Absolute Cross Sections of Multi-Photon Ionization of Xenon by the Comparison with Process of its Electron-Impact Ionization

A. A. Mityureva, A. A. Pastor, P. Yu. Serdobintsev, N. A. Timofeev

Abstract—Comparison of electron- and photon-impact processes as a method for determination of photo-ionization cross sections is described, discussed and shown to have many attractive features.

Keywords—Transition probability, cross section, photoionization, electron-ionization, multi-photon process.

I. INTRODUCTION

THE electron- and photon-impact excitation and ionization of atoms and molecules are the fundamental collision processes and occur in a wide variety of active mediums, thus a knowledge of their cross section data and an understanding of their mechanisms are required.

In recent years, the process of multi-photon ionization (MPI) of atomic and molecular species has been a subject of intensive experimental and theoretical studies and rapid progress in this field has been largely driven by advancement in high-power shot-pulse laser techniques. However, the calculation of these cross sections is still difficult, and in spite of numerous experimental investigations, uncertainty of the reliable values has stayed. Moreover it isn't always cleared up on which definitely way, by means of what atomic levels MPI process is carried out!

It doesn't concern the determination the probability of electron-impact excitation (EIE) and ionization (EII) of atoms which has not been only a subject of investigations over decades but also has given the accurate values of electron excitation and ionization cross sections, in particular for the rare gases. Such measurements for rare gaseous targets have a very long history and the determination of the cross sections for EIE and EII was studied rather well from the ground states [1], thus we can use them to a benchmark item. Besides that the cross sections for other some more exotic processes such as the excitation and ionization from the excited states especially from or to the metastable ones have been measured particular by us [2]–[6] too and we are going to use them as a benchmark item too.

In this paper we offer the way for the determination of total MPI cross sections some atoms by means of comparison efficacy of MPI and EII processes in the same experiment and the determination of MPI-cross section values over the wellknown EII - cross section. We have performed measurements of the multi-photon ionization of electrons from the outer shell of xenon.

Xenon is a particularly appealing ionization medium for this application because it is a heavy atom that requires only three steps of photo-ionization what is rather convenient for our experimental facilities.

This paper reports an initial step toward extending the technique of comparison of two different impact processes.

II. METHOD AND EXPERIMENTAL SETUP

In this section we consider 1) – the basic relations and 2) – the experimental technique.

1) The Basic Relations

The considered approach consists in the accurate measurement of ratio of total cross sections for MPI- and EII-processes, followed by the determination of the total MPI cross sections using the measured cross sections ratios and well-known total EII cross sections. Thus the ionization process of Xe atoms as a result of both three-photon and electron beam impacts have been studied. We have employed atom of xenon as an initial step of studying because it is the best object for investigation of MPI rare gases at present.

The total MPI ionization cross section of the gas under study is denoted as $\sigma_{ph}^{(p)}$, where p – is a parameter of the process multiplicity and p=3 in our case for Xe, is determined by as

$$\sigma_{ph}^{(3)} = \gamma \cdot v_{el} \cdot \frac{V_{el}}{V_{ph}} \cdot \frac{T_{el}}{T_{ph}} \frac{1}{\Phi_{ph}^3} \quad \left(cm^6 c^2 \right)$$

In this equation, γ is the fraction of ionized atoms:

$$\gamma \equiv \frac{N_{ph}^{+}}{N_{el}^{+}}$$

where

and

$$N_{el}^+ = N_0 \cdot V_{el} \cdot v_{el} \cdot T_{el}$$

 $N_{ph}^{+} = N_0 \cdot V_{ph} \cdot v_{ph}^{(3)} \cdot T_{ph}$

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are the full atomic populations resulted in multi-photon and electron impacts, respectively, and registered by a massspectrometer setup; N_0 is the population of the ground state; V_{ph} and V_{el} are the volumes of domains of the multi-photons and electron impacts processes; T_{pf} and T_{el} are the laser and electron beam pulse lengths and $v_{ph}^{(p)}$ and v_{el} are the rates (or probabilities) of multi-photon and electron processes. The value v_{el} is calculated on base of well-known literature data and the value $v_{ph}^{(p)}$ is to be determined. The cross-section of the multi-photon ionization is related with its rate (probability) by equation

$$\sigma_{ph}^{(p)} = \frac{v_{ph}^{(p)}}{\Phi_{ph}^{p}} \left(cm^{2p} \cdot c^{p-1} \right)$$

where $\Phi_{ph} = n_{ph} \cdot c$ - is photons flux density and p - is multiplicity parameter (here is equal to 3). The photons flux density can be estimated as P_{hv} , where P is laser energy flux density, hv - is photon energy.

This approach overcomes many of the limitations of conventional techniques, most essential being unnecessary to measure the absolute values of atoms and ions populations, and is capable of providing accurate absolute cross sections.

2) The Experimental Technique

So for a practical application of the method, which idea is shown in Fig. 1, we used experimental facilities of Saint-Petersburg State University optical center equipped with appropriate laser system. The experimental equipment and arrangement will have been described in detail later [11]. General experimental scheme is represented in Fig. 1.

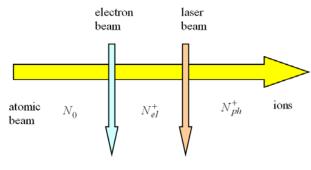


Fig. 1 Scheme of the experiment

Briefly the setup consisted of the ionization chamber with supersonic atomic beam spread along the axis of the chamber, a time-of-flight mass spectrometer with position-sensitive detector in the end of camera, and the electron gun, and the laser beam that crossed atomic beam.

We have studied the multi-photon ionization of 136 Xe atoms from the 5p ${}^{6}{}^{1}$ S₀ ground state to the Xe⁺ ions. Laser pulse has following parameters: laser beam energy 95 µJ, laser energy flux density was about $P \sim 10^{10} \frac{W}{cm^2}$ and duration about 10

ns at 223 nm wavelength.

The electron impact ionization of 136 Xe atoms has been studied by 17eV energy electron beam with current densities about 200 μ A.

The electron and photon ionization measurements were made with the specially calibrated detection system "Stefan Kaesdorf".

III. RESULTS

The photon ionization of xenon has been studied both theoretically and experimentally by various groups and for total, single-, two-, three-, multi-photon and single-, ... multicharge processes ionization and certainly all of them have highly different values. A discussion of the experimental and theoretical distinctions in their values and processes is out of this paper scope. Here we are presenting new approach to the determination of the absolute values of multi-photon ionization cross sections. Gained result for the cross section of three-photon xenon atom ionization is equal to

$$\sigma_{ph}^{(3)} = 6.4 \cdot 10^{-83} \ cm^6 \cdot c^2$$

We compared our result with several theoretical predictions [7]–[9] and made a comparison with the experimental data [10]. The theoretical estimations have given a good self-agreement and don't contradict to our result. The experimental value [10] for the cross section of three-photon non-resonant ionization of xenon equal to $1.1 \cdot 10^{-82} cm^6 \cdot c^2$ has shown rather good agreement with our result.

Thus we can see from these comparisons that it is worthy to use new method for determination of multi-photon ionization cross sections of atoms.

IV. CONCLUSION

We have reported the measurement of the multi-photon ionization cross section of the outer valence shell of xenon atoms by a comparison the results of two impacts – electronic and photon with atoms, an equal registrations of theirs and on the basis of its measured ratios and well-known total electron ionization cross section data the determination of the multiphoton ionization cross section.

Our result for three-photon ionization xenon atoms is found to be in quite good agreement with the data of theoretical estimations [7]–[9]. An agreement of our total cross section with result Schechter et al. [10] was also obtained. It shows that we have a reliable tool for studying and determination of multi-photon ionization cross sections of atoms.

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