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Phil. Trans. R. Soc. Lond. A 1982 **307**, 521-530
doi: 10.1098/rsta.1982.0127

Phil. Trans. R. Soc. Lond. A **307**, 521–530 (1982)
Printed in Great Britain

Mode-locked semiconductor lasers and their spectroscopic applications

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Double heterojunction semiconductor diode lasers operating in an external cavity can be actively and passively mode-locked to produce continuous trains of picosecond pulses of around 1 W peak power. These lasers, which are also frequency tunable, provide a convenient and cheap source of coherent ultra-short laser pulses for time-domain spectroscopy of semiconductor and molecular materials. With developments in laser diode processing techniques the spectral range could be extended to cover the visible to near i.r. region (up to 4 μm). Intracavity spectroscopy, with increased sensitivity, is also possible, particularly for the study of semiconductor carrier dynamics. Other applications include the study of coherent pulse propagation, two-photon spectroscopy and high-resolution spectroscopy when the diode lasers are operated in a single mode.

Introduction

The dominance of the organic dye laser in present-day spectroscopy is based upon the relative ease with which the wide homogeneously broadened fluorescence emissions of lasing dyes can be manipulated to produce tunable narrow-line or coherent ultra-short laser pulse outputs. After the initial experiments in producing narrow-band (Bradley *et al.* 1968*a*, *b*) and ultra-short pulse (Schmidt & Schafer 1968; Bradley & O'Neill 1969) dye laser modes of operation, these systems were quickly applied to atomic spectroscopy (McIlrath 1969; Bradley *et al.* 1970*a*) and to the investigation of coherent pulse propagation in resonance materials employing an electro-optical ultra-fast streak camera (Bradley *et al.* 1970*b*). Over the next decade there followed rapid and intensive development of instrumentation and techniques for high-resolution laser spectroscopy, and the flowering of this field can be seen in the work reported at this Discussion Meeting.

Similarly, there has been over the same decade equally rapid development of dye laser sources of ultra-short (picosecond and femtosecond) coherent pulses and of new picosecond time-resolving instruments based on linear (Bradley 1970) and nonlinear (Giordmaine *et al.* 1967; Weber 1967; Maier *et al.* 1966) detection systems. Although in frequency-domain laser spectroscopy the laser produced a revolution in accuracy and sophistication of the measurements, nevertheless the basic concepts and instruments existed before the invention of the laser. This was not so with time-domain laser spectroscopy. Here, although the laser, and in particular the dye laser, permitted the generation for the first time of ultra-short coherent pulses tunable in frequency, it took nearly a decade and a half for the development and expansion of the laser and measurement techniques into the picosecond and subpicosecond region (see Bradley (1977) for references). These dramatic advances in the techniques of generating ultra-short light pulses from mode-locked lasers, and of picosecond and shorter detection and measurement, are being increasingly applied by chemists, biologists, physicists and engineers for the study of previously unresolved ultra-fast processes of significance in their respective fields of science and technology. Previous developments were surveyed in an earlier discussion meeting (Bradley *et al.* (eds) 1980).

A major factor inhibiting the wider application of coherent picosecond and femtosecond time-domain spectroscopy has been the cost of the laser sources and the associated measurement equipment, particularly if sensitive linear detection is required. Reduction of the cost of synchronously driven electron-optical picosecond streak cameras (Bradley (1978), and references therein) could be achieved by employing image dissection read-out inside the streak image-tube (Bradley 1972). Experiments towards this end are under way in our laboratory. However, there remains the considerable cost of a mode-locked continuous wave (c.w.) dye laser, pumped by an argon ion laser, as commonly used for time-domain spectroscopy. With the successful development of room-temperature c.w. double heterojunction (d.h.) semiconductor diode lasers (see Casey & Panish (1978) and Thompson (1980)) capable of operation in the spectral region from *ca.* 600 nm to *ca.* 4 μm, it is now possible to consider replacing dye lasers as tunable light sources for both time-domain and frequency-domain spectroscopy. So far, d.h. room temperature laser diodes have been based upon III–V semiconductors, but work on II–VI laser materials is beginning to show promise for operation in the blue, green and yellow spectral regions (Yao *et al.* 1979). Thus, in the same way that the flashlamp-pumped frequency-tunable dye laser played a key role in 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 (see Bradley & New (1974) for references), the later developments in mode-locked c.w. dye lasers can be immediately transferred to diode lasers.

For these reasons some 5 years ago we decided that it would be worth while to investigate methods of mode-locking d.h. diode lasers to produce tunable coherent picosecond pulses. While similar work started at M.I.T. under H. A. Haus produced picosecond pulses from c.w. diode lasers (Ho *et al.* 1978), the first bandwidth-limited coherent optical pulses were obtained by the use of a special d.h. laser designed to suppress internal mode structures completely in a diode laser operating in an external cavity (Holbrook *et al.* 1980*a*, *b*). Passive mode-locking produces the shortest laser pulses (Ippen *et al.* 1980; Yokoyama *et al.* 1982) but the lasers employed have to be specially aged or proton bombarded (van der Ziel *et al.* 1981) to produce a region of saturable absorption. The most stable mode of operation, combined with the production of high peak-power bandwidth-limited picosecond pulses, is achieved when the diode laser is electrically pumped by a train of short (electrical) pulses in a manner analogous to the synchronously pumped c.w. dye laser in which the dye jet stream is optically pumped by the pulses from a mode-locked ion laser.

Actively mode-locked GaAlAs diode laser

Ultrashort optical pulse generation with semiconductor diode lasers is currently of wide interest for such applications as high bit-rate optical communications and very fast data processing as well as for picosecond and femtosecond time-resolved spectroscopy. While much work has been carried out on eliminating self-pulsations in laser diodes (Walpole *et al.* 1980), emphasis has now switched to investigations into the mode-locking of semiconductor diode lasers by using an external cavity configuration to produce ultrashort fully coherent pulses.

The first actively mode-locked c.w. diode lasers (Ho *et al.* 1978; Glasser 1978) produced pulses lasting about 20 ps. However, residual reflectivity at the cleaved diode facets interfered with the mode-locking process inside the external cavity and the pulses contained temporal substructures. By employing an intracavity etalon to limit the lasing bandwidth to only one longitudinal mode of the subcavity formed by the diode facets, temporally smooth pulses of duration *ca.* 60–30 ps were obtained (Ho 1979; Ito *et al.* 1980). The complete suppression of internal mode structure in a diode laser operating in an external cavity has been achieved (Holbrook *et al.* 1980*a*) by the combination of antireflexion coatings and the use of a GaAs–GaAlAs d.h. laser with the stripe contact making an angle of 5° to the normal of the diode facets. This approach permitted the generation of the first bandwidth-limited pulses from an actively mode-locked c.w. diode laser (Holbrook *et al.* 1980*b*). Employing a streak camera driven continuously in synchronism (Adams *et al.* 1978) with the laser pulse train made for easy optimization of the mode-locking process and direct measurement of the pulse shapes and durations. With indirect nonlinear pulse duration measuring techniques, such as second-harmonic generation optical intensity correlation, unambiguous pulse shapes are not obtained

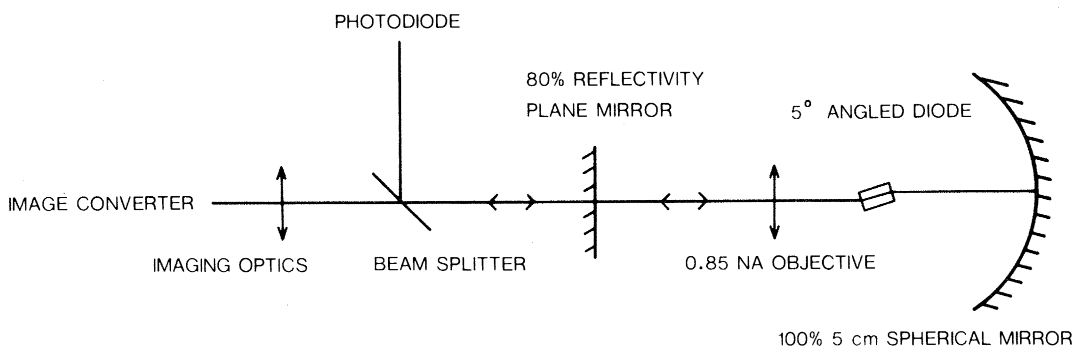

FIGURE 1. Experimental arrangement of the actively mode-locked diode laser.

and it is not possible to detect low-intensity but long-lasting sections of the laser output. These may contain a substantial proportion of the laser energy outside the ultrashort pulses themselves, as was found with the first synchronously pumped mode-locked dye lasers (Bradley *et al.* 1969).

With the experimental arrangement shown in figure 1, bandwidth-limited pulses of 16 ps duration are obtained with very low background energy content between the pulses of the mode-locked pulse train. The GaAlAs d.h. injection laser had an 18 μm wide insulated stripe, of length 0.5 mm, rotated on the chip by 5° from the conventional position normal to the cleaved facets. Following the approach that first gave bandwidth-limited picosecond pulses from dye lasers (Bradley 1978; Arthurs *et al.* 1971), bandwidth control was obtained by the introduction of an optically contacted, 50 μm gap, wedged Fabry–Perot interferometer filter (Bradley *et al.* 1968*a*) with 60% reflectivity coatings. The lasing bandwidth was thus reduced to 0.008 nm. An iris of 3 mm diameter was combined with a 1 mm slit aperture mounted perpendicular to the junction plane of the diode to prevent laser filamentation and multi-transverse-mode operation. With the Fabry–Perot filter tuned to 860 nm the d.c. threshold was 142 mA. The laser spectrum was monitored by a scanning Fabry–Perot interferometer of free spectral range 0.41 nm and a finesse of more than 60. Without the intracavity apertures the lasing spectrum was unstable above threshold and several components of the same (0.008 nm) bandwidth were generated.

Active mode-locking is produced when the d.c. bias current is reduced to 120 mA and the diode is modulated by r.f. power at 375.5 MHz, corresponding to the round-trip frequency of the laser resonator. Bandwidth-limited pulse generation requires that the d.c. bias current be kept between 120 and 122 mA and that the r.f. modulation frequency be maintained on resonance with the laser cavity to within 10 kHz. The streak camera has a Photochron II streak tube (Bradley (1978), and references therein) operating at a photocathode (S20) extraction field strength of 20 kV cm^{-1}. The tube was driven at the fourth-subharmonic frequency of the laser diode r.f. modulation and the relative phases were arranged (Adams *et al.* 1979) so that pulses were recorded on the linear portions of the sine-wave deflecting voltage. Thus in figure 2 the two pairs of pulses generated in a Michelson interferometer optical delay line (Ryan *et al.* 1978) are dispersed temporally in opposite directions. Calibration of the streak camera writing

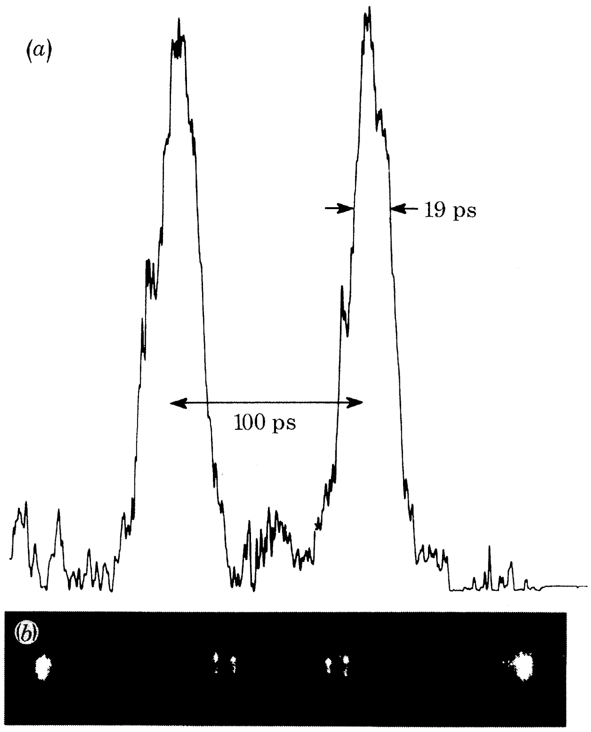

FIGURE 2. (*a*) Microdensitometer trace of streak camera record of pair of pulses (separated by 100 ps) generated in Michelson interferometer optical delay line. The ordinate is a linear density scale. (*b*) Streak photograph of bandwidth-limited pulses. The unresolved pulses at either end occur at the turning points of the sinusoidal deflexion voltage waveform when streak camera writing speed is momentarily zero.

speed was easily obtained from the known separation of the pulse pairs, and linearity of the streak was confirmed by moving the positions of the pulses by changing the relative phases. At the writing speed employed (10^9 cm s^{-1}), the camera resolution was *ca.* 10 ps. This experimental arrangement allowed simultaneous real-time observation of both the spectral and temporal profiles of the laser pulses. It was then relatively easy to adjust the laser for optimum mode-locking performance. The streak camera record and the corresponding linear density microdensitometer trace are shown in figure 2. After deconvolving the camera resolution of 10 ps, a pulse duration of 16 ps is obtained. From the corresponding spectral profile a time–bandwidth product value of $\Delta\nu\Delta t = 0.36$ is derived, to be compared with the theoretical values of 0.44 and 0.32 for gaussian and $sech^2$ pulses, respectively.

Effect of modulation frequency drift on pulse shapes

To investigate the dependence of the laser pulse shape on the modulation frequency, the film recording medium previously employed was replaced by a Hamamatsu type 145 optical multichannel analyser. This device provides a continuous video picture of the streak camera phosphor screen and an intensity profile can be recorded, at will, with signal integration if desired. The effect of detuning the modulation frequency can be seen in figures 3 and 4. It is clear that the active mode-locking process is sensitive to a detuning of as little as 10 kHz

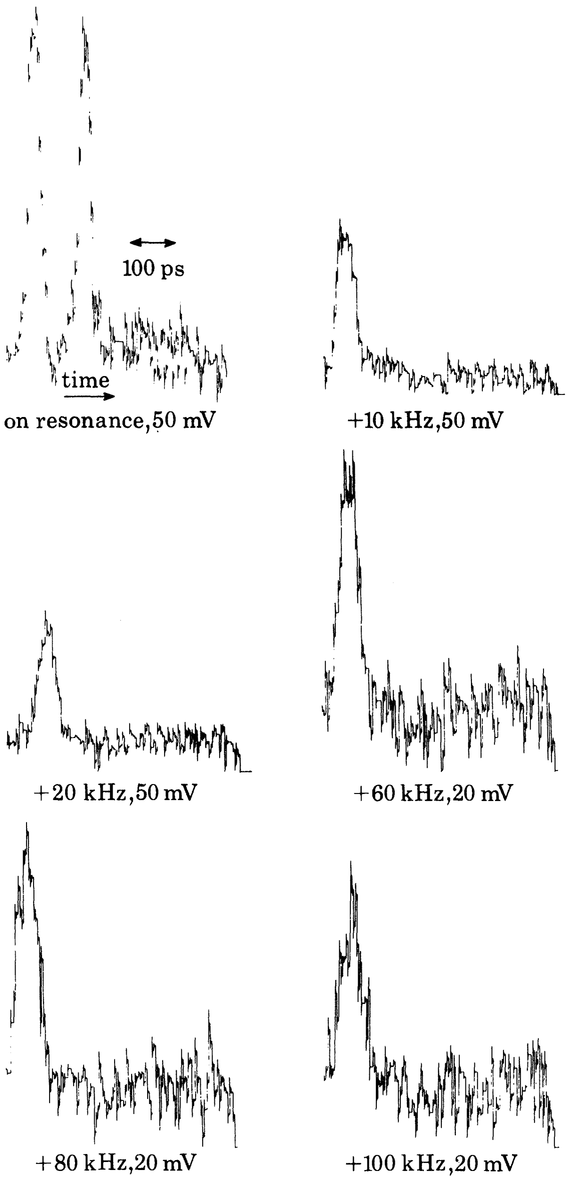

FIGURE 3. Series of streak camera (linear intensity) traces showing the effect of positive frequency detuning of the diode laser r.f. modulation. The temporal scale is shown in the first trace but in subsequent traces one arm of the calibrating Michelson interferometer was blocked. Each trace represents five frame integrations and the digital gain was ×50. Two intensity scales were used for the ordinate: the scale used is indicated on each trace, together with the amount of detuning from the optimum modulation frequency.

(*ca.* 0.002 % of the modulating frequency). This sensitivity is comparable with that found with other actively mode-locked lasers, such as the argon ion laser with a sensitivity to frequency detuning of 0.05 % (Ryan 1978), and the synchronously pumped dye laser with a sensitivity of 0.0001 % (Ausschnitt *et al.* 1979). Positive frequency detuning (figure 3), which produces a delay so that the optical pulse arrives later at the diode than the pumping pulse, leads to a gradual broadening of the output pulse shape. Negative frequency detuning (figure 4) in which the laser pulse arrives at the diode before the peak of the gain, leads to the production of a pulse tail. When detuning exceeds 40 kHz a double pulse evolves. The secondary pulse increases

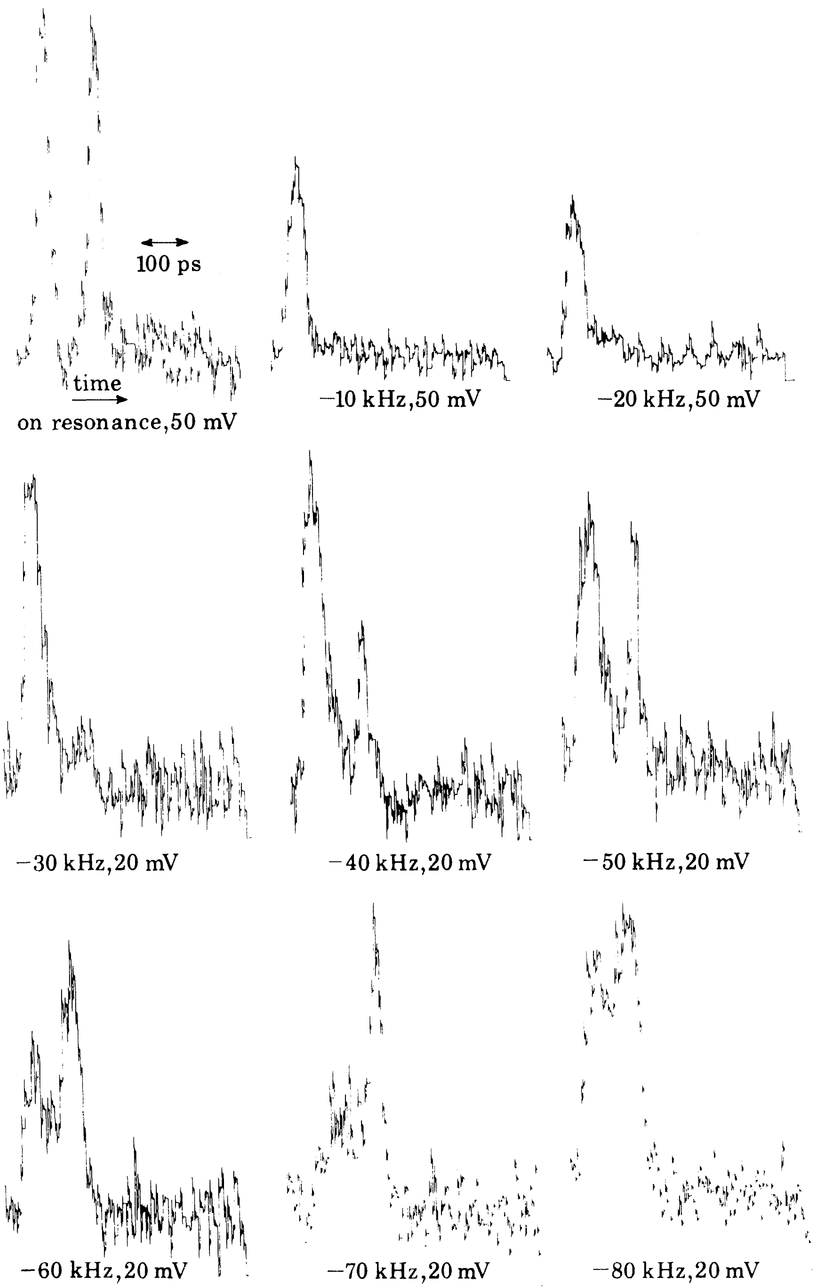

FIGURE 4. As in figure 3 but with negative frequency detuning.

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in magnitude, becoming dominant at 60 kHz detuning. At a detuning of 80 kHz the two sub-pulses merge into a broad pulse of *ca.* 100 ps duration. An interesting feature of these results is that the pulses are still clearly recorded with the streak camera being driven synchronously at the modulation frequency. If the pulses were emitted at the laser cavity frequency the streak camera would be constantly 'strobed' at the difference frequency and no well defined pulse profile would be observed. The intensity profiles of figures 3 and 4 were obtained after five television frame integrations.

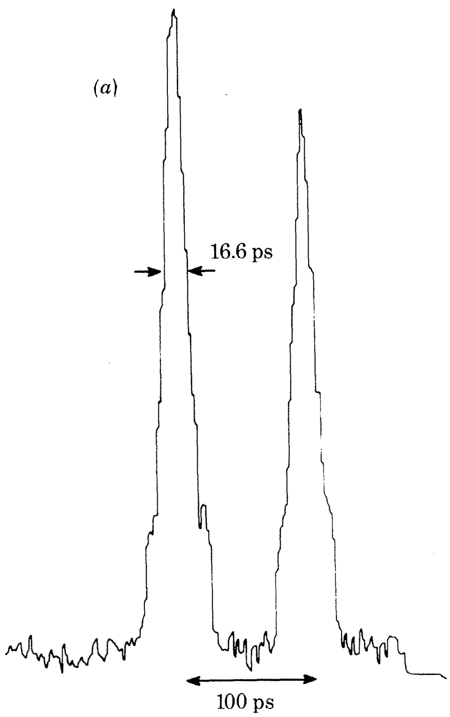
Saturation of the optical gain by the pulse circulating inside the external cavity results in pulse shortening (New 1974), with the eventual establishment of a bandwidth-limited pulse. If the pulse arrives too late, noise immediately preceding the pulse sees gain and the front edge of the pulse is broadened. Likewise, too early an arrival leaves sufficient gain for amplification of the trailing edge, or even amplification of a second pulse when the gain recovers. In both cases the output pulse train has the periodicity corresponding to the laser modulation frequency, since the pulse shape continually adjusts its shape to maintain synchronism (Frigo *et al.* 1977).

Electrical synchronous pumping with a snap diode

In an effort to produce shorter pulses and to permit operation above threshold to obtain both higher output power and greater stability, the sinusoidal modulation was replaced by the repetitive waveform produced by a 'snap' or 'step recovery' diode (Hewlett–Packard type 5082/0822), shunted antiparallel to the laser diode. The resulting output voltage pulse has a rise time of less than 150 ps and an f.w.h.m. of *ca.* 300 ps. With this more rapid pumping pulse the d.c. bias current could be significantly increased before gain recovery caused multiple pulsing. A further consequence was that extra loss could be introduced into the laser external cavity without quenching the laser action. Thus, alignment drift became much less critical. Figure 5*a* shows the pulse profiles when a snap diode was employed in the modulation circuitry. Deconvolving the camera resolution limit of 10 ps gives a pulse duration of 16 ps. From the corresponding scanning Fabry–Perot spectral width of figure 5*b*, a time–bandwidth product of 0.8 was obtained. This slight departure from strict bandwidth-limited operation may arise from modulation jitter in the driving electronics or from jitter in the periodicity of the pulse train. In either case, with synchronously driven streak camera detection, the recorded pulse profiles would be broadened, as already found with synchronously mode-locked and synchronously detected c.w. dye lasers (Adams *et al.* 1979). Thus the pulses could have been shorter than 16 ps. The sharper r.f. modulation did definitely produce the other beneficial effects expected. A pulse duration of not more than 16 ps could be obtained routinely because alignment was not so critical above threshold and the value of the d.c. bias current itself was no longer as critical for maintaining good mode-locking operation. As can also be seen from figure 5*a*, the snap diode modulation removed practically all of the residual interpulse noise seen in figures 3 and 4.

Further developments

Active mode-locking of a GaAlAs c.w. diode laser has produced pulses as short as 16 ps with a time–bandwidth product of 0.36. This was the first observation of bandwidth-limited pulses from an actively mode-locked injection laser. With sinusoidal modulation of the diode current at the external cavity round-trip frequency, the occurrence of good mode-locking was found to be critically dependent upon maintaining the d.c. bias current just on threshold. In addition, the modulation frequency of *ca.* 375 MHz has to be maintained in resonance with the cavity transit time to one part in 10^5. When the sine-wave r.f. modulation was replaced by repetitive pulses of 300 ps duration, more stable mode-locking was obtained when the laser was biased significantly above threshold with a 30% increase in output power to give pulses of *ca.* 1 W lasting 16 ps or less. By following the approach adopted for mode-locked c.w. dye lasers (Ryan *et al.* 1978), pulses as short as *ca.* 1 ps or less should be obtainable from a hybrid system in which active mode-locking is supplemented by the insertion of a saturable absorber into the laser cavity. The most convenient method of achieving this would be the inclusion of a second d.h. diode laser inside the external laser resonator cavity. Changing the d.c. electrical



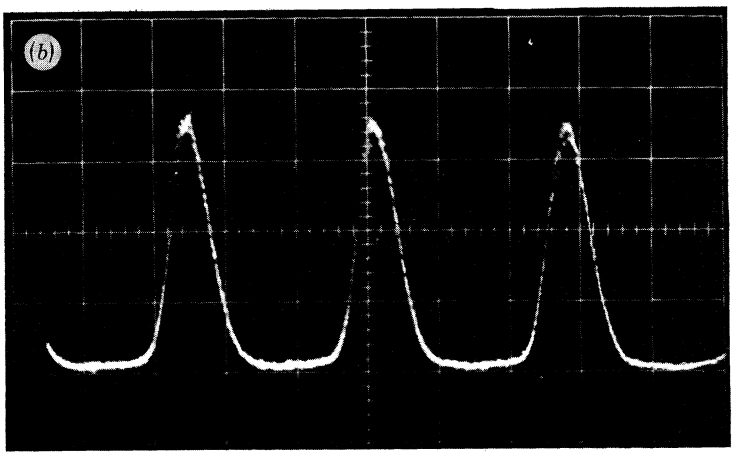


FIGURE 5. (*a*) Streak camera (linear intensity) recorded pulses from mode-locked diode laser modulated by snap diode circuit. Deconvolving synchronously driven camera resolution limit of 10 ps gives a pulse duration of 16 ps. (*b*) Simultaneously recorded scanning Fabry–Perot spectrum of pulses of (*a*). F.w.h.m. spectral bandwidth is 0.12 nm.

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current through the second laser diode affects the amount of gain or loss introduced. By connecting two separated regions of a homostructure semiconductor laser to a common r.f. source, ultra-short light pulses were first generated from a diode laser (Basov *et al.* 1966). Later, when the two parts of the laser diode (operating at cryogenic temperatures) were excited by independent current generators, the pulse duration (*ca.* 10^{-10} s) and repetition frequency could be controlled by variation of the electrical current through one half of the diode structure. We are studying the mode-locking performance of two independently modulated d.h. diode lasers in both Fabry–Perot and ring external resonator configurations. This seems the most promising approach to the generation of coherent subpicosecond pulses from diode lasers being an arrangement analogous to that which has produced pulses shorter than 100 fs in a ring c.w. dye laser (Fork *et al.* 1981). If successful in operation, such a double laser diode resonator would provide an excellent source for intracavity absorption and fluorescence spectroscopy of semiconductor and molecular samples.

Summary and discussion

In conclusion, it is clear that semiconductor d.h. diode lasers hold great promise as convenient and reasonably cheap sources of frequency-tunable coherent picosecond and subpicosecond pulses for time-domain spectroscopy. With improvements in the techniques of laser growth arising from new processes based upon molecular beam epitaxy and metal–organic chemical vapour deposition highly advanced semiconductor heterostructures could become routinely available. Indeed, it is possible to anticipate an integrated spectrometer, with the laser source, its driving and control circuits, the spectral or temporal dispersion elements and the detector grown on a single substrate. This approach could revolutionize spectroscopy since it would be possible to incorporate such a micro-chip spectrometer, for example, in the wall of a chemical reaction vessel. However, it is likely that we shall first of all have to investigate the detailed physics of diode lasers (in the same way as it was necessary to carry out detailed photochemical studies of laser dyes and saturable absorbers) before we can bring the semiconductor laser to the same state of perfection that the dye laser has now achieved, particularly for femtosecond coherent pulse generation. Recent results in controlling the spectral characteristics of external-cavity semiconductor lasers has produced narrow-band (*ca.* 1 MHz) emission tunable over 10 nm from a d.h. laser (Fleming & Mooradian 1981) and picosecond pulses tunable from 770 to 890 nm have been obtained by intense photoexcitation of a thin GaAs crystalline film pumped by a mode-locked dye laser (Damen *et al.* 1981). In addition, power-independent line-width broadening of a c.w. single-frequency diode laser, attributed to refractive index fluctuations resulting from statistical fluctuations in the number of conduction electrons in the small active volume of the device, has also been experimentally observed (Welford & Mooradian 1982). These are examples of the interesting physics involved in semiconductor laser investigations.

The authors acknowledge the financial support of the National Board for Science and Technology, and the S.E.R.C.

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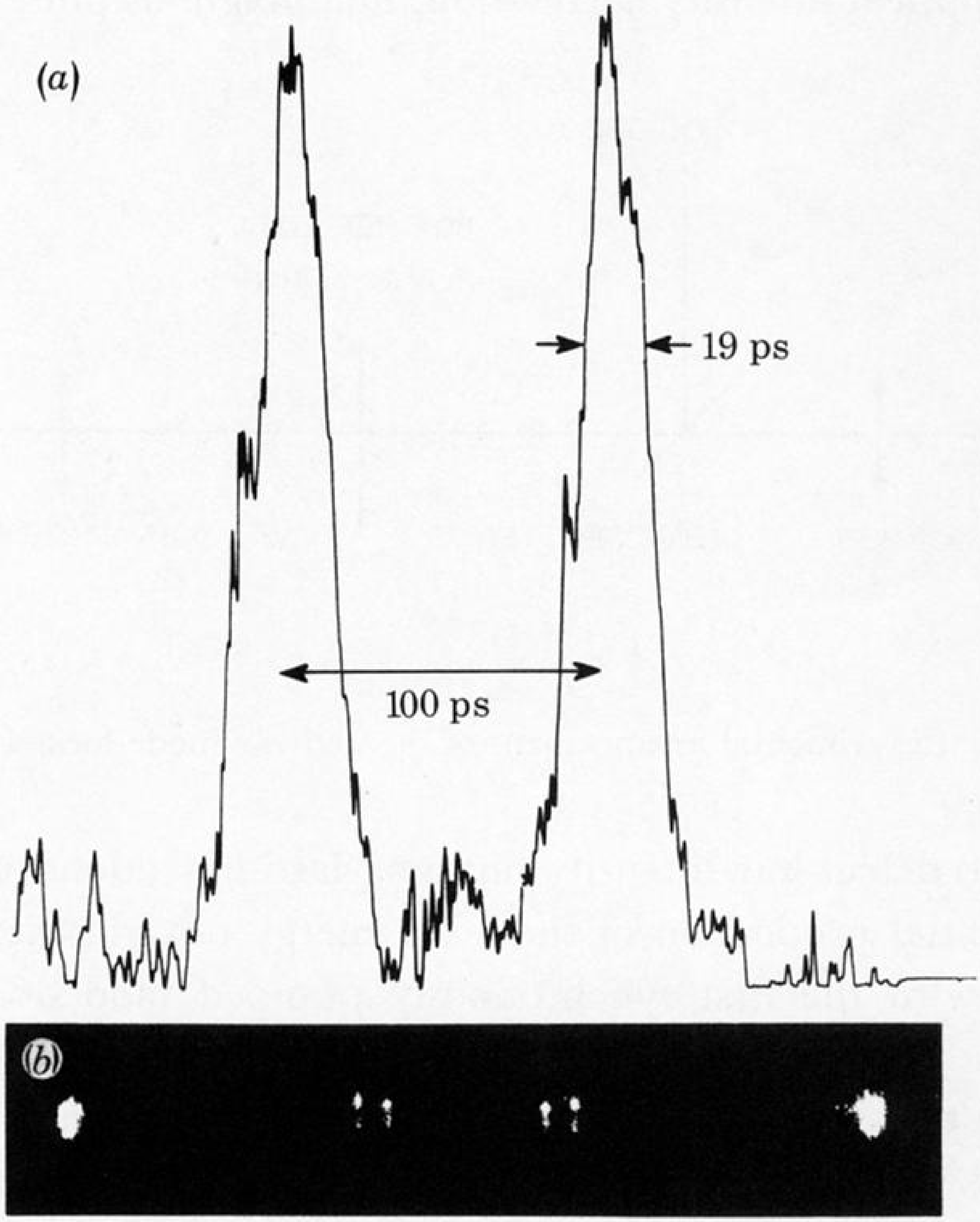

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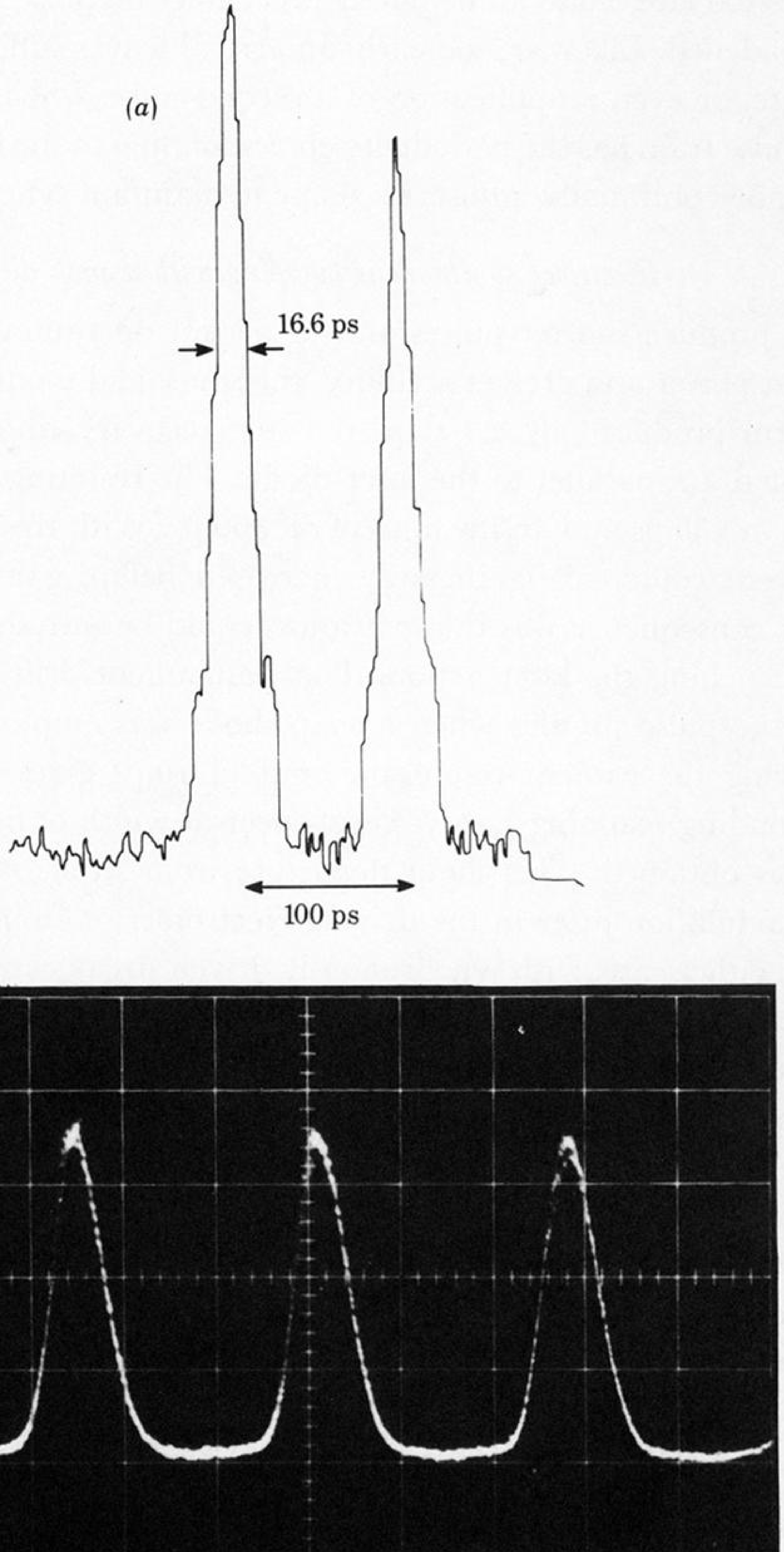

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