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M. Hemici, S. Mottin, M. Bon, J.-Y. Roncin, P. Laporte. Time resolved transient absorption probing of a laser plasma. Journal de Physique III, 1991, 1 (12), pp.2061-2065. 10.1051/jp3:1991251 . jpa-00248721

HAL Id: jpa-00248721 https://hal.science/jpa-00248721

Submitted on 4 Feb 2008

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Time resolved transient absorption probing of a laser plasma

M. Hemici (1), S. Mottin (2), M. Bon (1), J.-Y. Roncin (1) and P. Laporte (1)

(¹) Laboratoire Traitement du Signal et Instrumentation, CNRS URA 842, 23 rue P. Michelon, 42023 Saint-Etienne Cedex 2, France

(²) Ecole Nationale Supérieure des Mines, 158 Cours Fauriel, 42023 Saint-Etienne Cedex, France

(Received 6 May 1991, revised 4 July 1991, accepted 19 August 1991)

Résumé. — On décrit un dispositif expérimental qui permet l'étude de la dynamique de déclin d'un plasma, créé dans un gaz par un laser en impulsion, par spectroscopie d'absorption transitoire dans le visible, avec une résolution temporelle de 10 ns et une résolution spectrale de $0,3 \text{ cm}^{-1}$. On donne comme exemple l'absorption à partir des premiers niveaux excités 4s vers des niveaux 5p de l'argon. La dépendance temporelle est donnée pour la population du niveau $4s[3/2]_2$.

Abstract. — The described experimental setup allows the kinetic analysis of a pulsed-laser generated gas-plasma by transient visible absorption spectroscopy at a time resolution of 10 ns and a spectral resolution of 0.3 cm^{-1} . Absorption from the first-4s level manifold towards the 5p level manifold of the argon atom is reported in the spectral range around 420 nm. An example of time dependence is given for the $4s[3/2]_2$ level population.

Since the advent of lasers their ability to generate plasmas has been recognized and widely used and the physical properties of such plasmas extensively studied [1]. Quite a number of those works dealt with rare gas laser plasmas but very few have been devoted to spectroscopical properties of these plasmas. However during the last two decades dense rare gases have attracted considerable attention due to their potentiality as laser media [2]. In this respect a detailed knowledge of plasma kinetics is of great importance. The present report describes a method allowing the kinetic study of a gas plasma by transient visible absorption spectroscopy at high spectral and temporal resolution.

The experimental setup is sketched in figure 1. The pulsed-laser generated plasma, recently used as a vacuum ultraviolet source [3], is created by focusing the 1 064 nm radiation of a pulsed 585-20 Quantel Nd : YAG laser (YAG 1, 350 mJ) at the center of a 6-way gas cell by means of the spherical lens L1 of focal length 20 mm. The stainless steel gas cell, mounted with metallic Helicoflex gaskets from Cefilac, is evacuated with a turbopump and refilled before each run with a gas at a pressure typically in the atmospheric range (10^5 Pa). This is a very clean technique as the medium is excited far from the cell walls. The probe beam is obtained by irradiating a laser dye cell by part of the frequency tripled radiation at 355 nm of a



Fig. 1. — Sketch of the experimental setup. L2, L5: cylindrical lenses. L1, L3, L4: spherical lenses. W2, W3: wedged coated windows. OMA: Optical Multichannel Analyzer.

581-20 Quantel Nd : YAG laser (YAG 2). The two YAG lasers are run at 20 Hz and deliver pulses of 10 ns. The stainless steel dye cell is of the type described in [4]. It is known that the high resolution spectrum of a broad-band dye-laser exhibits bright and narrow hopping hot spots. It has been demonstrated in [5] that it is necessary to avoid any cavity if one wants a good spectral quality of the continuum. That is why the two wedged side windows W2 and W3 are tilted and have antireflection coating. Then the light coming out of the dye cell is just an amplified spontaneous emission that is focused a few cm beyond the plasma by means of the tilted spherical lens L3 of focal length 80 mm. The lens L4, of focal length 600 mm, sends quasi-parallel light towards the 2.5-m Ebert spectrometer manufactured by Sopra. The cylindrical lens L5 of focal length 90 mm concentrates the light onto the 30 μ m entrance slit. Such an optical arrangement leads to a negligible amount of light from the plasma entering the spectrometer as compared to the probe light.

In a first stage we can use a $9 \times 12 \text{ cm}^2$ photographic plate to get an overview of the spectrum. For intensity measurements we replace the plate by a 1452A-1461 EG & G optical multichannel analyzer (OMA) made of an array of 512 photodiodes 25 µm each so that a width of 12.7 mm is scanned for any setting of the grating. In the reported example of an argon plasma, which is probed around 430 nm, the grating was set in the 3rd order giving a plate factor of 0.06 nm/mm, i.e., $1.5 \times 10^{-3} \text{ nm/pixel}$.

A home made electronic monitor starts the Pockels cells of the two YAG lasers with a variable time delay. In addition the light from the plasma and that from the dye cell are viewed by two photodiodes, respectively 1 and 2, whose signals are sent to an oscilloscope so as to check the time delay between firing the plasma and starting the probe beam. The delay can be adjusted from 0 to 2 μ s with an accuracy of a few ns. It is well known that, for a null delay, the probe beam, going through the plasma, is considerably attenuated by the

background absorption [6-8] making eyeview optical adjustment straigthforward by looking at the shadow pattern similar to those described in [1]. Final adjustment onto the spectrometer slit by L5 is viewed on the video control of the OMA signal which is stored and processed in a Macintosh microcomputer. Data are handled with Macoma software and the resulting absorption curves come out of a laser printer. Previous transient absorption studies have been performed using somewhat related techniques. Nevertheless to the best of our knowledge, they were restricted to plasmas created by focusing a laser beam onto a solid target [9-11] and did not offer the wide capability of the present setup, that is to say synchronized lasers and high temporal and spectral resolutions.

Our experimental setup has been tested by filling the gas cell with argon of purity N60 at pressures in the range 10^5 Pa and the laser dye was stilben 420. It is observed that, when the time delay is increased from 0 onward, the background absorption decreases and gives way to discrete absorption lines corresponding to transitions starting from the first-4s level manifold of the argon atom. Discrete absorption is the strongest for a delay in the 100 ns range, depending on pressure, and decreases afterwards. In figure 2 are displayed several transmission curves for argon at 10^5 Pa and with time delays from 0 to 70 ns. Lines A and B correspond to the transitions $4s[3/2]_1 \rightarrow 5p[1/2]_0$ and $4s[3/2]_2 \rightarrow 5p[5/2]_3$, respectively. The very direct characterization of these transient atomic excitations is fully consistent with recent selective vacuum ultraviolet photoexcitation studies of neutral rare gases which have shown that the first excited atomic levels act as intermediate steps in decay processes [12-14]. For line B we have plotted the amount of absorption in function of time delay. There results a typical rise and decay curve, displayed in figure 3, giving the time dependence of the absorption resulting from the $4s[3/2]_2$ excited atoms. The curve was fitted with the formula $\left[\exp\left(-t/\tau_1\right) - \exp\left(-t/\tau_2\right)\right]$ with a rise time $\tau_1 = 18.5$ ns and a decay time $\tau_2 = 550$ ns. It has to be pointed out that absorption occurs along a highly inhomogeneous medium in which expansion and thermalization takes place so that kinetic information can result only from modeling. Nevertheless the present setup may provide direct information about the decay scheme from observation of transiently populated levels and oscillator strength ratio of intervening transitions. Previous observations of the above mentioned transitions in absorption have been reported from experiments using a cell containing a rare gas excited by an electron beam and performed at smaller resolution [6-8]. The advantage of a laser plasma is to give a higher concentration of excited species so that the plasma still absorbs at a lower pressure.



Fig. 2. — Transmittance of the argon plasma at 10^5 Pa with time delays of 0, 15, 20, 40, 50, 60 and 70 ns from bottom to top, in the wavelength range around 420 nm.



Fig. 3. — Time dependence for the absorption of the $4s[3/2]_2$ argon level : experimental values and fit as explained in text.

In conclusion the present method, which is of wide applicability, will be first devoted to study spectroscopy and kinetics of rare gases. Detailed information about the various transient species intervening in laser plasma decay may be expected by combining time dependence as illustrated above, density effect and extended wavelength range. The high resolution of our system will allow, in particular, determination of oscillator strength ratio.

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