Measurements of two-photon absorption cross-section of Xe for laser spectroscopic techniques calibration

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Abstract. Two-photon transition cross section for one xenon transition (5p^6 1S_0-5p^56p[1/2]_0) will be measured with very low uncertainty by two-photon polarization spectroscopy. The method, which is explained in this paper, is based on a comparison of direct measurements of the two-photon absorption in xenon gas at two closed wavelengths (resonant and no resonant).

1. Introduction
In the last years, the Laboratory of Laser Spectroscopy in Valladolid University (Spain) has been dedicated to the development of new spectroscopy techniques for the diagnosis of plasmas far off thermodynamic equilibrium, i.e. plasmas for technological application and combustion systems. In this field of plasma diagnostics, tools are demanded for quantitative measurements of atomic ground state density for species like hydrogen, carbon, nitrogen and oxygen. In our laboratory we have established a high precision two-photon polarization spectroscopy technique, which is not affected by quenching and allows measuring directly the two-photon absorption. It has been used e.g. to determine absolute atomic hydrogen density in quite different environments with an uncertainty as low as 10% [1, 2].

In general, one of the most extended methods in order to determine the absolute concentration of ground state atoms in plasmas is Multi-photon Laser-Induced Fluorescence (MP-LIF). A quantitative interpretation of the signal requires the knowledge of the thermodynamical state of the plasma and all interaction mechanisms. Different calibration methods have been described in the literature in order to obtain quantitative results. Nowadays one of the most popular is the use of calibrated two-photon LIF signal in noble gases, like Xe and Kr, with a two-photon resonance line spectrally close to the investigated atomic transition [3, 4]. This kind of comparisons needs a precise knowledge of several atomic parameters like radiative lifetimes of the excited states, their quenching coefficients for different collisions, and the relevant ratios of two-photon excitation cross sections. The objective of our present work is to use two-photon polarization spectroscopy as a new method for the determination of two-photon excitation cross-sections of noble gases.

In this paper a procedure is described for the determination of the cross section of the Xe transition: 5p^6 1S_0-5p^56p[1/2]_0.

2. Principle of two-photon polarization spectroscopy
The schematic set-up for measuring two-photon absorption by polarization spectroscopy is shown figure 1. A circularly polarized pump beam of high irradiance is focused into the investigated medium,
where it overlaps with a weak linearly polarized signal beam. The signal beam experiences the two-photon resonant optical anisotropy induced in the medium by the circularly polarized pump beam. As a consequence, the linear polarization of the signal beam is rotated by the dispersion part and becomes slightly elliptic by the absorption part. The polarization change can most sensitively be detected behind a polarization analyzer set at a right angle with respect to the original polarization direction [5].

Figure 1: Schematic set-up of two-photon polarization spectroscopy

The signal irradiance behind the analyzer, $E_S$, is found to be for weak absorption [5-7]:

$$
\frac{E_S}{E_R} = \frac{1}{16} \left( \frac{\sigma_{ij}^{(2)} N \langle E_p \rangle}{h \nu_p} p(J_i, J_f) \right)^2 P(\Delta \nu) + R
$$

Here, $N$ is the number density of the absorbing species (in units of m$^{-3}$), $\langle E_p \rangle$ is a suitably defined mean value for the product of the pump irradiance $E_p$ (W/m$^2$) and the length $l$ of the overlap volume including the correlation function $g^{(2)}$, $R$ denotes the residual transmittance of the crossed polarizers, i.e. $E_S/E_R$ as measured without pump beam. The polarization line shape function $P(\Delta \nu)$ (in units of s$^2$) is given by the sum of the squares of the absorption line shape and the related dispersion as well, both are connected by the Kramers-Kronig relation. Where $\Delta \nu = \nu_s + \nu_p - \nu_0$, is the frequency difference between the sum of the laser-frequencies $\nu_s + \nu_p$ (of the signal and pump beam), and the resonance frequency $\nu_0$. The factor $p(J_i, J_f)$ results from the polarization dependence of the two-photon cross section $\sigma_{ij}^{(2)}$ and its value for the transition in this work is: $p (0,0) = 1$.

Two-photon polarization spectroscopy allows measuring directly the spatial variation of the atomic density, even if there are strong spatial variations of the thermodynamic state and the corresponding quenching rates in the medium [1, 2, 6]. In the case of counter-propagating beams, like in figure 1, the first order Doppler shift cancels and there is no Doppler-broadened background at all, because only absorption of one photon from each beam causes the signal. Additionally, collinear beams allow to measure Doppler broadened profiles, hence the determination of kinetic temperatures. Indeed, the application of the two-photon polarization spectroscopy is only limited by the fact, that the signal has to be measured against the background signal due to the residual transmittance $R$.

3. Determination of Xe cross section via a standard of two-photon absorption

According to Eq. (1), $\sigma_{ij}^{(2)}$ can be determined without further provision if the quantities $N$, $\langle E_p \rangle$, $P(\Delta \nu)$ and $R$ are known. For a Xe gas, the only crucial contribution to the total uncertainty of $\sigma_{ij}^{(2)}$ is the absolute measurement of $\langle E_p \rangle$. Thus, this uncertainty can be reduced if the measurement of $\langle E_p \rangle$ can be avoided by the experimental concept. This is possible by combining two-photon polarization spectroscopy with a standard of two-photon absorption. Such a standard is given by an absolute known two-photon absorption cross section and a well defined number density.

In a previous paper [6], and in order to measure the absolute number densities in a hydrogen plasma, a standard of atomic hydrogen densities was established by a stationary hydrogen arc plasma; with the well known two-photon cross section of the 1S-2S transition this hydrogen standard also
represents a standard of two-photon absorption. This standard is a rather complex plasma source which is not easily available in other laboratories, therefore in the same experimental set-up, xenon gas was established to be the transfer standard of absolute two-photon absorption at the frequency of the 1S-2S resonance of hydrogen. This was possible because xenon gas at room temperature and 500 mbar pressure yields to sufficiently large non resonant two-photon polarization signal. It was proved in detail, that the non resonant signal shows the same dependence on particle density and pump irradiance as a resonant one. To account for this non resonant polarization signal Eq. (1) can be written:

$$\frac{E_S}{E_R} = \frac{1}{16} \left( c_{Xe} N_{Xe} \frac{\langle E_p I \rangle}{h \nu_p} \right)^2 + R \quad (2)$$

where the constant factor $c_{Xe}$ substituted the product: $\sigma_{H}^{(2)} P_{if}(\Delta \nu)$. As a consequence, the comparison of the non resonant polarization signal of xenon with the resonant of the hydrogen standard obtained at the 1S-2S transition frequency ($p(0, 0) = 1$) keeping unchanged the overlap parameter $<E_P I>$, yields the following relation:

$$c_{Xe} = \sigma_{H}^{(2)} N_{Xe} \frac{N_{Xe}}{N_{Xe}} \left( \frac{E_S / E_R}{E_S / E_R} - \frac{R_{Xe}}{R_{H}} \right)^{1/2} = (1.05 \pm 0.06) \times 10^{-57} \text{ m}^4 \text{ s} \quad (3)$$

Although the comparison is performed at the same frequency, the residual transmission present in the actual measurement may be different, e.g. due to different pressure used for xenon and operating the standard.

Based on this calibration, the two-photon xenon cross section can be related to the factor $c_{Xe}$ via a comparison of resonant and non resonant signals, keeping the pump beam unchanged:

$$\sigma_{Xe, if}^{(2)} = c_{Xe} \frac{1}{P_{Xe}(J_i, J_f)} \left[ \frac{E_S / E_R}{E_S / E_R} - \frac{R_{Xe, if}}{R_{Xe}} \right]^{1/2} \quad (4)$$

Hence, measurements of various Xe two-photon cross section can be performed in the same xenon gas cell.

The experimental arrangement needed for this kind of measurements is already operative in our Laboratory. A comprehensive description of the experimental set-up can be found elsewhere [8], here we only provide details concerning this work. The radiation for Xe two-photon excitation is generated by an advanced tuneable single longitudinal mode UV-laser spectrometer of nanosecond pulse duration, that provides up to 10 mJ pulse energy in 2.5 ns with spectral quality of 300 MHz bandwidth. This laser can be tuned from 243 nm to 249 nm, i.e. over the entire wavelengths interval from the non resonant to the resonant two-photon transition. The term $<E_P I>$ during this process can be considered constant, because the laser power can be measured and the overlap volume can be kept unchanged.

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