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The absolute optical oscillator strengths of the $3p^54s$ and $3p^54s'$ excitations of argon measured by the dipole (γ, γ) method

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Abstract

The optical oscillator strengths of the $3p^54s$ and $3p^54s'$ excitations of argon are of crucial importance in determining the elemental abundance of argon from astronomical observations. Substantial efforts have been made to determine the optical oscillator strengths of the $3p^54s$ and $3p^54s'$ transitions of argon, however, significant discrepancies among the reported values still exist. In this work, the absolute optical oscillator strengths of the $3p^54s$ and $3p^54s'$ transitions of argon have been measured by a newly developed high-resolution dipole (γ, γ) method. A comprehensive comparison with the previous experimental and theoretical data was then made as a cross-check.

Keywords: optical oscillator strength, photoabsorption cross section, argon, dipole (γ, γ) method, inelastic x-ray scattering

(Some figures may appear in colour only in the online journal)

1. Introduction

Argon, as a product of stellar nuclear processes, is widely distributed in the Universe [1–5]. The elemental abundance of argon is extensively used in the astronomic research of the components of interstellar media near the Sun, the evolution of cometary matter, the planetary atmospheres and even the Galaxy. As the basis of spectral analysis, absolute optical oscillator strength (OOS), or the equivalent quantity of a photoabsorption cross section of argon plays a crucial role. Moreover, from the viewpoint of fundamental research, the study of the electronic excitation of argon provides valuable quantitative information for testing and developing theoretical methods.

The absolute OOSs of the $3p^54s$ and $3p^54s'$ transitions of argon have been extensively studied both experimentally and theoretically. The experimental measurements were carried out by the dipole (e, e) [6–9], the electron impact [10, 11], the self-absorption [12–16], the lifetime [17–22] and the pressure-broadening profile [23–26] methods. Among these, some early measurements [8, 9, 11, 20–22] deviate greatly from recent experimental results, so they are excluded from discussion in this paper. For other measurements [6, 7, 10, 12–19, 23–26], the experimental OOS of the $3p^54s'$ can be classified into two groups, with one being about 0.22 [10, 12–15, 18, 25] and the other being about 0.26 [6, 7, 16, 19, 23, 24, 26]. However, for the OOS of the $3p^54s$, there is no clear classification. Considering the important application values of the OOSs of the $3p^54s$ and $3p^54s'$, it is significant that they be determined by a new experimental technique to provide independent cross-checking, and that is the purpose of this work.

As for the theoretical investigation, the OOSs of the $3p^54s$ and $3p^54s'$ have been calculated by different theoretical methods [27–40] since the 1960s. Considering the tremendous advancements in theory and calculational ability, it is believed that recent calculations based on the B-spline method [33], the relativistic distorted wave (RDW) [34], the relativistic many-body perturbation theory [35] and the random phase approximation with exchange (RPAE) [36] are the more reliable ones. Unfortunately, most experimental results are not consistent with these recent calculations. It has also been found that the OOSs of the $3p^54s$ and $3p^54s'$ have been evaluated [41, 42] by averaging previous experimental results.

In this paper, the newly developed dipole (γ, γ) method, which involves inelastic x-ray scattering (IXS) operated at a negligibly small momentum transfer, has been applied to measure the absolute OOSs of excitations from the ground state to $3p^54s[3/2]_1$ and $3p^54s'[1/2]_1$ of argon. The details of the experimental measurements are presented in section 2, while the experimental results and discussions are presented in section 3; finally, a summary of this work is given in section 4.

2. Experimental method

The present experiment was performed on the Taiwan Beamline BL12XU at SPring-8 [43, 44], and the experimental procedures employed are similar to our previous work [45, 46]. In brief, a Si (333) monochromator and a Si (555) spherical analyzer were applied to achieve a high energy resolution of about 70 meV. During the experiment, the analyzer energy of the scattered photon was fixed at 9888.11 eV, while the incident photon energy varied, allowing the energy loss to be easily determined. The samples of argon and helium were sealed in a gas cell with Kapton windows and put on the experimental platform for measurement in turn; the helium sample was used here for normalization. The pressure in the gas cell was maintained at 0.143 MPa for argon and 0.98 MPa for helium, respectively. In addition, the actual transmissivities of the two sample gases were measured, and all the spectra of helium and argon were measured at room temperature. The spectrum of the excitations for $3p^54s[3/2]_1$ and $3p^54s'[1/2]_1$ of argon with an energy resolution of about 70 meV is shown in figure 1.

In order to determine the intensity of the individual excitation, a least-squares fitting was used to fit the experimental spectra. In the fitting procedure, the peak profile was described by a Gaussian function. Then the relative OOS is given as:

$$f_0^r(\omega_n) = B'_\gamma(\omega_n)\frac{N(\omega_n)}{N_0}\frac{1}{D_0\alpha}\frac{1}{l_{\text{eff}}}\frac{1}{n_0P}. \qquad (1)$$

B'_γ is the so-called Bethe–Born conversion factor, which is determined accurately by simulating the actual arrangement of the light path considering the rectilinear propagation of light. $N(\omega_n)$ and N_0 stand for the counts of the scattered photons and the intensity of the incident photons, respectively. l_{eff}, n_0 and P are the collision length, the density of the target at 0.1 MPa, and the pressure of the target in units of MPa, respectively. D_0 is a constant determined by the detection efficiencies of the ionization chamber and the detector of the scattered photon. The transmissivity α is determined by the sample species and its pressure, and it can be measured accurately with and without the sample gas in the gas cell by an ionization chamber after the gas cell. In order to obtain an absolute OOSs, a simple normalization method is used through:

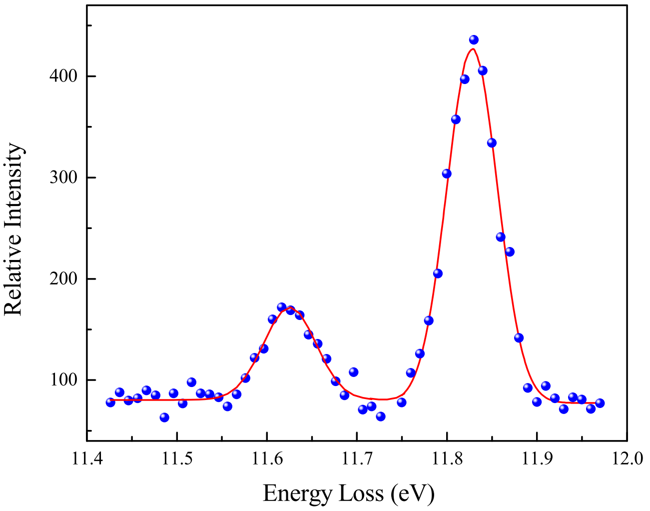

Figure 1. The dipole (γ, γ) spectrum of argon for the excitations for $3p^54s[3/2]_1$ and $3p^54s'[1/2]_1$ at a scattering angle of 2°. Solid blue circles: the experimental data; red line: the fitted result.

$$f_0(\omega_n) = \frac{B'_\gamma(\omega_n)}{B'_\gamma(2^1P)_{\text{He}}}\frac{[N(\omega_n)/N_0]}{[N(2^1P)/N_0]_{\text{He}}}\frac{\alpha_{\text{He}}}{\alpha}\frac{P_{\text{He}}}{P}f_0(2^1P)_{\text{He}}. \qquad (2)$$

Where the 2^1P of helium, whose OOS has been investigated with a high accuracy both experimentally [47] and theoretically [48], was measured at the same scattering angle and used to normalize the results of the argon. A similar normalization procedure was used in our previous papers [45, 46].

According to equation (2), the experimental errors of the OOSs of argon come from the contributions of the Bethe–Born conversion factors, the statistical counts, the transmissivity and the pressures of both argon and helium, as well as the OOS of the 2^1P of helium. The above-mentioned uncertainties have been investigated carefully and listed in table 1. It should be mentioned that the Bethe–Born conversion factor of the dipole (γ, γ) method have an accuracy of better than 0.2% due to the clear geometric arrangement of the setup and the rectilinear propagation of light. It can be seen from table 1 that the main experimental error contributions are due to the statistical counts. These counts can, in principle, be greatly reduced by accumulating them—although it is hard to obtain a long beam time. The total experimental errors are also listed in table 1.

Table 1. Uncertainty budget.

Source	σ
Bethe–Born conversion factor(4 s′, 4 s, 2^1P)	<0.2%
N(4 s′)	2.8%
N(4 s)	7.8%
$N(2^1$P)	2.2%
N_0(4 s′, 4 s, 2^1P)	<0.3%
α(4 s′, 4 s, 2^1P)	<0.2%
P(4 s′, 4 s, 2^1P)	<0.5%
f_0 (2^1P)	$<10^{-6}$
total(4 s′)	3.6%
total(4 s)	8.1%

3. Results and discussion

The absolute optical oscillator strengths for the $3p^6 \longrightarrow 3p^54\,s$ and $3p^54s'$ transitions of argon are listed in table 2 and shown in figures 2 and 3 along with the previous theoretical and experimental ones. It should be emphasized that some early measurements [8, 9, 11, 20–22] and calculations [27–32, 37–40] are not shown in figures 2 and 3 for clarity, as pointed out in the introduction.

It can be seen from figures 2 and 3 that the present dipole (γ, γ) OOSs are in excellent agreement with those of the dipole (e, e) [6, 7] and the recent lifetime measurements [17, 19]. It should be emphasized that the lifetime measurement for the $3p^54s$ of Chornay *et al* [17] should be more reliable than the other lifetime measurements due to the fact that it is free from the cascade effect as a result of the scattered electron being energy-selected as a start signal. Considering the excellent agreement of the OOSs of the $3p^54s$ of Chornay *et al* [17] and Federman *et al* [19], the OOS of the $3p^54s'$ of [19] should be reliable, which agrees well with the present dipole (γ, γ) and those of the dipole (e, e) results. Although the OOSs of the $3p^54s$ measured by Westerveld *et al* [14] (with self-absorption) and Lewis [24] (with the pressure-broadening profile) are consistent with the one measured in this work, the OOSs for the $3p^54s'$ do not match the present result. Other measurements [12, 13, 15, 16, 18, 23, 25, 26] shown in figures 2 and 3

Table 2. Experimental and theoretical OOSs for the $3p^54s$ and $3p^54s'$ transitions of argon.

	$^2P_{3/2}4\,s$	$^2P_{1/2}4s'$
Experiment		
Dipole (γ, γ)		
Present work	0.0664 ± 0.0054	0.257 ± 0.0091
Dipole (e, e)		
Wu *et al* [6]	0.0676 ± 0.0040	0.259 ± 0.015
Chan *et al* [7]	0.0662 ± 0.0033	0.265 ± 0.013
Electron impact		
Li *et al* [10] 1988	0.058 ± 0.003	0.222 ± 0.02
Absolute self-absorption		
Gibson and Risley [12]	0.0580 ± 0.0017	0.2214 ± 0.0068
Tsurubuchi, Watanabe and Arikawa [13]	0.057 ± 0.003	0. 213 ± 0.011
Westerveld, Mulder and Eck [14]	0.063 ± 0.005	0. 240 ± 0.02
Jongh and Eck [15]		0.22 ± 0.02
Ligtenberg *et al* [16]	0.0717 ± 0.0024	0. 2683 ± 0.0075
Lifetime		
Chornay, King and Buckman (*electron-photon coincidence*) [17]	0.065 ± 0.005	
Lawrence (*delayed coincidence*) [18]	0.059 ± 0.003	0.228 ± 0.021
Federman *et al* (*Beam foil*) [19]	0.064 ± 0.003	0.257 ± 0.013
Pressure-broadening profile		
Copley and Camm [23]	0.076 ± 0.008	0.283 ± 0.024
Lewis [24]	0.063 ± 0.004	0.278 ± 0.02
Vallee, Ranson and Chapelle [25]	0.051 ± 0.007	0.210 ± 0.030
Stacey and Vaughan [26]	0.036 ± 0.004	0.275 ± 0.02
Recommended values:		
Recommended by Verner *et al* [41]	0.0665	0.244
Recommended by Gargioni and Grosswendt [42]	0.060	0.233
Theory		
Zatsarinny and Bartschat [33]	0.0640	0.242
Vos, McEachran and Zhu [34]	0.0736	0.222
Avgoustoglou and Beck [35]:		
(1) length	0.0672	0.248
(2) velocity	0.0670	0.242
Amusia [36]		$0.278(f_1+f_2)$

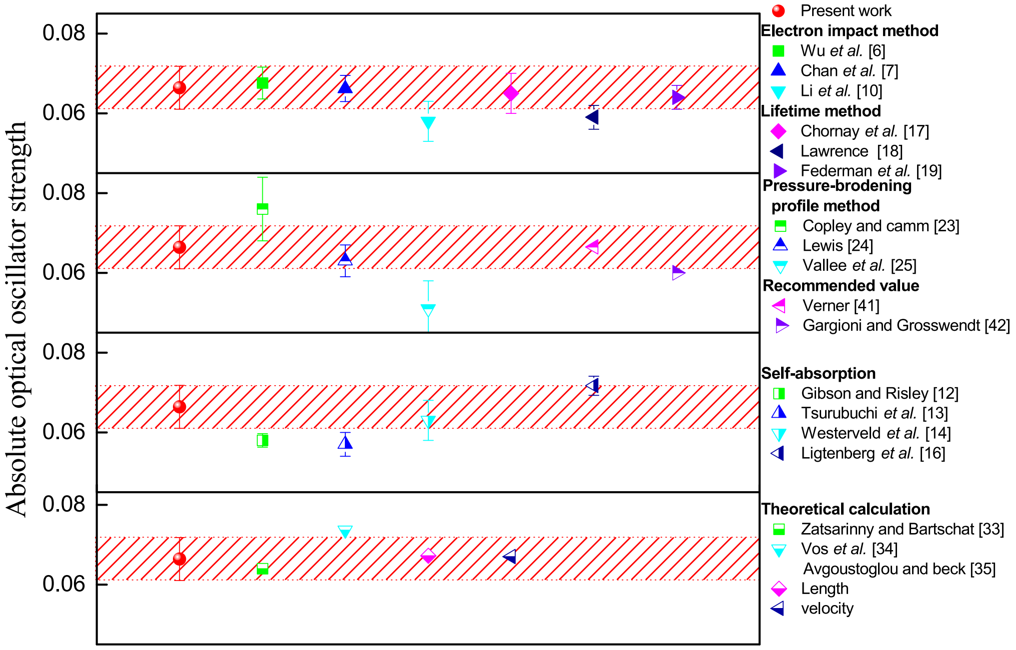

Figure 2. The OOS of the $3p^5 4s$ measured by the present dipole (γ, γ) method along with the previous experimental and theoretical results.

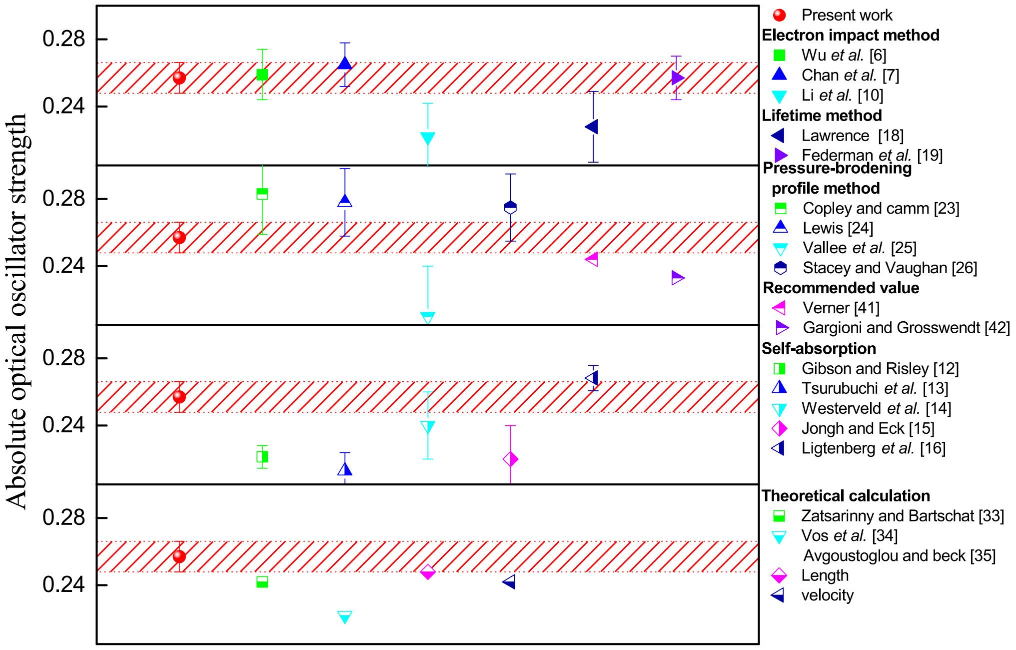

Figure 3. The OOS of the $3p^5 4s'$ measured by the present dipole (γ, γ) method along with the previous experimental and theoretical results.

deviate from the present data systematically. Considering the inherent demerits and limitations of the electron impact, the self-absorption, the lifetime and the pressure-broadening profile methods, as summarized and pointed out by Chan *et al* [49], it is reasonable that the present dipole (γ, γ) results, those of the dipole (e, e) [6, 7] and the lifetime measurements [17, 19] should be more reliable. It should be pointed out that although the dipole (e, e) method is limited by the rapid variation of its Bethe–Born conversion factor with the excitation energy, the systematic investigation of Brion's group shows that this limitation is not severe and their OOSs are free from any systematic uncertainty for a lot of atoms and molecules. To the best of our knowledge, the only shortcoming of the dipole (γ, γ) method is its low count rate [45, 46], and the resulting statistical error can be determined accurately and listed in table 1.

As for the theoretical calculations, except for those of the recent RDW [34] and the RPAE [36], the calculated results are highly consistent among themselves, with the OOSs of the $3p^5 4s$ being 0.064−0.0672 and the OOSs of the $3p^5 4s'$ being 0.242−0.248. It can clearly be seen from figures 2 and 3 that for the $3p^5 4s$ the present OOS is in good agreement with those calculations and the recommended value [33, 35, 41], while the OOS of the $3p^54s'$ in this work is slightly larger than the results of references [33, 35, 41]. In order to elucidate this discrepancy, more theoretical works are greatly recommended.

4. Summary

Considering the importance and the application values of the OOSs of the $3p^54s$ and $3p^54s'$ excitations of argon, they have been measured by the newly developed dipole (γ, γ) method. The present results provide an independent cross-check with the previous experimental and theoretical data. It was found that the present OOSs of the $3p^54s$ and $3p^54s'$ are in good agreement with the dipole (e, e) measurements [6, 7] and some new lifetime results [17, 19]. However, apparent discrepancies compared with the self-absorption [12–15], the pressure-broadening profile [23–26] and the lifetime [18] measurements have been observed—especially for the OOSs of the $3p^54s'$. Most recent theoretical calculations regarding the $3p^54s$ are in agreement with the present value, while the calculated OOSs of the $3p^54s'$ are generally lower than the value in this work. Detailed analysis shows that the present dipole (γ, γ) method is a relatively simple and reliable experimental method for determining the OOSs of the discrete transitions of atoms and molecules.

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