Two-photon excitation cross-section of Xe revisited

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Abstract: Two-photon absorption laser-induced fluorescence (TALIF) is a technique commonly used for density and/or temperature diagnostics, mainly of Oxygen atoms, in flames and plasmas. The quantitative analysis is based on the comparison with the signal obtained, at a similar wavelength, in pure Xenon. The cross-section of Xenon is therefore a key parameter, but has not been confirmed by independent measurements. The present experiment aims to provide a direct measurement of this two-photon cross-section.

Keywords: Two-photon cross-section, TALIF, Xe.

1. TALIF and two-photon cross-section

Laser diagnostics in plasmas frequently resorts to the so-called TALIF technique, which consists in recording the fluorescence that follows two-photon excitation of an atomic species (cf. Fig. 1). The density of oxygen atoms in flames can for instance be monitored via the 3p ^{3}P→3s ^{3}S^o 844.6 nm fluorescence that follows 2 photon-excitation of the 3p ^{3}P level from the 2p^4 ^{3}P ground level, at the wavelength λ = 225.6 nm. A widespread protocol is to compare the fluorescence yield to that measured in Xe vapor, the density of which can be known straightforwardly, when illuminated by the same optical system. Xe atoms can be excited at the nearby wavelength of 224.1 nm to a 6p'[3/2]$_2$ state, then de-excited by spontaneous emission to the 6s'[1/2]$_1$ level at the wavelength λ = 834.9 nm, in an atomic scheme quite similar to the scheme implemented in oxygen, which makes dealing with calibration issues easier, both for the exciting light and for fluorescence detection.

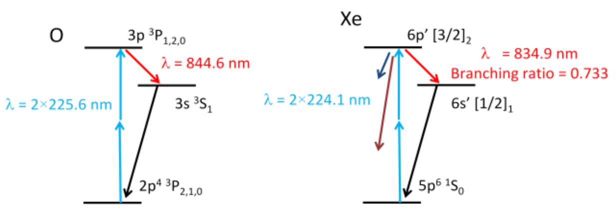

Fig. 1. Relevant energy levels for the two-photon excitation of atomic oxygen and xenon.

Along these lines quantitative analysis, however, relies on the knowledge of the ratio of the two two-photon cross-sections $\sigma^{(2)}$ involved, namely $\sigma^{(2)}$(Xe)/$\sigma^{(2)}$(O). Unfortunately, this ratio was measured only by one team [1] and only the cross-section of oxygen $\sigma^{(2)}$(O) has been measured [2] or calculated [3] directly. A direct measurement of the cross-section $\sigma^{(2)}$(Xe) of Xenon has thus appeared quite desirable.

2. Experiment

We have built an experimental set-up to independently determine the two-photon absorption cross-section of Xenon. When the atomic density becomes high, the attenuation of the laser beam is no longer negligible and the transmission is directly connected to the cross-section [4]. Figure 2 shows the transmission signal around the 6p'[3/2]$_2$ resonance with a single-mode pulse laser excitation of about 1 mJ, 6 ns, 350 µm in diameter passing through a 51 cm cell with an atomic density of 2.12×10^{18} at/cm^3. We report also on this figure the difference between the inverse of the transmitted energy and the inverse of the incident energy. This quantity is directly linked to the spectral line-shape g(ω).

To control photon statistics and get a good reproducibility of the results, we have used a home-made injected single-mode Ti:Sapphire laser [5].

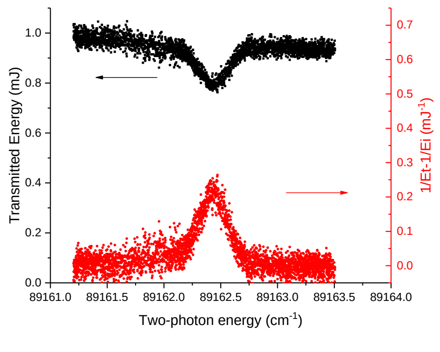

Fig. 2. Two-photon absorption spectrum around the 6p'[3/2]$_2$ resonance of Xe. The cell length is 51 cm and the pressure is 8.61 kPa. The laser pulses have a duration of about 6 ns and the beam waist is about 350 µm.
Left axis: transmitted energy.
Right axis: 1/(transmitted energy) - 1/(incident energy).

We have recorded absorption spectra obtained under different experimental conditions (pressure from 0.75 to 28 kPa and photon flux from 0.6 to 2.6×10^{25} cm^{-2}.s^{-1}).

3. Model and results

When the laser beam (photon flux ϕ) is collimated over the length (L) of the sample (with a density n_0) and has a profile rectangular in time (duration τ) and cylindrical in space ($a = \pi w^2/2$ where w is the beam waist), a simple analytic expression can be obtained for the excitation rate and the absorption of photons. In the weak field regime, the fraction of excited atoms ($\Delta n_e(\tau)$) and of absorbed photons ($\Delta\phi(\tau)$) are given by:

$$\Delta n_e(\tau) \approx \sigma^{(2)} \varphi^2 \tau$$

$$\Delta\phi(\tau) \approx 2\, n_0\, \sigma^{(2)} \varphi L$$

Using this approximation, it appears that the attenuation of the laser beam is no longer negligible when the product $2N_0\sigma\phi L \sim 1$. In contrast, these expressions explain why there is no measurable attenuation in current laboratory plasmas, where atomic densities are of the order of 10^{18} atoms/cm^3.

When attenuation of the photon flux is taken into account, in the frame of the laser pulse, the relative variation of the flux is given by:

$$d\varphi(x,y,z) = -2\,\sigma^{(2)}\, n_0\, (\varphi(x,y,z))^2$$

Thus, the variation of the flux of photons at every position *z* in the sample follows:

$$\varphi(x,y,z) = \frac{\varphi(x,y,0)}{1 + 2\,\sigma^{(2)}\, n_0\, z\, \varphi(x,y,0)}$$

In the experiment, we do not measure an instantaneous flux, but the energy integrated for the pulse duration. We can use numerical calculations to connect both quantities and to take into account a more realistic spatial and temporal description of the laser beam. A numerical example of the flux attenuation is given in figure 3.

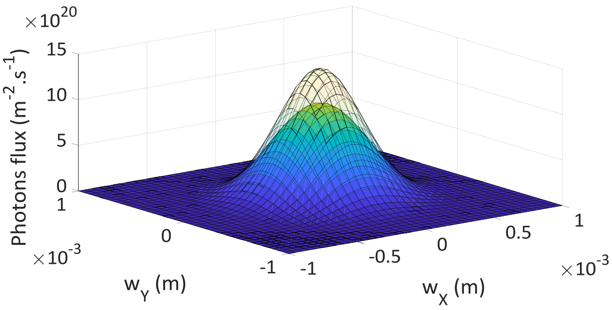

Fig. 3. Variation of the photon flux for a Gaussian beam (w = 500 µm, t = 6 ns, E = 500 µJ, λ = 226 nm) with a cell of 51 cm, an atomic density of 10^{18} atoms/cm^3 and an effective cross-section of 1.215×10^{-35} cm^4.

Our preliminary measurements lead to a value of the effective cross-section of the 6p'[3/2]$_2$ state of about $\sigma_0^{(2)} = 1.5\times10^{-35}$ cm^4, a value lower by a factor 2 than the commonly accepted value [1]. We have also measured the cross-section of the 6p'[1/2]$_0$ state and found a preliminary value of $\sigma_0^{(2)} = 1.9\times10^{-35}$ cm^4.

We are now modeling the beam propagation more carefully, taking into account previously neglected effects such as photo-ionization. Preliminary calculations indicate that the cross-section value should not vary by more than 10%. We are also trying to determine an uncertainty in our measurement, which can be quite high due to the non-linearity of the process and the difficulty of measuring certain quantities, not least the energy of the laser pulse at 226 nm.

4. References

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