PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Introductory Remarks

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Introductory remarks

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The first day of this Discussion Meeting is mainly devoted to ultra-short laser sources and the methods of measurement of picosecond and subpicosecond pulses of radiation over the electromagnetic spectrum from X-rays to the infrared. The last decade has seen dramatic advances in the understanding of the complex physical and chemical processes occurring in mode-locked lasers and in the interactions, linear and nonlinear, with matter of the ultra-short light pulses so produced. The flashlamp-pumped frequency-tunable dye laser has played a key role in these advances. It provided the shortest and most controllable pulses for the development of time-resolving instruments and, later, was an experimental model for the detailed measurements that eventually led to the elucidation of the mechanisms by which ultra-short pulses evolve in a mode-locked laser from the initial fluorescence intensity fluctuations (see Bradley & New (1974) for references). These new concepts, in turn, were applied to the development of mode-locked continuous wave (c.w.) dye lasers and, more recently, to mode-locked semiconductor diode lasers. Both of these types of c.w. lasers will be discussed in some of the following papers.

The expansion of the laser and measurement techniques into the subpicosecond region (see Bradley (1977) for references) held out exciting possibilities for accurate measurements of previously unresolved ultra-fast processes in physics, chemistry and biology. These scientific opportunities are being increasingly exploited, as this volume shows. New information was soon obtained about primary photobiological interactions and excited state relaxation kinetics in molecules (Shapiro 1977) and on the dynamics of carrier relaxation in highly excited semiconductors (Bryant *et al.* 1978).

Electron-optical chronoscopy, which is still the only technique permitting the direct linear measurement of picosecond, and faster, luminous phenomena (see Bradley (1978) for references) has been extended to shorter wavelengths in the visible–ultraviolet and X-ray regions (Bradley *et al.* 1975), for experiments in nonlinear optics and for the study of high density, high temperature plasmas generated in laser fusion experiments. These studies could, in turn, lead to the production of femtosecond (10^{-15} s) pulses of coherent radiation at extreme ultraviolet and X-ray wavelengths, with revolutionary implications for molecular biology and chemistry. The analogy with the progress of spectroscopy over a century ago is close. The development of narrow linewidth spectroscopic sources, such as the hollow-cathode lamp, electrodeless discharges and atomic beams, enabled new phenomena arising from isotope and other nuclear effects to be quantitatively studied. While in atomic spectroscopy high resolution spectrometers and interferometers were first developed, in the time domain the light sources and diagnostic techniques have developed in step. As a result the methods of ultra-short laser pulse generation and measurement were relatively quickly brought to their present advanced state. They are now sufficiently refined and catalogued for them to be used with confidence for investigations on a picosecond–femtosecond timescale into the basic mechanisms of matter, animate and inanimate. This pattern,

of course, follows the historical pattern in that major developments in science and technology are almost always related to advances in instrumentation. It is thus interesting to briefly look at the origins of the measurement techniques with which we are concerned in this meeting.

The basic concepts of high-speed optical chronoscopy have been developed over a period of nearly two centuries. The principles of time measurement were first set out by Aristotle: 'We apprehend *time* only when we have marked motion . . . not only do we measure the movement by the time, but also the time by the movement, because they define each other'. The first experimental exploitation of this Aristotelian principle for accurate time measurement must be credited

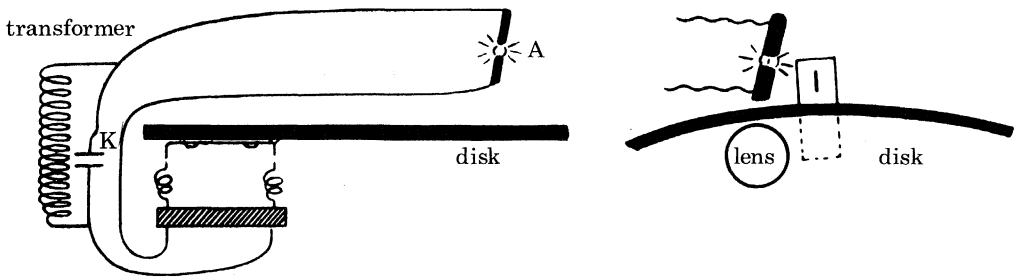

FIGURE 1. Arrangement of 'synchrono-phosphoroscope' of R. W. Wood (1921).

to Galileo Galilei for his discovery of the isochronism of the pendulum around 1580. However, fast optical chronoscopy really began when Wheatstone (1834) employed a rotating mirror to measure the duration of an electric spark and to study the propagation of electricity through copper wires. The speed of rotation of the mirror was determined from the note emitted by a siren mounted on the mirror spindle. By visual observation of the streaked image Wheatstone was able to show that a high voltage electric spark had a duration of less than 10^{-6} s. It is interesting to note that the results of Wheatstone's investigations were communicated to the Royal Society by Faraday. Also using a spark light source, Abraham & Lemoine (1899) were able to demonstrate that the Kerr cell shutter could be operated with an exposure time of less than 10^{-8} s. In the period between these two experiments Talbot (1852) had shown how electric sparks could be used for high-speed 'stop-motion' photography of rapidly moving objects and demonstrated his method with a newspaper page fixed to a rapidly spinning platform.

Closer to the subject matter of our meeting were the measurements carried out by R. W. Wood (1921) of the time interval between optical absorption and subsequent emission of fluorescence. Using his 'synchrono-phosphoroscope', Wood was able to set an upper limit of 2.5 μs to the lifetime of the emission from uranium glass and barium platino-cyanide, 'about ten times as brief as any observed phosphorescence' previously recorded. In this instrument (figure 1) a thin disk was mounted on the shaft of a synchronous motor, operated by the same 60 cycle alternating current that fed the primary of a 30 000 V transformer used for charging a Leyden jar capacitor. The capacitor was discharged through an electric spark in synchronism with the rotating disk and the emission of the sample following illumination by the spark was observed visually, or could be recorded photographically.

The use of image-tubes for high-speed photography was pioneered by Courtney-Pratt (1949) who showed that a magnetically deflected, streak image tube could give submicrosecond time resolution. However, the attainment of picosecond time resolution had to await the arrival, two decades later, of the Photochron family of streak-tubes based on the high-field photocathode

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

extraction mesh electrode principle (Bradley 1970). With this simple expedient the limit of time resolution was steadily improved to 700 fs (Bradley & Sibbett 1975) and the practicable limit of *ca.* 100 fs for electron-optical chronoscopy is now in sight.

Thus the picosecond era in optics, which can be said to have its origins in the first mode-locked ruby laser in 1965 (Mocker & Collins), rapidly developed with the first frequency tunable picosecond pulses from dye lasers in 1969 by Bradley & O'Neill, closely followed by the introduction of the picosecond streak camera in 1970. With increased understanding and sophistication in diagnostic equipment it became possible to generate bandwidth limited pulses as brief as *ca.* 1 ps from flashlamp pumped dye lasers. Transform-limited subpicosecond pulses were then obtained for the first time directly (Ruddock & Bradley 1976) and indirectly (Ippen

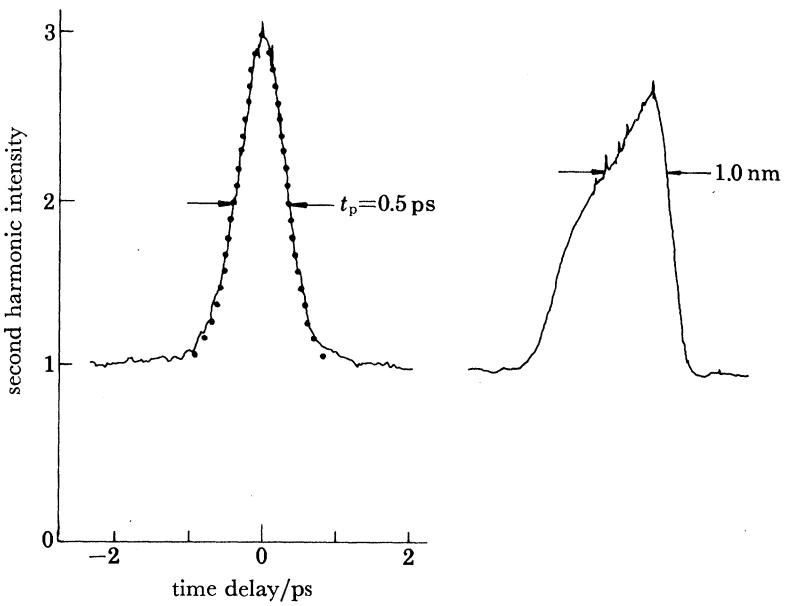

FIGURE 2. Second-harmonic autocorrelation trace of the first subpicosecond bandwidth-limited pulses obtained (Ruddock & Bradley 1976) from a mode-locked c.w. dye laser. The discrete points, corresponding to a sech^2 laser pulse intensity profile, show the excellent agreement with the theoretical soliton pulse shape.

& Shank 1975) from c.w. dye lasers. These pulses, which had sech^2 temporal profiles (figure 2), in excellent agreement with the theoretical model of Haus (1975), contained as few as 150 optical cycles. Since there are only 50 optical cycles (at 600 nm) in a 100 fs pulse it is clear that we are approaching the limit for ultra-short pulse generation in the visible spectral region. To produce shorter duration pulses it will be necessary to go to shorter wavelengths, e.g. for 10 fs pulses to use radiation of 60 nm wavelength (Bradley 1975, 1979). Work towards the generation of shorter pulses at shorter wavelengths is under way at the Blackett Laboratory.

Before beginning our discussions it is fitting to refer to Wheatstone's concluding remarks in his 1834 paper. 'By prosecuting these researches with instruments of higher power and of greater accuracy in their indications, numerical laws may be established for a large class of phenomena, the relations of which we have had hitherto no means of observing.' While Wheatstone could only refer to the study of electric current and discharges, today the list of such phenomena can now be extended to many fields of science and technology (table 1).

Finally, it should be noted that perhaps there is one great advantage that we have today over the earlier pioneers. Thanks to Planck and Einstein we know that ultra-short light pulses are indeed commonplace (figure 3) and that our eyes see with picosecond and femtosecond light pulses. What the well mode-locked laser does is to produce isolated bandwidth-limited pulses in a reproducible manner. All of the energy inside the laser cavity is concentrated into a single well behaved pulse propagating backwards and forwards between the two mirrors so as to emit a train of regularly spaced pulses through the partially transmitting mirror.

TABLE 1. APPLICATIONS OF PICOSECOND PULSES

1. *photochemistry*	intramolecular and intermolecular relaxation processes
2. *biology*	photosynthesis; dynamics of haemoglobin, visual molecules (rhodopsin) and DNA
3. *geophysics*	satellite and lunar ranging; continental drift; earthquake prediction
4. *solid state*	hot electrons; exciton dynamics; picosecond electronics; nonlinear optics
5. *extreme u.v. and X-ray coherent radiation*	medical diagnosis; phototherapy; holographic X-ray optics; integrated circuits
6. *compression*	fusion and fission: laboratory astrophysics
7. *stereometry*	3D measurement including oscillating or rotating bodies

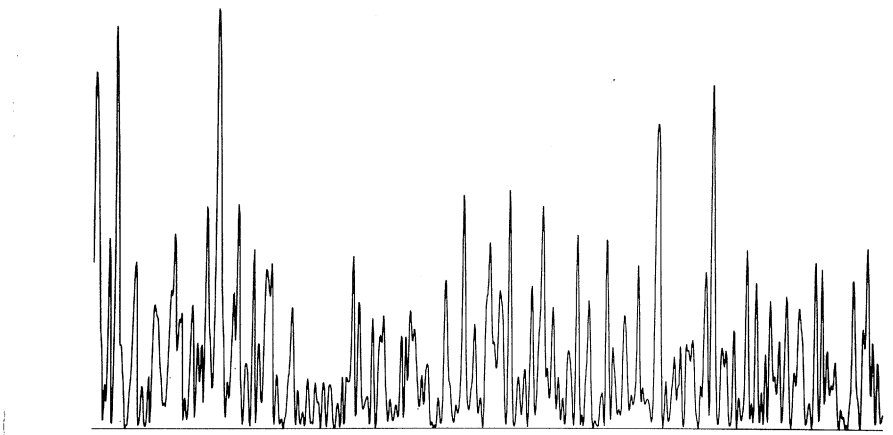

FIGURE 3. Intensity fluctuation pattern of 'thermal' light source, or of the below-threshold fluorescence emitted by laser-active media. In either case the shortest fluctuation has a duration determined by the inverse spectral bandwidth.

The first two papers deal with mode-locked c.w. dye lasers which are now capable of generating continuous trains of pulses with durations in the femtosecond range in the most controllable way. When properly mode-locked with a saturable absorber, the c.w. Rhodamine 6G dye laser can produce bandwidth-limited pulse durations as short as 200 fs with hyperbolic secant temporal profiles. The mode-locked laser then operates with 20 000 longitudinal modes (in a single transverse TEM_{00} mode) rigorously locked together in phase and amplitude. It is clear that such a unique light source will have many applications in science and technology, particularly in the field of coherent nonlinear interactions.

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

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PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES