in this particular configuration the gating pulse overlaps the image of the 0.53 μ pulse in the OKE cell with an effective velocity of $2c/n$, where c is the velocity of light, and n is the refractive index of CS_2. This overlapping process within the CS_2 cell leads to a time resolved image of the 0.53 μ pulse being transmitted by the OKE shutter. In the present case the time resolution is limited, since the gating pulse has approximately the same duration as the pulse being probed. However, with the provision of much shorter gating pulses the temporal resolution of this technique can be improved considerably.

In order to demonstrate this capability of the transverse, focal plane gating technique for producing a picosecond time resolved image, the spectrum of the individual second harmonic light pulses was investigated. A knowledge of the temporal behavior of the spectrum of the pulse is of interest in view of previous evidence of frequency chirping within mode-locked Nd:glass laser pulses [9, 11, 12]. The transmitted signal of the OKE shutter was imaged by means of an $f/1.8$, 86 mm focal length lens onto the slit of a 3/4 meter Czerny–Turner spectrograph and the optical system arranged such that the time axis of the transmitted signal was aligned along the slit of the spectrograph. The resulting spectrograms consequently displayed a wavelength scale along one dimension and a picosecond time scale along the other. A typical time resolved spectrum of the second harmonic of a single amplified mode-locked laser pulse is shown in fig. 2. The total spectral width of the pulse is ~30 Å, and its duration (fwhm), ~10 psec. However, as can be seen, there is a progressive sweep of the spectrum of the pulse toward longer wavelength, with a total wavelength shift of ~30 Å per 10 psec. This is more clearly shown in fig. 3, which shows a series of densitometer traces obtained from the spectrogram shown in fig. 2. The value of the wavelength shift within the second harmonic of one amplified 1.06 μ mode-locked pulse, is similar to that which Treacy [11] observed in the pulsetrain output of a mode-locked Nd:glass laser (~100 Å per 8 psec) by using dispersive delaying and two photon fluorescence techniques. However, it should be noted that the spectral resolution of the spectrogram of fig. 2 is limited to no better than

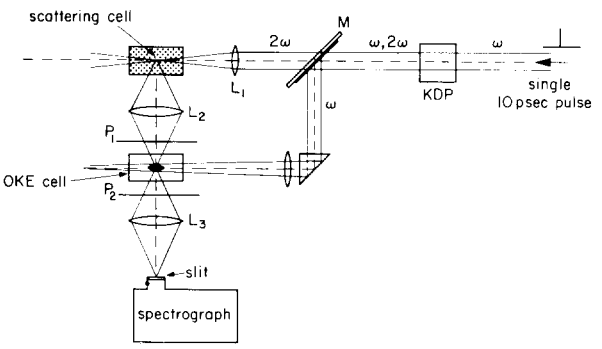

Fig. 1. Schematic representation of experimental arrangement for the time resolved spectroscopy of 0.53 μ mode-locked laser pulses using transverse, focal plane, optical Kerr effect photography. Legend: M – dielectric mirror which fully reflects fundamental radiation, but transmits second harmonic, P_1, P_2 – polarisers; L_1 – focusing lens; L_2, L_3 – quality, high power lens in photographic system.

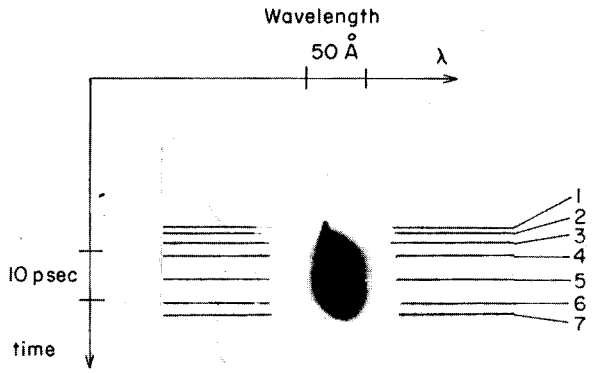

Fig. 2. Time resolved spectrum of the second harmonic of a single amplified mode-locked Nd:glass laser pulse.

~3 Å, corresponding to the dynamic spectral resolution of any instrument for a 10 psec pulse at 1.06 μ.

Time resolved spectra of the second harmonic of the oscillator output pulsetrain were obtained, and found to be similar to the spectrum shown in fig. 2. In addition, in some of the spectra obtained, both of the single amplified pulse, and of the output pulsetrain of the laser, a time-independent spectral component was observed. The origin of this stucture is at present unclear, however it did not appear to have originated from the second harmonic generation crystal, and may be intrinsic to the output spectrum of the mode-locked laser itself.

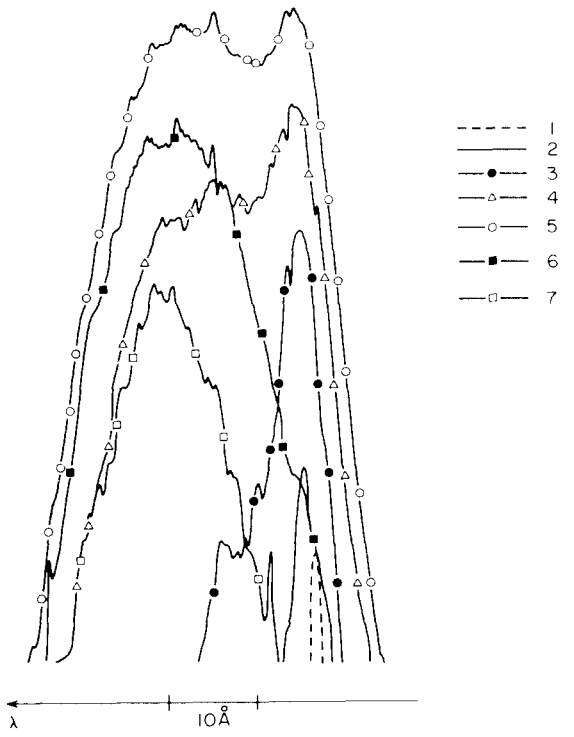

Fig. 3. A series of densitometer traces obtained from the spectrogram shown in fig. 2. The number on each trace corresponds to the appropriate line in fig. 2.

laser itself.

The results presented above, although preliminary, do illustrate the potential of a new technique of ultrafast photography. In particular, the provision of laser light pulses of significantly shorter duration, as may be afforded by mode-locked dye lasers, or with the use of optical compression techniques, in conjunction with tranverse gating, focal plane optical Kerr effect photography, will extend the range of time resolved spectroscopy into the subpicosecond range. However, this can only be achieved at a heavy cost in spectral resolution, and thereby constitutes a final limination on the applicability of this, or any other, ultrafast time resolved spectrographic technique.

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References

[1] A.A. Malyutin and M. Ya. Schelev, JETP Lett. 9 (1969) 266.
[2] V.V. Korobkin, A.A. Malyutin and M. Ya. Schelev, JETP Lett. 11 (1970) 103.
[3] V.V. Korobkin, A.A. Malyutin and A.M. Prokhorov, JETP Lett. 12 (1970) 150.
[4] M. Ya. Schelev, M.C. Richardson and A.J. Alcock, Appl. Phys. Lett. 18 (1971) 354.
[5] D.J. Bradley, B. Liddy and W.E. Sleat, Opt. Commun. 2 (1971) 391.
[6] R.C. Eckhardt, C.H. Lee and J.N. Bradford, Appl. Phys. Lett. 19 (1971) 420.
[7] S.D. Fanchenko and B.A. Frolov, JETP Lett. 16 (1972) 101.
[8] R.V. Dewhurst, H. Al-Obaidi, S.A. Ramsden, R.V. Holland and R. Harris, Opt. Commun. 6 (1972) 356.
[9] M.C. Richardson, IEEE J. Quant. Electron. QE9 (1973) 768.
[10] D.J. Bradley and W. Sibbett, Opt. Commun. 9 (1973) 17.
[11] E.B. Treacy, Appl. Phys. Lett. 14 (1969) 112.
[12] D.H. Auston, Opt. Commun. 3 (1971) 272.
[13] M.C. Richardson and K. Sala, Appl. Phys. Lett. 23 (1973) 420.
[14] K. Sala, M. Eng, thesis submitted to Fac. of Engng., Carleton University, Canada (1973).
[15] M.C. Richardson and K. Sala (to be published).