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LETTER TO THE EDITOR

Two-colour time-resolved spectroscopy of helium using high-order harmonics

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Abstract. The radiative lifetime of the $1s2p\ ^1P$ state of helium is measured in a two-colour ionization experiment with a 5% accuracy. The state is excited by the 13th harmonic of a tunable 80 ps laser and ionized by a synchronous ultraviolet laser, with a variable time delay. This experiment demonstrates that the high harmonics generated in a jet of rare gas exposed to an intense laser field provide a tunable short-pulse XUV source ideally suited for pump/probe type of studies and, in particular, short lifetime measurements.

Pump/probe techniques using visible or ultraviolet lasers have become an essential tool in atomic, molecular and solid-state physics. One of the most spectacular applications is the so-called 'femtochemistry', pioneered by Zewail and co-workers (1988), where femtosecond laser pulses are used to investigate a photodissociation process in real time. The extension of these techniques to processes where one of the photons, the pump or the probe is in the extreme ultraviolet (XUV) region presents several difficulties. Traditional laser-based techniques, such as multi-step excitation (Baumert *et al* 1991) or frequency mixing (Cromwell *et al* 1992, Eikema *et al* 1993) are limited in spectral range (typically above ~ 60 nm) and/or time resolution. Synchrotron radiation light sources (Nenner *et al* 1994), continuously tunable in the XUV, can be synchronized with pulsed lasers, as shown recently by Lacoursihre *et al* (1994), but not easily, and moreover are limited in temporal resolution to a few tens of picoseconds at best.

A domain where pump/probe time-resolved methods with the pump in the XUV range can be very useful in atomic spectroscopy for the accurate measurements of the radiative lifetimes of high-lying selectively-excited atomic or ionic states. Time-resolved fluorescence techniques (Svanberg 1991, Zerne *et al* 1994) cannot be employed easily in the XUV range due to the poor collection efficiency. However, by ionizing the excited atoms or ions with a second short-pulse excitation delayed in time and detecting the ions produced, a high collection efficiency as well as a high temporal resolution can be obtained.

In the present work, we show that the high harmonics emitted when an intense laser is focused into a rare gas can be an ideal XUV source for pump/probe types of experiment, and in particular for the measurement of short (subnanosecond) lifetimes. High-order harmonics

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generated in a jet of rare gas have been studied for several years and the physics of these highly nonlinear processes is now reasonably well understood (Krause *et al* 1992, Corkum 1993, L'Huillier *et al* 1993). The harmonic spectra are characterized by a plateau of nearly constant intensities and can extend in energy up to 170 eV (Macklin *et al* 1993, L'Huillier and Balcou 1993). Experiments demonstrating the usefulness of this new source for applications in solid-state physics (Haight and Peale 1993, 1994) or in photoionization spectroscopy (Balcou *et al* 1994) have recently been performed. The harmonic radiation is collimated and spatially coherent. Its spectral and temporal characteristics follow those of the pump laser. For example, a tunable picosecond laser generates tunable harmonic radiation of relatively narrow-bandwidth, allowing selective excitation of atomic states. The short pulse duration of this radiation enables one to perform time-resolved experiments with a high temporal resolution. Moreover, the harmonic pulses can easily be synchronized with another short-pulse laser the frequency of which can be chosen in order to ionize the excited atoms in an efficient as well as selective process. To illustrate and demonstrate these ideas, we have measured the lifetime of the $1s2p^1P$ state of helium, using the 13th harmonic of a tunable 80 ps laser generated in a jet of krypton. The harmonic radiation excited resonantly the $1s2p$ state, which was subsequently ionized by ultraviolet laser light. The lifetime was measured with an accuracy comparable with previous measurements performed using other methods (self-absorption or electron scattering).

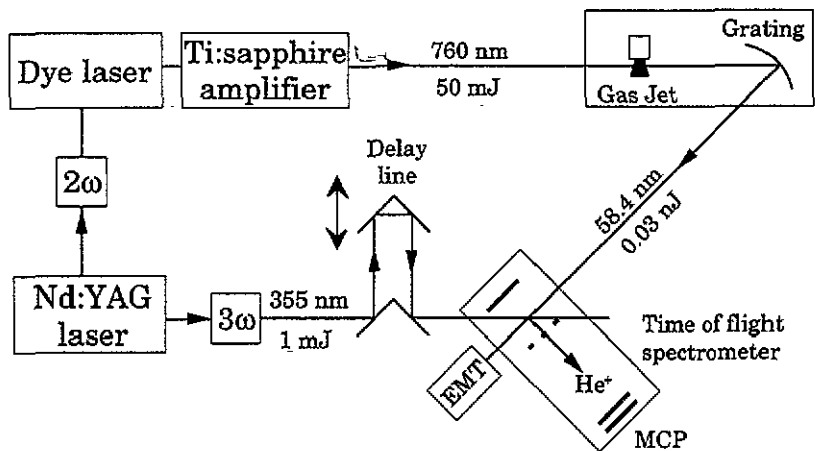


Figure 1. Experimental setup.

The experimental setup is shown schematically in figure 1. A distributed feedback dye laser oscillator (Shank *et al* 1971, Bor and Schaefer 1983, Schade *et al* 1990) generates 80 ps pulses tunable between 715 and 900 nm. These pulses are amplified in two dye cells and in a titanium-sapphire crystal up to an energy of 50 mJ (Larsson 1994). The laser bandwidth is 0.08 nm and the repetition rate is 10 Hz. Harmonics are generated in a pulsed jet of rare gas and separated by a 1200 grooves/mm normal-incidence spherical grating with a 1 m radius of curvature. The odd harmonics of the fundamental and those of the second harmonic generated in a doubling crystal together provide a continuously-tunable narrow-band XUV light source from 200 to 35 nm (21st harmonic) as described by Larsson (1994). By using lasers with shorter pulse duration this limit can be extended to much shorter wavelengths (Macklin *et al* 1993, L'Huillier and Balcou 1993), to the detriment, however, of the harmonics bandwidth.

In the present experiment we tuned the laser wavelength around 760 nm and selected the 13th harmonic (58 nm, 21 eV) to resonantly excite the $1s2p^1P$ state of He. The excited atoms were subsequently ionized by the third harmonic (355 nm, 3.5 eV) of a fraction of the 80 ps Nd:YAG laser the second harmonic of which is used to pump the dye laser (see figure 1). This photon energy of the probe laser was chosen in order to barely reach the threshold, thus optimizing the ionization cross section. (For measurements of the radiative lifetimes of higher-lying states the ionization of other, lower states in the cascade decay, is prevented by choosing the probe energy to be just above the ionization limit.) The two beams were crossed at 45° inside a time-of-flight spectrometer (Balcou *et al* 1994). Both the 13th harmonic and the 355 nm light were approximately focused at the crossing point (over a diameter estimated to be ~ 0.5 –1 mm). A variable delay line placed in the beam path of the third harmonic allowed us to adjust the relative time between the two light pulses. The generated ions were separated in mass in the time-of-flight tube and detected by a microchannel plate (MCP).

Ideally, the bandwidth of the harmonic light should be comparable with the Doppler broadening of the $1s^2 \rightarrow 1s2p$ transition (3×10^{-4} nm), in order to optimize the number of 'useful' photons. On the other hand, the harmonic pulse duration should be short enough (~ 50 ps) to enable lifetime measurements with good accuracy. The spectral profile of the 13th harmonic, obtained by tuning the laser wavelength and by recording the ion signal, is shown in figure 2. The width at half maximum is about 10^{-2} nm ($\Delta\lambda/\lambda \simeq 10^{-4}$). The temporal width of the harmonics, smaller than that of the pump, can be estimated to be approximately the perturbative limit, i.e. 20 ps (Faldon *et al* 1992). In the present experiment, the temporal resolution is limited by the pulse duration of the probe ~ 50 ps. Figure 3 shows in a logarithmic scale the number of He^+ ions in arbitrary units as a function

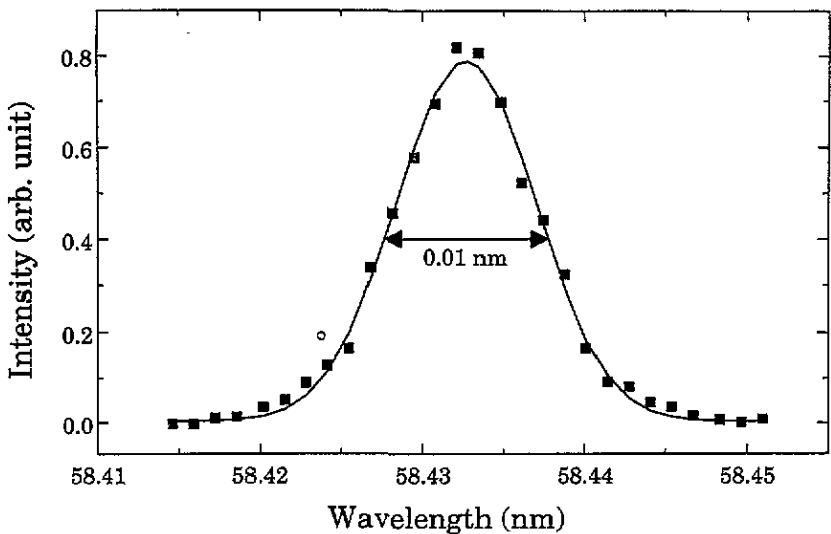


Figure 2. Spectral profile of the 13th harmonic beam. The full curve is a fit to the data.

of the delay time between the pump and the probe. Each point is an average of 200 shots, selected so that the harmonic intensity, recorded by an electron multiplier (EMT) (see figure 1), is within a certain window. The pressure for this measurement was 7×10^{-6} mbar. The number of ions detected per second varies from 10 to 1000, which makes the acquisition time for a full decay curve small, about half an hour, thus minimizing systematic errors.

Note also that the signal/noise ratio is quite high, owing to the absence of other light, second orders, etc, which could be an additional cause of the ionization of the gas. The logarithm of the signal varies linearly with the delay time over more than four lifetimes (2 ns), allowing the lifetime to be determined with a good accuracy. From the estimated

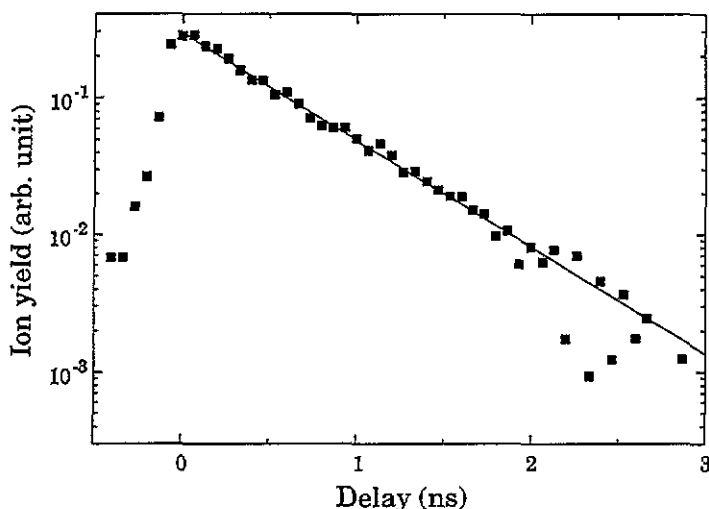


Figure 3. Number of ions as a function of the delay time. A linear fit of the data obtained at positive delay times, indicated by the full line, allows us to determine the lifetime.

number of ions produced at each laser shot we can deduce the number of harmonic photons in the interaction chamber. When the probe and pump beams were spatially overlapped, the number of ions detected was of the order of 100 per shot. The collection efficiency is 100% and the probe photon flux was sufficiently high to ionize all the excited atoms in the interaction region. The absorption cross section at the maximum of the Doppler-broadened spectral line is $7 \times 10^{-14} \text{ cm}^2$. Assuming an interaction length of about $500 \mu\text{m}$, for an atomic density $\rho = 1.4 \times 10^{11} \text{ cm}^{-3}$ ($7 \times 10^{-6} \text{ mbar}$), the number of photons used for exciting the $1s^2 \rightarrow 1s2p$ transition was therefore equal to 2×10^5 . By multiplying this number by the ratio of the harmonic linewidth over the Doppler width, we can deduce an estimation of the total number of harmonic photons in the interaction chamber: 10^7 per laser pulse, in relatively good agreement with another direct estimate (Larsson 1994).

In figure 4, we present the results of several lifetime measurements performed at various static pressures. The aim of the repeated measurements was twofold: to test the reproducibility of the data and to investigate the influence of the atomic density. Further data were also obtained at even higher pressures ($\sim 10^{-3} \text{ mbar}$), using a pulsed helium gas jet. No effect due to, for example, multiple scattering or collision processes, was observed over the whole range investigated. This could be due to the small volume intercepted by the probe beam (estimated to $\sim 10 \text{ mm}^3$). The measurement was possible even at quite low pressures: $2 \times 10^{-7} \text{ mbar}$ ($4 \times 10^9 \text{ atoms/cm}^3$). The line indicates the mean value of this ensemble of measurements. The deduced lifetime is $0.57 \pm 0.03 \text{ ns}$.

The branching ratio between the $2p \rightarrow 1s$ and $2p \rightarrow 2s$ decays is 10^4 (Radzig and Smirnov 1985). The lifetime of the $1s2p^1P$ state can therefore be converted into the $1s \rightarrow 2p$ oscillator strength and compared with previous measurements and calculations. The oscillator strength for the $1s^2 \rightarrow 1s2p^1P$ transition in helium has been studied experimentally by various methods from self-absorption (Ligtenberg *et al* 1994, Tsurubuchi *et al* 1989),

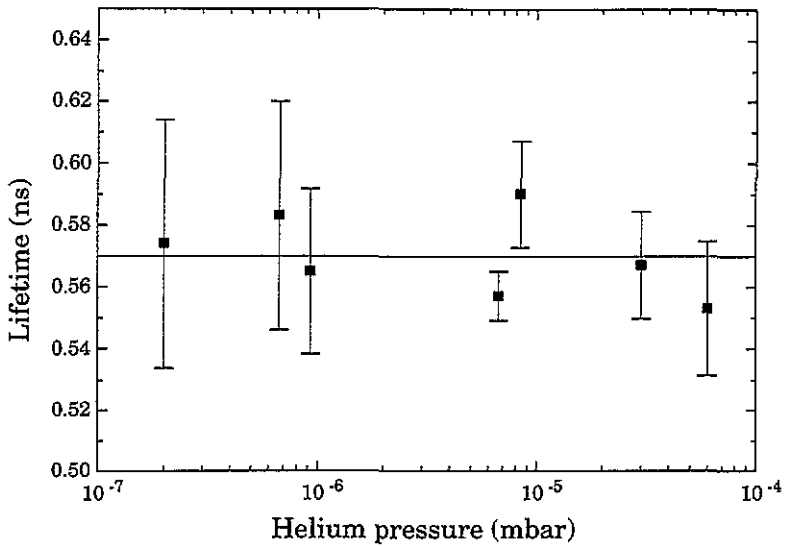


Figure 4. Lifetime measurements at different atomic pressures. The straight line is the mean value. The statistical error bars are indicated.

lifetime measurements using level crossing (Burger and Lurio 1971) or beam-foil (Martinson and Bickel 1969), and electron scattering (Chan *et al* 1991). Our result is in good agreement with these previous experimental data, with about the same level of accuracy. The theoretical calculation recognized as being most accurate in the literature is that of Schiff *et al* (1971), using a variational method. Table 1 summarizes some of the most recent measurements, together with the theoretical value.

Table 1. Recent measurements of the $1s \rightarrow 2p$ absorption oscillator strength. We also indicate the theoretical calculation of Schiff *et al*.

Ligtenberg <i>et al</i>	Self-absorption	0.2683 ± 0.0075
Tsurubuchi <i>et al</i>	Self-absorption	0.273 ± 0.008
Burger and Lurio	Lifetime: level crossing	0.275 ± 0.007
Martinson and Bickel	Lifetime: beam-foil	0.27 ± 0.01
Chan <i>et al</i>	Electron scattering	0.280 ± 0.014
Present	Lifetime: two-colour	0.269 ± 0.015
Schiff <i>et al</i>	Theory	0.2762 ± 0.0001

In conclusion we have demonstrated that the high harmonics produced during the interaction of an intense laser beam with a jet of rare gas can be used for two-colour pump-probe experiments. It enables cascade-free studies of radiative properties of atomic and ionic systems in the XUV spectral range. It is also possible to go to much shorter wavelength (less than 10 nm) and shorter pulse duration (~ 100 fs or less) than in the present experiment, by using a pump laser with a shorter pulse duration. This could be useful for measurements of very short radiative processes.

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