# Single-shot autocorrelator for KrF subpicosecond pulses based on two-photon fluorescence of cadmium vapor at $\lambda = 508$ nm

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By excitation of cadmium vapor with a high-peak-power KrF excimer laser pulse, fluorescence of an atomic transition at $\lambda = 508$ nm is induced by a two-photon ionization process followed by fast recombination. The nonlinear response of the medium is used to develop a simple single-shot autocorrelator for subpicosecond KrF excimer laser pulses operating down to intensities of less than $10^9$W/cm.$^2$ We have measured 360-fs (FWHM) pulses at $\lambda = 248$ nm with a time resolution of 15 fs.

High-power subpicosecond UV lasers play an important role in multiphoton excitation and ionization processes.[1,2] These lasers are also proposed as pump sources for XUV and soft-x-ray lasers.[3] For KrF laser pulses at $\lambda = 248$ nm the commonly used technique for measurement of the pulse width is a nonlinear multiple-shot autocorrelation method based on multiphoton ionization of NO.[4] However, often the pulse width has to be controlled accurately in a single shot to eliminate instabilities. Autocorrelators for single-shot pulse-width measurements of femtosecond pulses at $\lambda = 248$ nm have been realized by two-photon fluorescence[5,6] of $Xe_2$,[7] three-photon fluorescence of XeF,[8,9] and recently by two-photon ionization of NO.[10] All these single-shot autocorrelators have some disadvantage for practical use. For example, the $Xe_2$ autocorrelator requires the detection of VUV fluorescence at 172 nm, and the one operating with NO requires the registration of charged particles with high spatial resolution. In addition, all correlators demand high pump laser energies ($E_p > 1$ mJ) and the proper alignment of colliding focused beams. Therefore they are not well suited for a permanent on-line control of KrF excimer laser pulses.

Recently we observed in cadmium vapor strong laser oscillation at $\lambda = 508$ nm by pumping the vapor with a subpicosecond KrF excimer laser system.[11] The excitation mechanism of the upper laser level was found to be a two-photon ionization process followed by fast recombination, as shown schematically in Fig. 1. Here we present investigations of a single-shot autocorrelator based on this process. Autocorrelation experiments have been performed with unfocused counterpropagating laser beams down to laser intensities of less than $10^9$ W/cm.$^2$

In the experiments a KrF excimer laser with a pulse width of 360 fs (FWHM) and an energy of 10 mJ (Refs. 11 and 12) was used to produce test pulses for the autocorrelator. The pulse width of this laser could be controlled by a multishot correlator based on two-photon ionization of NO.[4] In the experiments, the size of the original probe laser beam (1 cm × 3 cm) was cut by a pinhole to a diameter of ~1 cm and its energy was reduced to ~0.4 mJ. Subsequently the laser pulse was divided by a 50/50 beam splitter and directed antiparallel into a cadmium heat pipe with an active length of ~5 cm. The heat pipe was operated at a temperature of ~450°C (cadmium-vapor pressure 1 Torr) and a helium buffer-gas pressure of 20 Torr (Fig. 2). The temperature of the heat pipe was controlled within an accuracy of 5°C. The fluorescence of the cadmium vapor at a wavelength of 508 nm could be observed by a window transverse to the pump beam direction.

For the detection and documentation of the transversally emitted nonlinear fluorescence either a camera with a macro objective or a vidicon camera (B&M Spectronic SIT 500/OSA 500, resolution 25 $\mu$m) with a stereo microscope (Wild Leitz stereo microscope, magnification 20×, numerical aperture 0.1) as the objective was used. To block unwanted scattered light, an interference filter was inserted in front of the micro-

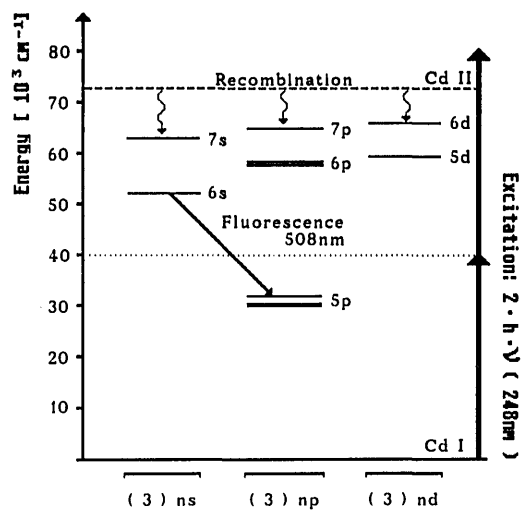


Fig. 1 Level scheme of atomic cadmium (part) with the relevant excitation–emission process used for the autocorrelation measurements.

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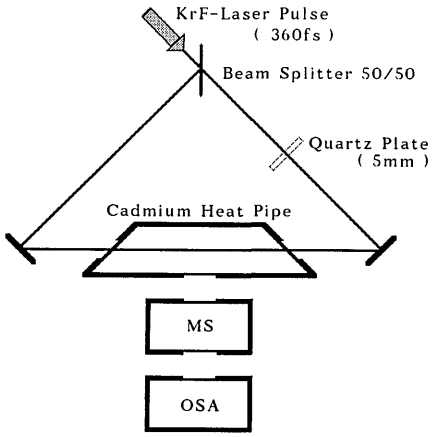


Fig. 2. Experimental arrangement of the single-shot autocorrelator. The quartz plate is used for calibration. MS, microscope for magnification of the fluorescence trace; OSA, vidicon camera system.

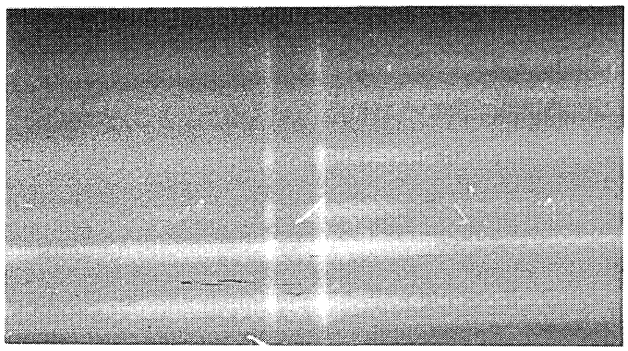

Fig. 3. Photograph of ten superimposed fluorescence traces generated by the counterpropagating laser pulses. The vertical lines (distance 1.25 mm) indicate the regions of the overlapping pulses with (right-hand line) and without use of the quartz plate.

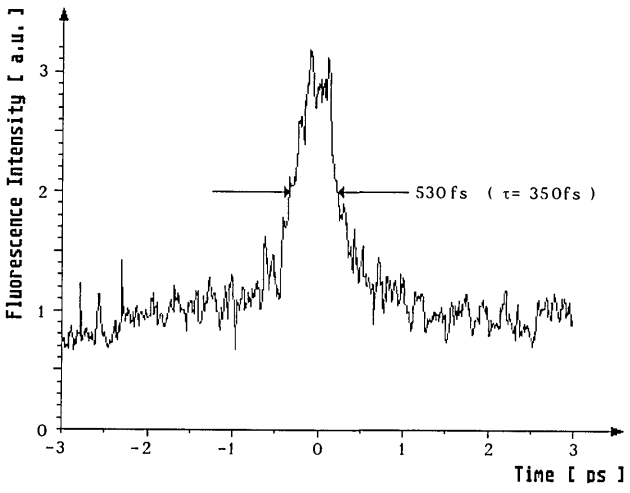


Fig. 4. Single-shot autocorrelation trace taken with the microscope–vidicon imaging and detection system.

scope or camera. The calibration of the autocorrelator was done by insertion of a plane-parallel quartz plate (thickness 5 mm) into one beam of the counterpropagating pulses.

A photograph of the transverse fluorescence (508 nm) of ten successive laser shots at a laser intensity within the heat pipe of less than $10^9$ W/cm$^2$ is shown in Fig. 3. The horizontal structure in the photograph is caused by inhomogeneities in the cross sections of the colliding beams. After five laser shots the quartz plate in one arm of the autocorrelator was removed. A densitometer trace analysis of this photo agrees well with a single-shot fluorescence intensity profile taken by the microscope–vidicon camera system (Fig. 4). The vertical-line structure of Fig. 3 can also be clearly observed through the microscope by eye; therefore with a calibrated superimposed scale a direct estimate of the pulse width is possible.

For a pulse form of a quadratic hyperbolic secant function the intensity profile of Fig. 4 corresponds to a pulse width of 350 fs (FWHM).[5,13] This value is in good agreement with that of the pulse width measured by the multiple-shot autocorrelator based on NO and indicates that no noticable broadening is caused by the optical imaging of the fluorescence trace within the heat pipe. Under the present experimental conditions the spatial resolution, determined by the resolution of the vidicon and the magnification and the numerical aperture of the microscope, is ~5 $\mu$m, corresponding to a time resolution of 15 fs.

In conclusion, a single-shot autocorrelator for subpicosecond pulses of KrF excimer laser radiation has been realized by two-photon ionization of cadmium vapor with subsequent atomic fluorescence at 508 nm. Because of the special nature of the nonlinear process, only relatively low intensities, less than $10^9$ W/cm,$^2$ are required for the pulse-width measurement. Therefore the autocorrelator can be operated with unfocused beams, is easy to align, and requires only a small amount of the total laser energy. This permits permanent on-line control of the pulse width. In the present heat-pipe system the distance from the laser beams to the observation window is relatively large (10 cm), making the imaging difficult and limiting resolution. With a more compact heat-pipe design or even with a sealed-off quartz tube this distance may be strongly reduced. In this way and by use of an adequate imaging system in combination with a diode stripe for detection, it should be possible to develop a compact, simple, and low-cost single-shot autocorrelator for KrF laser pulses with a time resolution below 5 fs.

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