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# High harmonic generation at 1 kHz repetition rate with a pulsed valve

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**Abstract.** High harmonic generation at 1 kHz repetition rate is reported. A piezoelectric pulsed valve together with a femtosecond Ti:Sapphire laser both operating at 1 kHz are employed to generate high harmonic radiation in xenon and argon. The characteristics of the valve have been analyzed by using a fast ion gauge and a time-of-flight mass-spectrometer.

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With advances in laser technology the generation of high harmonics of intense laser radiation has become a rapidly evolving area of research. One of the most spectacular examples is the generation of wavelengths in the water window (2.3–4.4 nm) [1–3] and of pulse durations close to the attosecond ($10^{-18}$ s) time domain [4].

The generation process of high harmonics is by itself an interesting example of the behaviour of atoms and molecules in intense laser fields. However, most of the current interest in high harmonic generation lies in the realization of ultrashort sources in the vacuum–ultraviolet (VUV) and extreme-ultraviolet (XUV) spectral regions. High temporal and spatial coherence [5, 6] – unique features of high harmonic generation – make it feasible to use harmonic pulses for time-resolved experiments in these spectral regions. Only recently, a cross-correlation of a high harmonic pulse with the fundamental laser pulse [4] showed the pulse duration of harmonics to be significantly shorter than the fundamental. Here a pulse duration below 2 fs was measured. This illustrates the potential of high harmonics for generating pulses in the attosecond time domain. The conversion efficiency of high harmonic generation has been improved up to $10^{-5}$ by proper phase matching [7, 8] and pulse energies in a single harmonic of up to 0.2 nJ have been reported [7].

Both for the study of the fundamental aspects of high harmonic generation as well as for the experimental application of high harmonic radiation, an increase in the average photon flux is desired. Nowadays, typical table-top chirped pulse amplification (CPA) laser systems with a 1 kHz repetition rate are used in many laboratories working in the field of high harmonic generation. Clearly, it is highly desirable to generate high harmonics at the full repetition rate of the laser system used. High harmonic generation at the kHz-repetition rate has been achieved by the use of static gas cells [9–12] or hollow fibers [13–15]. It has been shown [8, 15] that under optimized conditions the efficiency of high harmonic generation is approximately equal in both geometries. The drawback of static gas cells is absorption of the VUV-radiation emerging from the gas and the high gas load in the vacuum system. This makes the use of pulsed gas jets desirable. However, with pulsed gas jets, the repetition rate of harmonic sources was limited by the pulsed valve to several 100 Hz. In [16] harmonic generation from a kHz-repetition rate Ti:sapphire laser in a pulsed gas jet was reported. In this case, the only limitation on the repetition rate of the VUV-source was the valve, which was only capable of running at a maximum repetition rate of 750 Hz. Recently photoelectron spectroscopy with high harmonic radiation at 1 kHz was reported by Nugent-Glandorf et al. [28].

The ideal nozzle valve for high repetition rate harmonic generation would have the following characteristic design specifications: it should produce short and intense gas pulses at a repetition rate of 1 kHz or above. Furthermore, a high shot-to-shot stability with respect to the intensity and the temporal characteristics of the gas pulses is required.

In this paper we describe the development of a piezoelectric pulsed valve applied to the generation of high harmonics at a 1 kHz repetition rate.

## 1 Experimental setup

For the experiments presented in this paper, we used a commercial all-solid-state Ti:sapphire laser system from Spectra-Physics. Femtosecond laser pulses from an oscillator are temporally stretched, regeneratively amplified at a 1 kHz repetition rate and finally recompressed to a 80 fs pulse duration. The pulses have an energy of 900 μJ at a center wavelength of

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800 nm. The laser is focused by a 300-mm-focal-length lens into the particle beam, very close to the exit of the nozzle of a piezoelectric valve. We estimate the focused laser intensity to be $2 \times 10^{14}$ W/cm$^2$. This estimate is based on measurements of the ion yield intensity dependence of different noble gases. The valve is mounted on two translation stages for $x$- and $y$-adjustment and placed inside a vacuum chamber. This vacuum chamber is pumped by a 1000-l turbomolecular pump to obtain a pressure in the chamber of $\sim 10^{-4}$ mbar during the operation of the valve. The radiation emerging from the interaction of the laser pulses with the gas jet was analyzed using two different monochromators. For the lower-order harmonics (H3–H17, $\lambda$ between 300 nm and $\sim 45$ nm) we used a home-built 0.5-m Seya-Namioka monochromator with a 1200 grooves/mm platinum-coated spherical grating blazed for 70 nm. For the higher-order harmonics above H13 we used a commercial grazing incidence monochromator (Jobin-Yvon, LHT30) with a palladium-coated, 550 grooves/mm toroidal grating (wavelength range 150–15 nm).
In both cases the VUV-radiation is converted into fluorescence in the visible wavelength range by using Na-salicylate ($C_7H_5NaO_3$) as a scintillator material [17]. This scintillator was attached to a BG40-filter (Schott) which served both as an exit window of the vacuum system and to block background radiation from the scattered fundamental at 800 nm. A photomultiplier tube (Hamamatsu 1P28) was placed behind this window to detect the fluorescence from the harmonic photons. Data were collected using a box-car amplifier and stored in a computer. An overview of the experimental setup is given in Fig. 1. For gas pulse analysis a fast ionization gauge (FIG) [18] was also placed in the chamber. Furthermore, a time-of-flight mass spectrometer was mounted in the chamber to analyze the ions produced by the intense laser–matter interaction.

The piezoelectric pulsed valve was built following the design of Proch and Trickl [19]. A schematic view of the valve is shown in Fig. 2. The centerpiece of this valve is a piezo disk-translator (Physik-Instrumente P-286.23). In the center of this disk translator is a hole, where a plunger is mounted in an adjustable fashion. Actuation of the piezo translator by high-voltage pulses flexes the disk and moves the plunger up and down, resulting in the opening and closing of the nozzle. A vacuum seal is achieved by the use of a viton ring at the end of the plunger. This viton ring serves at the same

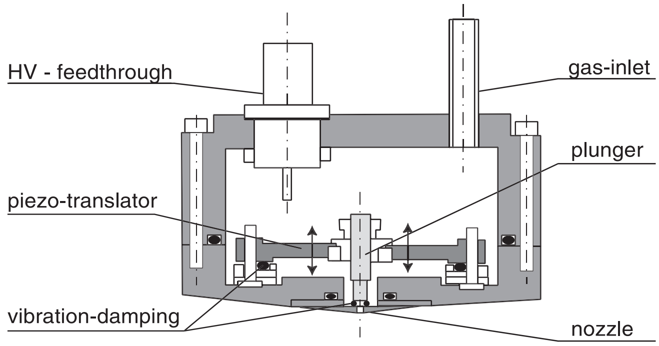


**Fig. 2.** Schematic view of the valve

time for vibration damping. It turned out to be necessary to attach a water cooling device to the valve. At high repetition rates, there is a substantial thermal load to the piezo, by which the shot-to-shot stability deteriorates unless cooling is implemented.

In order to synchronize the pulsed valve with the laser, trigger pulses from the laser were appropriately delayed in time with a Stanford Research delay generator. Pulses from this delay generator were then fed into a high-voltage pulse-generator. The generated high-voltage pulses were directly applied to the piezo-ceramics inside the valve, to open and close the nozzle.

## 2 Performance of the valve

A first test of the performance of the valve was achieved by observing the pulse train from the valve using the fast ion gauge (FIG). In Fig. 3 pulse trains from the valve operated with different stagnation pressures are shown. The pulses are separated in time by 1 ms, indicating the 1 kHz repetition rate. One can see stable operation ($\pm 1\%$) for stagnation pressures of up to 3 bar. At the higher stagnation pressures of 4 bar and 6 bar the shot-to-shot stability deteriorates to $\pm 3\%$.

Another test of the valve performance was done with the time-of-flight (TOF) spectrometer. We recorded the ions generated by the interaction of the laser pulses with the gas pulse and changed the delay between the high voltage pulses used to trigger the valve and the laser pulse. This enabled us to

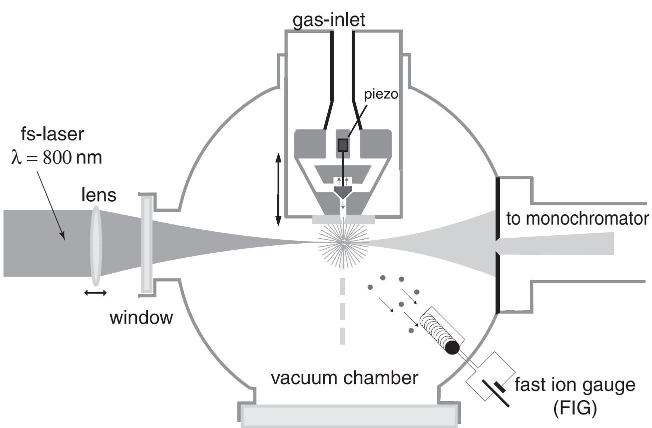


**Fig. 1.** Overview of the experimental setup

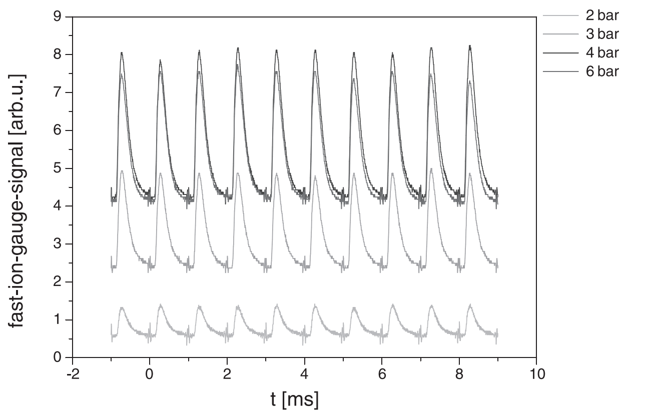


**Fig. 3.** Pulse trains from the valve for different stagnation pressures, measured with the fast ion gauge

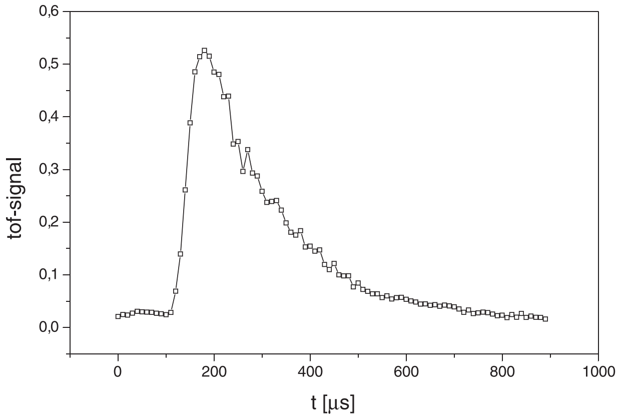


**Fig. 4.** Temporal profile of the gas pulses from the valve. The ion signal was recorded with the time-of-flight spectrometer while the delay between the gas and laser pulses was varied

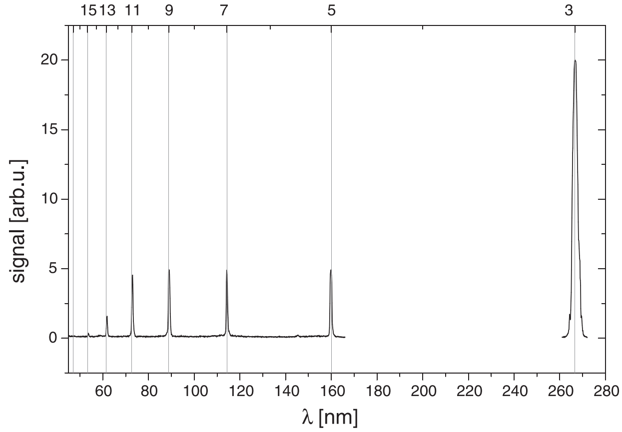


**Fig. 5.** High harmonic generation in a xenon gas jet at a 1 kHz repetition rate

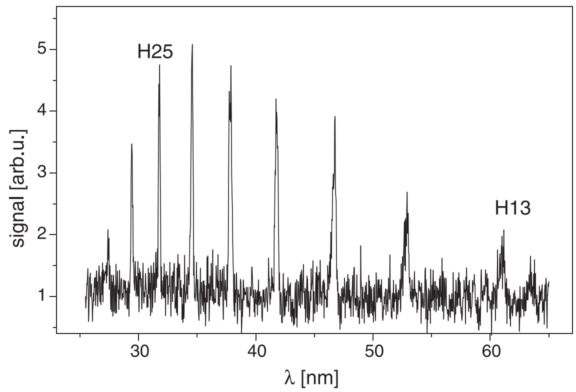


**Fig. 6.** High harmonic generation in an argon gas jet at a 1 kHz repetition rate

measure the temporal profile of the gas pulse, as shown in Fig. 4. To avoid saturation and space charge effects the laser intensity was attenuated appropriately. We used trigger pulses of 50 μs duration to generate the high-voltage pulses. The actual gas pulse, however, does not follow the trigger pulse profile closely, but exhibits a steep rise and a slow decay. From the area under the gas pulse we can estimate the reduction in gas load to the vacuum system as compared to a continuous operation of the valve. It should be noted that a similar way to measure the temporal profile of the gas pulse was reported in [20]. In this article the third harmonic of the fundamental laser pulse was recorded while changing the delay between the laser and gas pulses.

## 3 High harmonic generation

Gas pulses from the valve and femtosecond laser pulses at 800 nm have been used to generate high harmonics. The process of high harmonic generation is now well understood in the framework of the so-called three-step model [21]. In the first step an atom is ionized by field- or tunnel-ionization. In the second step this electron is accelerated in the strong electric field of the laser and eventually driven back to the ionic core with which it can recombine in the third step. The energy emitted in a harmonic photon during this recombination is the energy which the electron gained by the acceleration in the laser field plus the ionization potential of the atom. Harmonic spectra are typically of the following shape: a strong decrease of the intensity for the low-order harmonics followed by a plateau consisting of harmonics with nearly equal intensity and then a steep decrease at the highest harmonics, the so-called cut-off region. The photon energy $E_c$ at which this cut-off occurs is roughly given by the so-called cut-off law $E_c = I_p + 3.2U_p$ [21] with the ionization potential $I_p$ of the specific atom used and the ponderomotive potential $U_p[\mathrm{eV}] = (9.33 \times 10^{-14})\ (I\ [\mathrm{W/cm^2}])\ (\lambda^2\ [\mu\mathrm{m}])$ corresponding to the energy of the free electron in the oscillating laser field.

Typical high harmonic spectra obtained by focusing the fs-laser pulses into the gas jet produced by the valve described here are shown in Figs. 5 and 6. In both cases raw data is shown without any correction for the detection efficiency of the specific monochromator.

In Fig. 5 we used xenon as a gas and detected the harmonics with the home-built Seya-Namioka monochromator. One can clearly distinguish the single harmonics from $3\omega$ (H3) to $15\omega$ (H15). For the lowest orders one can see the steep decrease in the intensity and the plateau from H5 to H11. The highest observed harmonic is H15. Since the transmission efficiency of this monochromator allows us to observe higher harmonics, we have to assume that we really observe the cut-off in this experiment. That result agrees well with the spectra reported in [22–24], where the highest harmonic observed in xenon was also H15. The relatively low harmonic order at which the cut-off occurs is given by tunneling and barrier-suppressed ionization of the neutral xenon atoms already in the rising edge of the laser pulse [25].

In Fig. 6 argon was used to generate harmonics and the grazing incidence monochromator to detect these. We see harmonics from H13 to H29. With H29 as the cut-off we estimate the corresponding laser intensity from the cut-off law to be $1.5 \times 10^{14}\ \mathrm{W/cm^2}$. This measurement demonstrates the capability of our setup to generate harmonic radiation even at wavelengths around 30 nm.

## 4 Conclusion

In the work presented we have used a piezoelectric pulsed valve to generate gas pulses at a repetition rate of 1 kHz. The gas pulses from the valve were analyzed by using a fast ion gauge and the temporal profile of the gas pulse was measured with a time-of-flight spectrometer. A strong reduction in the gas load as compared to continuous gas jets could be achieved with this pulsed valve. Stable operation of the valve has been accomplished and the valve has now been used for over half a year without the need for replacement of the piezotranslator. The valve's repetition rate of 1 kHz is matched to that of state-of-the-art femtosecond laser systems. Therefore, this valve allows us to study intense laser–matter interactions at a 1 kHz repetition rate. As an example we have demonstrated the capability of this valve by generating high harmonics in xenon and argon gas jets. We believe that with this setup some interesting experiments are now made possible, which would be very difficult to perform with, for instance, a 10 Hz repetition rate. One of these experiments is the feedback-controlled optimization of high harmonic generation, already demonstrated in hollow fibers [26], using pulse-shaping technology in combination with an evolutionary algorithm [27]. The combination of this valve with the time-of-flight spectrometer will also allow for the simultaneous investigation of other high-field effects such as non-sequential double ionization or above-threshold ionization.

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