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Synchronization of a mode-locked Ar^+ laser with the synchrotron radiation: application to the two-photon ionization (VUV + visible) of helium

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Abstract : a two-color experiment combining and synchronizing a mode-locked Ar^+ laser and the synchrotron radiation has been set-up allowing to perform time-resolved pump and probe experiment on the ns time-scale. As a test experiment, the time-resolved (1+1) photoionization of helium via the 1s3p state is presented.

1.INTRODUCTION

Laser and synchrotron radiation (SR) are photon sources with very specific properties, monochromaticity and high photon flux for the former, and a very broad tunability and high photon energy for the latter. To take simultaneous advantage of these features, it is especially interesting to combine the two photon sources to perform pump and probe experiments in which a first photon prepares an excited state of the studied system or a transient species which is analyzed by a second photon via an excitation into either a discrete or an electronic or nuclear continuous state [1,2].

According to this scheme several experiments have been recently performed combining a visible cw laser to the SR to study the photoionization of excited [3] and aligned atoms[4] and of laser induced dissociation fragments [5]. In these experiments, the lifetime of the intermediate state in the interaction region is of the same order of magnitude as the SR interpulse period (T_{SR}=120 ns in the case of Super-ACO) so that the use of cw lasers is satisfactory. At the opposite, if the intermediate state has a much shorter lifetime, it is necessary to concentrate in time the two photons, *i.e.* to use a pulsed laser synchronous with the SR, with comparable repetition rates, in order to obtain an efficient pumping process. Note that MHz repetition rates are very well suited when space charging, false coincidences or optical damage have to be avoided. In addition, by scanning the delay between the two pulses of light, one can have access to the lifetime of the excited state and perform time-resolved spectroscopy, in which the dynamics of the excited state is sampled in time by the delayed probe photon while keeping a continuous detection scheme, as in a stroboscopic experiment. Therefore, the temporal resolution of such an experiment is limited by the temporal cross correlation between the two pulses and not by the speed of the detector [6]. Moreover, the high peak power of a pulsed laser allows the production of non-linear processes such as multiphoton absorption.

2.THE EXPERIMENT

Already done in the solid-state [7] and liquid phase [8] but never yet in the gas phase where the target density is very low, a time-resolved laser+synchrotron experiment is presented here on a simple test system : helium. The experimental set-up is described in detail elsewhere [9]. Briefly, we have synchronized a mode-locked Ar^+ laser at 514 nm

with the SR in the two bunches mode of Super-ACO (pulsed at 8.32 MHz) by triggering the radiofrequency of the mode-locker with the one of Super-ACO, in order to induce a resonant two-photon ionization on an effusive jet of helium via the 1s3p (^{1}P) state according to the following scheme :

$$He(1s^2) + h\nu_{SR}\ (23.086\ eV) \rightarrow He^*(1s3p) + h\nu_{laser}\ (514\ nm) \rightarrow He^+ + e^-\ (0.9\ eV)$$

An electronic dephaser allows to tune the laser-to-SR delay and thus to follow the population of the excited state as a function of time. In addition, the experimental set-up is composed of a time-of-flight electron spectrometer optimized for the collection of 0.9 eV electrons corresponding to the studied process, a synchronous detection system necessary to enhance the poor signal-to-noise ratio, and a fast photodiode located *in situ* to characterize very precisely the correct spatio-temporal overlap of the two beams.

3.RESULTS AND CONCLUSION

The recorded electron signal representing the evolution of the 1s3p state population versus the laser-to-SR delay is presented in figure 1. This signal is well described by a simulation in which the lifetime of the excited state is set to the known value of 1.7 ns, and a temporal instrumental cross-correlation, mostly due to the width of the SR, of about 1.1 ns is fitted.

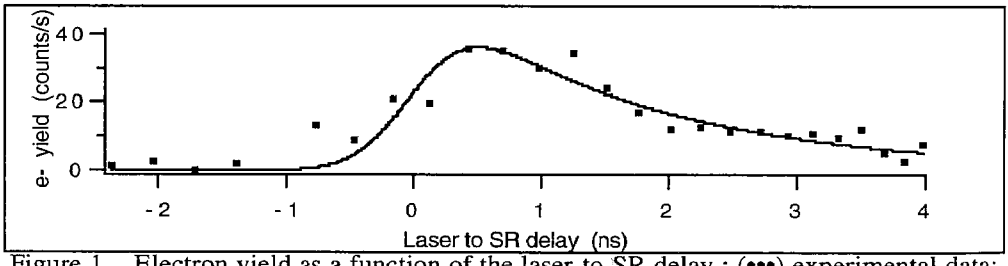

Figure 1 . Electron yield as a function of the laser-to-SR delay : (•••) experimental data; (---) simulation obtained with a 1s3p lifetime of 1.7 ns and a temporal resolution of 1.1 ns.

Considering these first results, the experiment seems to be very well adapted to the study of dynamics and time-resolved spectroscopy on the nanosecond time-scale. Its capabilities should be extended by the use of a dye laser synchronously pumped by the Ar^+ laser that would allow for instance the determination of excited states photoionization cross sections.

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