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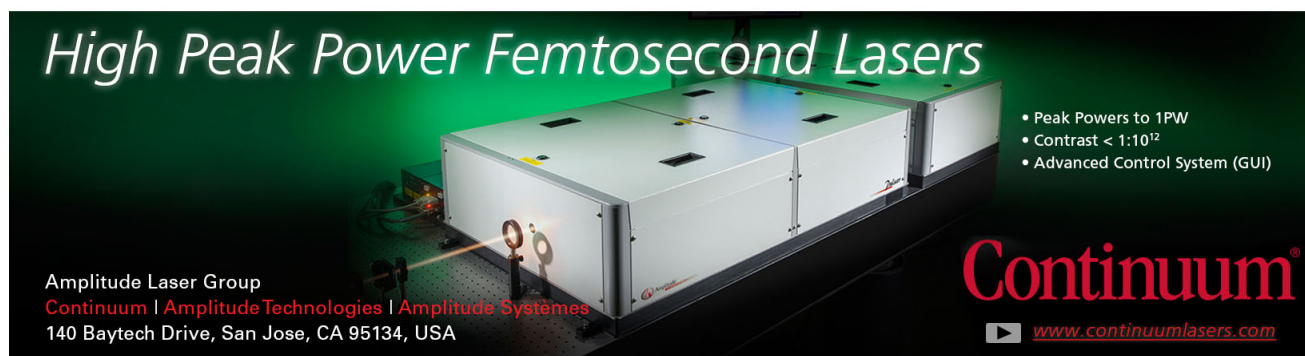
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A single-shot autocorrelator for the ultraviolet with a variable time window

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A single-shot autocorrelator for the ultraviolet based on multiphoton ionization of gases has been developed. It makes use of a modified Mach-Zehnder interferometer and a segmented two-photon-ionization chamber as a spatial square-of-intensity detector. The device has in principle a continuously variable temporal detection range from ~ 50 fs to 50 ps.

INTRODUCTION

Autocorrelation methods based on second harmonic generation or two-photon fluorescence are generally not adaptable to the ultraviolet wavelength region, because of the lack of suitable nonlinear crystals or the practical difficulty of detecting the two-photon excited signals. It has been demonstrated¹⁻⁴ that pulse duration measurements can be carried out by using multiphoton ionization (MPI) of gases as the nonlinear detection mechanism. The photo-ionization detection technique is a highly sensitive method; its unique property, compared to the other autocorrelation detection methods, is that the output to be observed is an electric current instead of a light signal, thus providing easy detection.

Nowadays, autocorrelation based on two-photon ionization (TPI) of gases is a commonly used technique for the temporal study of short excimer laser pulses.⁵⁻¹² By proper choice of the gas, autocorrelation measurements were performed at 308 nm,⁵⁻⁶ 351 nm,⁷ 248 nm,⁸⁻¹⁰ 276 nm,¹¹ and 193 nm,¹² using triethylamine,^{5,7} DABCO,⁶ and NO⁸⁻¹² gases. In the above experiments the TPI gas chamber followed a standard Michelson interferometer, which was used to produce a delay between the two partial beams at the output. This delay was changed in equidistant steps from shot to shot, and the time-averaged autocorrelation function was obtained by recording the ion current as a function of the delay, using multiple (10^2 - 10^4) shots.

The only single-shot autocorrelation measurement based on TPI is reported by Bourne *et al.*¹³ In that experiment, two counterpropagating picosecond pulses created a spatially dependent charged-particle cloud. By applying a dc electric field parallel to the two beams, the electrons were propelled towards the collection electrode. The electron flight time was thus proportional to the distance between their position and the collection electrode. The spatial distribution of the electron cloud could then be determined simply by measuring the temporal profile of the current at the collection electrode. The main drawback of this simple and elegant method is its limited resolution ($\sim 1:20$) and its limited time scale (26-600 ps). The resolution (and therefore the lower limit of the time scale) can be somewhat improved by a change of the detection geometry (as suggested in the same Ref. 13) by the use of a transverse drift field adjacent to the overlap zone and an electronically scanned diode array designed for electron detection. However, no subpicosecond

resolution is expected from this system, even with much improved resolution of the detection, while maintaining the counter-propagation-pulse geometry. By contrast, the lower limit of the temporal resolution can be extended to the femtosecond region, and can be made independent of the spatial resolution of the detection by using two beams propagating in the same direction, whose pulse fronts enclose a small angle. This can be produced by either having two parallel partial beams with tilted pulse fronts,¹¹ or two partial beams enclosing a small angle¹⁵ or the combination of both.¹⁶ However, in this case, the accuracy of single-shot autocorrelation measurements becomes strongly influenced by the homogeneity of the measured beam. Since the output of large-aperture, high-brightness excimer lasers is generally homogeneous and the spatial distribution of the output beam can be characterized by a flat-topped function, it is possible to perform accurate single-shot autocorrelation measurements even without the use of a reference beam. On the other hand there are cases, where instant information on the temporal behavior of a single pulse is needed even at the expense of an eventual decrease of the accuracy, which makes the availability of single-shot autocorrelation measurements necessary.

For the above reasons we have developed a single-shot autocorrelator for the ultraviolet based on MPI, which is free of the above-mentioned limitations even for the shortest pulse durations. In this paper, the results of this development are presented with special emphasis on the following aspects: (1) sensitive and high-resolution spatially-distributed nonlinear TPI detection geometry, using transversal drift field and segmented electrodes, (2) simple interferometer design for high power pulses of short wavelength, providing two spatially and temporally overlapping partial beams at the detector with a variable angle between them, and (3) elimination of the dependence of the shape of the autocorrelation function on the spatial intensity distribution of the beam.

I. CONSTRUCTION OF THE AUTOCORRELATOR

The schematic of the single-shot TPI autocorrelator is shown in Fig. 1. It consists of a "misaligned" Mach-Zehnder interferometer, whose output consists of two temporally and spatially overlapping partial beams at the position of the detection, where they enclose a small angle α . The

advantage of using this interferometer instead of the generally used Michelson interferometer is that it has less components and the conditions for the two partial beams being identical (number of reflections, refractions, etc.) can be easily set.^{9,17,18} A disadvantage when using a Mach-Zehnder interferometer for autocorrelation measurements is that the relative timing of the two partial beams cannot be changed as easily and in as well a controlled way as with a Michelson interferometer, where it can be simply done by moving one mirror in one of the two arms of the interferometer. In the Mach-Zehnder interferometer, tuning of the two partial output beams can be accomplished by synchronized movements of at least two mirrors as will be shown in the following. The simplest solution is to rotate the beamsplitter and the mirror of one arm (e.g., BS1 and M1 in Fig. 1) leaving them always parallel. Experimentally, we achieved this by mounting both mirrors on a common rotating holder having its center of rotation just symmetrically in between the two components. Then a small rotation of the holder ψ results in a delay D of the same sign at both components while leaving the direction of the output beam unchanged and introducing only a minor shift Δs of the output beam. The pathlength difference D is given by

$$D \approx \Delta d - \Delta s = (L/2)\sin\varphi - (L/2)(1 - \cos\varphi), \quad (1)$$

where L is the pathlength between the beamsplitter and the mirror (Fig. 1). It is seen from Eq. (1) that the delay is a function of φ . However, when working with short pulses, the necessary delay D is small compared to L . (In our case $D/L \approx 3 \times 10^{-3}$, thus the value of ψ is in the same order of magnitude.) Then Δd and Δs can be approximated as

$$\Delta d \approx (L/2)\varphi, \quad (2)$$

$$\Delta s \approx (L/4)\varphi^2, \quad (3)$$

resulting in

$$D \approx \Delta d. \quad (4)$$

The sum of the delays introduced by both components then is $2D$. The practical importance of Eq. (4) is that the delay can be a linear function of setting of a properly positioned micrometer screw, used for rotation of the beamsplitter-mirror pair mounting.

II. CONSTRUCTION OF THE TPI DETECTOR

The interferometer was followed by the TPI gas chamber, which was situated where the two output beams of the interferometer were in spatial overlap (Fig. 1). The construction of the TPI gas cell is shown in Fig. 2. It consists of an array of 16 collection electrodes ($20 \times 1 \text{ mm}^2$ each) acting as a common, segmented plane-electrode. The 16 collection electrodes—having 0.25 mm separation—cover a $20 \times 20 \text{ mm}^2$ -area, which is surrounded by a 7-mm-wide, electrically-grounded guard ring. This is used to get a well characterized E -field in the collection zone. Each collection electrode is held at ground potential by a 44-M Ω resistor and is connected to a separate charge amplifier (see below). The field-electrode is separated by 2.5 mm from the plane of the collection electrodes and connected to a -250-V dc potential. The polarity and the value of this potential are chosen to obtain high spatial resolution, when the lower electrode array is used for electron collection. Increase of the accelerating voltage resulted in an increase of the electric signal at the collection electrodes and a slightly improved spatial resolution. Above 250 V, the increase of the output signal and the

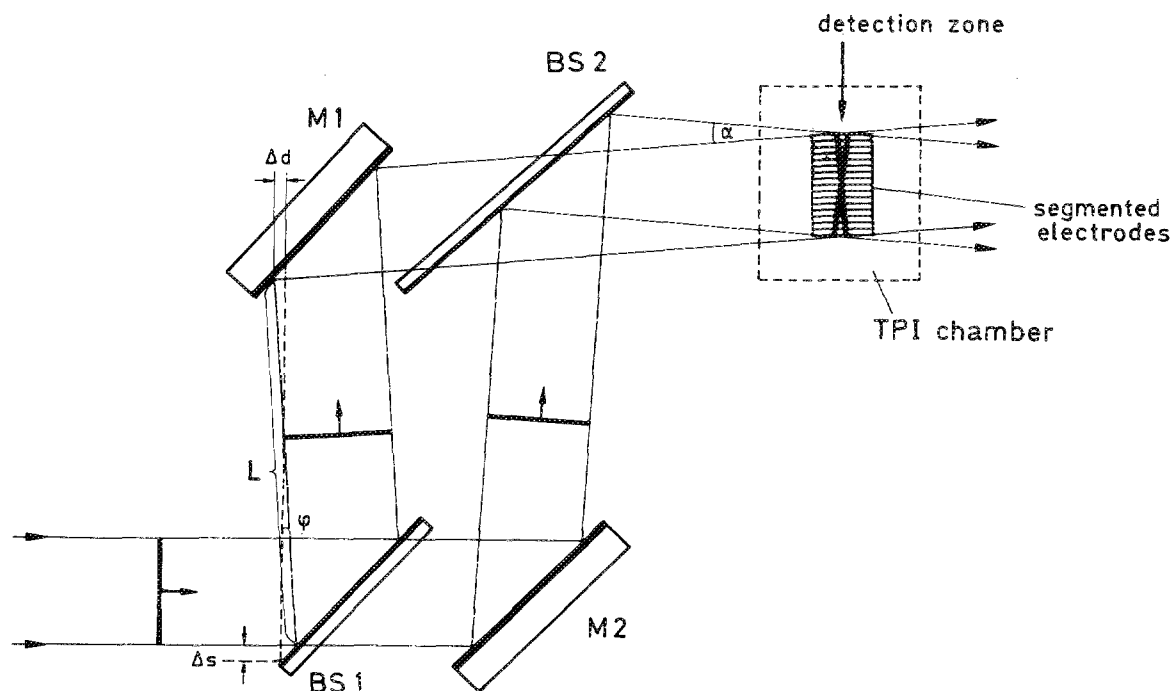


FIG. 1. Schematic of the single-shot TPI autocorrelator.

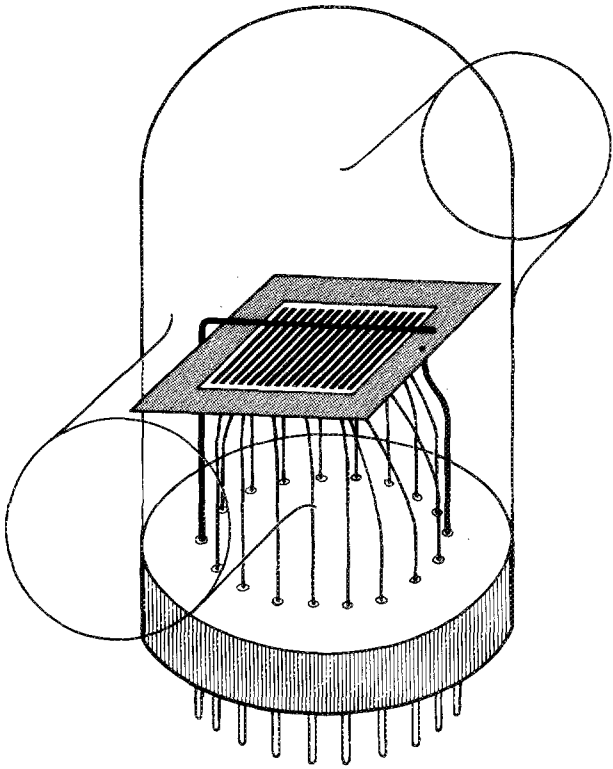


FIG. 2. Schematic of the spatial TPI square of incident intensity detector.

resolution showed saturation. In order to get optimum spatial resolution, the direction of the longer axis of the lower collection electrodes should be set parallel to the plane bisecting the angle between the two beams. The reason for using a wire as a field electrode (Fig. 2) is that in this case the electric field strengths has a maximum at the field electrode just in the middle of the collecting electrodes. Therefore,

mainly those electrons are collected which were created in the middle of the collection electrodes (called detection zone in Fig. 1) resulting in less sensitivity of the above-mentioned relative adjustment of the detector with respect to the beam. If a plane electrode was used as a field electrode, resulting in a homogeneous electric field along the collection electrodes, all the electrons would be collected from that volume, which is defined by the plane of the collection, and the upper plane electrode. In this case the adjustment of the detector is more critical. However, one would have less sensitivity on the spatial inhomogeneities of the beam as will be shown later (Sec. V).

The number of the collection electrodes was matched to the capacity of the multiplexer used in the electronics (see below) and was restricted to 16, imposing some limitation on the resolution. However, the problem related to the limited number of points from which the autocorrelation curves are obtained could be minimized by proper match of the width of the autocorrelation curve to the time window of the measurement. An example for the typical operational conditions is given in Fig. 6, where the autocorrelation width $\Delta\tau$ is $\sim 1/3$ of the time window T . This condition can be achieved for a given input pulse by proper choice of the time window of the measurement, which can be changed continuously by changing the angle α between the two beams. It can be shown that the time window T of the measurement is defined by the equation

$$\sin(\alpha/2) = cT/l, \quad (5)$$

where c is the speed of light, l is the length of the collection zone (parallel to the upper electrode). Considering that in optimum conditions $T \approx 3\Delta\tau$, the optimum value of α is

$$\alpha \approx 2 \arcsin(3c\Delta\tau/l) \quad (6)$$

for a pulse characterized by an autocorrelation width $\Delta\tau$.

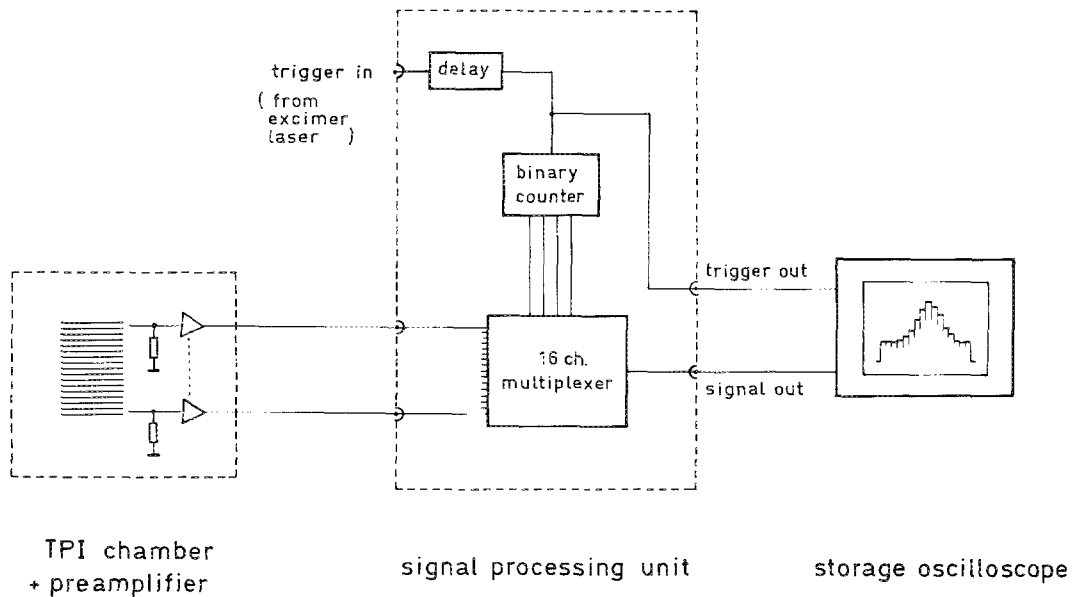


FIG. 3. Lay-out of the electronics.

III. THE ELECTRONICS

A schematic of the electronics is shown in Fig. 3. Each of the 16 collecting electrodes was grounded through a $R = 44\text{-M}\Omega$ resistor and connected to a JFET-input operational amplifier (TL 084), which was used in the unity-gain, noninverting mode as impedance transformer. The four TL 084 ICs—each containing 4 amplifiers—were mounted on the TPI chamber close to the electrodes to minimize the coupling capacitance of the connections to the collection electrodes, and to minimize the electromagnetic interference (EMI) pick-up. The outputs of the amplifiers were connected to the signal processing unit with a $\sim 2\text{-m}$ -long cable which also contained the supply leads. This construction made it possible to move the TPI chamber independent of the main electronics, and resulted in a very low noise level even in a high EMI environment. The output pulses of the amplifiers were sequentially read out—just at their maxima—by a 16 channel CMOS analog multiplexer (Analog Devices ADG 506 AKN). The four binary addresses of the multiplexer were driven by a binary counter, which was started by the delayed trigger pulse of the excimer laser.

The upper trace of Fig. 4 shows the temporal behavior of the output pulse from one of the electrodes after the preamplifier, under typical operational conditions. The horizontal scale is $50\ \mu\text{s}/\text{div}$, the vertical is $100\ \text{mV}/\text{div}$. Multiplexing of the 16 outputs of the preamplifiers was started just before the maximum of the electric signals, which could be adjusted by the delay in the processing unit. The speed of multiplexing was adjusted to read out each of the 16 channels at the plateau of the signals from the electrodes by proper choice of the frequency of the counter. The lower trace in Fig. 4 shows relative timing of multiplexing of the 16 channels with respect to the input electric signal shown in the upper trace. In this measurement, the oscilloscope was triggered externally by the trigger output of the excimer laser. The signal processing unit also has a trigger output to study the multiplexed signal output at a higher time resolution. The traces in Figs. 5 and 6 were recorded using this triggering.

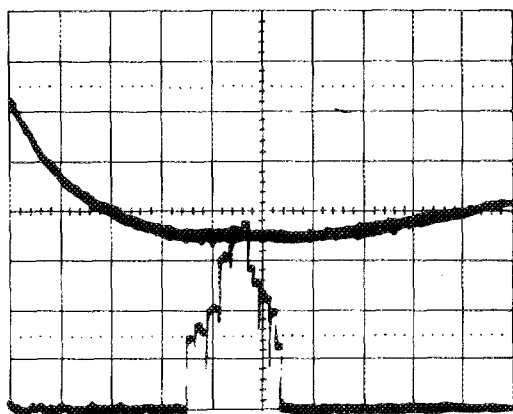
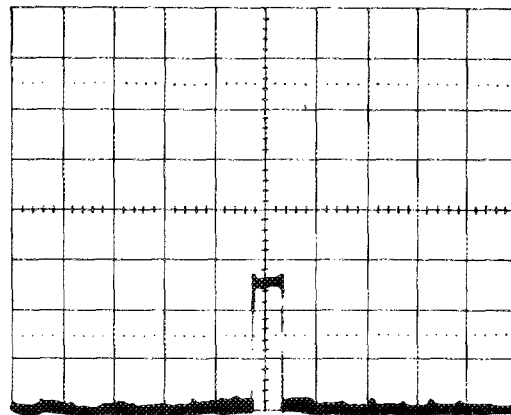
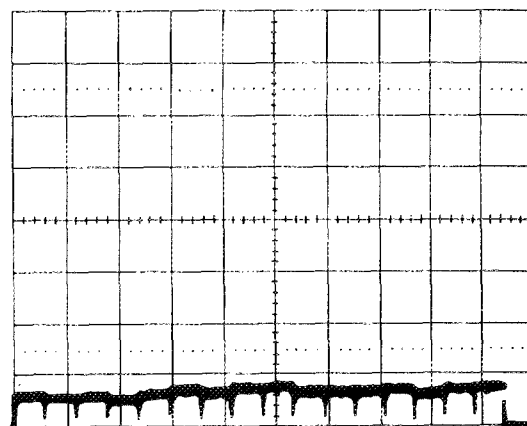


FIG. 4. Upper trace: Temporal behavior of the electric signal at the output of one of the preamplifiers. Lower trace: Output from the processing unit (horizontal scale: $50\ \mu\text{s}/\text{div}$, vertical scale: $100\ \text{mV}/\text{div}$).



a)



b)

FIG. 5. (a) Recording of a 0.8-mm -diam calibrating beam. (b) Output when only one of the partial beams of the interferometer is present.

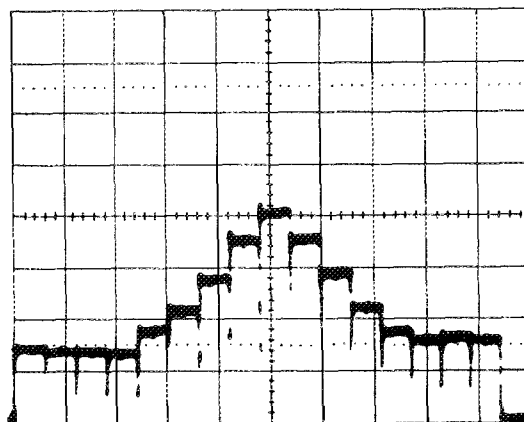


FIG. 6. Recording of an autocorrelation curve of a 248-nm 400-fs pulse.

IV. EXPERIMENTAL RESULTS

Figure 5(a) is the picture shown by the oscilloscope, when the input beam is apertured by a 0.8-mm-diam alignment pinhole, and both beams are adjusted to be in spatial overlap in the middle of the detection zone. This procedure could also be used to check the spatial resolution of the TPI detector, which was practically one channel in our case as can be seen in Fig. 5(a). With the standard field-electrode separation ($d = 2.5$ mm), the same resolution was obtained even for smaller electrode potentials than the $U = -250$ V normally used. Somewhat stronger dependence of the resolution was observed on the height of the probe beam. The resolution decreased when the probe beam was moved towards the field electrode. This effect was especially enhanced with large field-electrode separations. For $d = 5$ mm and for a probe beam just hitting the field electrode, a significantly decreased resolution is found and the relative amplitude of the neighboring channels reached 0.5. The electrode separation of $d = 2.5$ mm was found to minimize this effect, and to provide nearly one channel resolution.

Figure 5(b) is the recording of the square of intensity distribution of the beam, made by sending the whole beam coming from one arm of the autocorrelator through the TPI cell, while the other arm was blocked. This already indicates the main drawback of this method: The picture obtained always shows the square of the intensity distribution of the beam, which can significantly influence the shape of the autocorrelation function. In spite of the fact that the spatial intensity distribution of excimer lasers across the beam is expected to be described by a flat-topped function, for high brightness amplifiers, this function is found to be modulated by "hot spots" as already recognized in Refs. 19 and 20. This spatial modulation of the intensity appears as an uncontrolled modulation of the autocorrelation curves. Figure 5(b) is an optimized condition which was obtained by changing the relative position of the beam and the detector; in general, the intensity distribution seen by the detector was somewhat worse.

When both partial beams of the interferometer were sent through the TPI detector and spatial overlap of the two beams and the time window of the measurement were adjusted as described earlier, an autocorrelation function seen in Fig. 6 was obtained. For these measurements, the uncompressed pulses of our femtosecond-KrF-laser system were used,⁹ whose pulse duration was measured to be 450 fs, by an independent multiple shot measurement. The beam was attenuated; ~ 3 mJ pulse energy was enough to work with the TPI detector close to saturation. For these measurements the detector was filled with 100-Torr NO. The angle α was set to 5° . The whole 8×25 mm²-beam (with a longer axis lying horizontally) was sent through the interferometer. A 2.5×25 -mm² part of the beam was cut geometrically by the field electrode and by the lower plane electrode-array of the TPI detector. The time window of the single shot measurement corresponding to Fig. 6 was determined by two independent methods: by measuring the angle between the two beams [through Eq. (5)] and by introducing a controlled delay between the two beams, and monitoring the shift of the maximum of the autocorrelation curve on the screen. Both

methods resulted in $T = 2.9$ ps. From this value, $\tau = 650$ fs can be obtained for the autocorrelation width, corresponding to 420 fs pulse duration, with the same assumption (sech²) for the pulse shape as in the multiple shot measurement. The values obtained by the two measurements are the same within the estimated 15% accuracy of the single shot measurement.

V. OUTLOOK

This device can only be regarded as a nonoptimized first realization of a single shot TPI autocorrelator, using a transverse drift field, and segmented electrodes. The main drawback of this device is its limited accuracy, which is partly due to the limited number of collecting electrodes. This can be increased without any technical difficulty from 16 to 32, by proper extension of the multiplexing electronics. However, the degradation of accuracy due to the dependence of the shape of the autocorrelation curves on the intensity distribution of the beam is much more difficult to avoid. In the present setup, only a 2.5×1.25 -mm² part of the beam is seen by a single collecting electrode, and is spatially integrated (where 2.5 mm is the height of this part of the beam determined by the separation of the field electrode, 1.25 mm is the collection-electrode center-to-center spacing). Since in this case, it was already the inhomogeneity of the beam which determined the accuracy of the measurement, the increase of the number of electrodes, which necessarily decreases the spatially integrated area, is not expected to increase the real resolution and only results in more scattered points. To increase the height of the integrated area by increasing the field-electrode separation is no solution, because of the decreased spatial resolution. This means that it only makes sense to increase the number of the collection electrodes if the expected increased scatter of the measured points due to inhomogeneities of the beam can be prevented by some kind of integration of the intensity of the beam, which does not decrease the spatial resolution. The simplest way is to focus the beam in the vertical direction by a cylindrical lens, and get the whole beam through the detection area of the TPI detector. In this way, the vertical size of the integrated area of the beam can be increased from 2.5 mm to the original height of the beam (~ 8 mm); however, no increase of the horizontal size of the integrated area is expected. The latter problem might be solved by a modified detection geometry as shown in Fig. 7. Let us assume that the area determined by the collection electrodes is fully in the overlap of the two beams, and timing of the two beams is accomplished to have the maximum of the autocorrelation function in the middle of the electrode array. If we assume the two beams to have a hot spot in the position of the dashed line in the figure, it results in a maximum electric signal from the X electrode, if collection of the electrons is localized to plane A , as in our experiments by using a single wire as a field electrode. It is seen that if collection of the electrons is done at plane B , the position of the maxima of the electric signal due to the hot spots is changed, while the maximum of the autocorrelation function remains unchanged. Therefore, with the use of more separate field electrodes—which can be activated independently—one can have a spatial integration of the intensi-

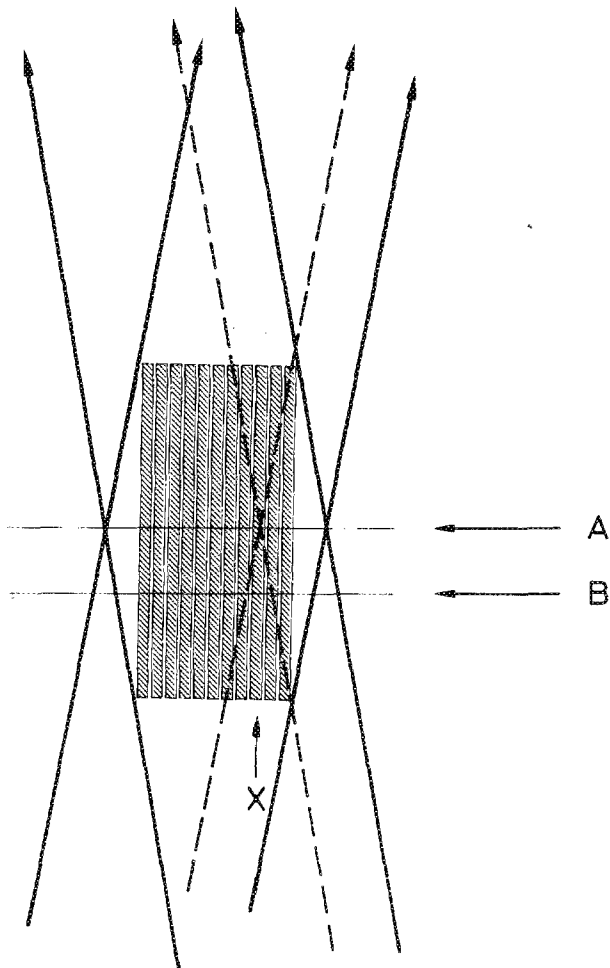


FIG. 7. Schematic of the proposed detection geometry.

ty across the beam, without any decrease of the temporal resolution. When using this method, the direction of the collecting electrodes must be set parallel to the bisector of the directions of propagation of the two beams, and the effect of the limited spatial coherence of the beam must be considered, limiting the maximum distance of these additional field electrodes from plane *A*.

The other possibility to improve the present system is the construction of the interferometer. In the present setup, while the relative timing of the two partial beams can simply be adjusted by a micrometer screw, complete spatial overlap of the beams at the detector, and their angle α determining the time window, can only be adjusted by complicated adjustment of two mirrors (M2, BS2 in Fig. 1) which also changes the timing of the two pulses. By the use of a special beam steering arrangement, it seems possible to continuously vary the angle between the two beams at the TPI detector, while leaving the temporal and spatial overlap unchanged. The development of such a device is in progress.

VI. DISCUSSION

In conclusion, we have demonstrated operation of a spatially resolving TPI detector, using transverse drift field and segmented electrodes. As a part of an autocorrelator, single-shot pulse duration measurements of ~ 500 fs pulses at 248 nm have been carried out. A modified Mach-Zehnder interferometer has been applied in these experiments, optimized for UV single-shot measurement. The method is very sensitive; only a fraction of the total output pulse energy of short pulse excimer systems is required for the measurements. This method is expected to have a very broad temporal and spectral range. By varying the angle between the two beams, the temporal range can be extended in principle from ~ 50 fs to ~ 50 ps. A spectral range from ~ 200 to ~ 500 nm seems possible to be covered by proper choice of the gas. The main drawback of this method is its limited accuracy, related partly to technical and physical reasons. Suggestions have been made to solve both problems.

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