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Compton Scattering Measurements of Ar and O_2 Using 59.54 keV Gamma-rays

Erdem ŞAKAR[1*], Ali GÜROL[1]

[1]Atatürk Üniversitesi, Fen Fakültesi, Fizik Bölümü, Erzurum, Türkiye
Erdem ŞAKAR ORCID No: 0000-0002-1359-4464
Ali GÜROL ORCID No: 0000-0001-9494-3533

**Corresponding author: erdem@atauni.edu.tr*

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Electron momentum distribution

Abstract: In this study, it has been investigated of the Compton profiles (CP) of the Ar and O_2 gases using a Compton spectrometer having annular ^{241}Am radioactive source. The photons emitted from the radioactive source and scattered photons from the samples were recorded using HPGe detector. In order to obtain experimental Compton profiles of the samples the Warwick codes were used. The experimental results obtained were compared with the theoretical results calculated by Hartree-Fock method. The results clearly show that the experimental and theoretical results are in good agreement, especially in the high momentum region. It was observed that while the difference between the experimental and theoretical results of Ar and O_2 gases in the $p_z = 0$ region is about 4%, this difference is below 1% after $p_z = 2$.

59.54 keV'lik Gama-ışınları Kullanılarak Ar ve O_2'nin Compton Saçılma Ölçümleri

Anahtar Kelimeler
Compton profile,
Saçılma,
Hartree-Fock metodu,
Gama-ışınları,
Elektron momentum dağılımı

Öz: Bu çalışmada, dairesel ^{241}Am radyoaktif kaynağa sahip bir Compton spektrometresi kullanılarak Ar ve O_2 gazlarının Compton profilleri araştırılmıştır. Radyoaktif kaynaktan yayınlanan fotonlar ve numunelerden saçılan fotonlar HPGe detektörü kullanılarak kaydedildi. Numunelerin deneysel Compton profillerini elde etmek için Warwick kodları kullanıldı. Elde edilen deneysel sonuçlar Hartree-Fock yöntemi ile hesaplanan teorik sonuçlarla karşılaştırıldı. Sonuçlar, özellikle yüksek momentum bölgesinde deneysel ve teorik sonuçların uyumlu olduğunu açıkça göstermektedir. $p_z = 0$ bölgesindeki Ar ve O_2 gazlarının deneysel ve teorik sonuçları arasındaki fark yaklaşık % 4 iken $p_z = 2$'den sonra bu farkın % 1'in altında olduğu görülmüştür.

1. INTRODUCTION

In Compton scattering, which is the inelastic scattering of X or gamma rays from electrons, scattered photons are exposed to a Doppler broadening due to the velocity of the electrons. This broadened line shape is directly related to the momentum of electrons in the target material and is called Compton profile. The theory of the CP is mainly based upon impulse approximation (IA). According to this approximation, the collision process occurring between the photon and target material electrons takes place in a very short period of time, and accordingly, the potential energy of the material remains constant in this process [1,2].

The spectrum of the inelastically scattered photons is related to the electron momentum distribution of the investigated material. For this reason, the CP of any material can be obtained using ground-state electron momentum density $(n(\boldsymbol{p}))$ [3,4]:

$$J(p_z) = \iint n(\boldsymbol{p}) dp_x dp_y \qquad (1)$$

where p_x , p_y and p_z are the electron momentum components in Cartesian coordinates, and $n(\boldsymbol{p})$ can be calculated by following:

$$n(\boldsymbol{p}) = \sum_i \left| \iint \psi_{i(r)} \exp\langle -i\vec{p} \cdot \vec{r} \rangle \, dr \right|^2 \qquad (2)$$

where $\psi_{i(r)}$ is the electron wave functions (WFs). Based on this, the CP involve knowledge with regard to the WFs and the orbital-wise electron occupation. This matchless property of the inelastic scattering technique for researching the WFs has been utilized to analyze the force of the approximation techniques in the electronic structure calculations.

There are many studies in literature for determining the CP of solids and liquids [1,2,5]. Conversely, there are a limited number of studies on CP measurements of gases. Eisenberger and Platzman [6] determined the electron momentum distributions of He and H_2 gases by Compton scattering and demonstrated that the experimental results obtained for He were in good agreement with the Hartree-Fock theoretical results. In experimental measurements made by Eisenberger and Reed [7] using 160 keV gamma rays emitted from ^{123}Te source, the CPs of helium, nitrogen, argon and krypton gases were measured. The data obtained were compared with HF results and they obtained the HF calculations for Ar and Kr gases differ less than 1% at q = 0 from experimental data. This study also revealed the conclusion that gamma ray use in CP measurements will produce more accurate results than X ray use. Tong and Lam [8] made CP measurements for some inert gases. They compared the experimental results with the theoretical profiles obtained using both HF and LDA (local density approximation). Accordingly, the LDA method exhibited good results in elements with high atomic numbers, excluding the tail region where contains the effects of multiple scattering.

Kobayashi et al. [9] determined the Compton profile of nitrogen gas using synchrotron radiation, and compared the obtained experimental results with previous studies. They showed that the new results were about 0.05% different from the previous results at q = 0. Sakurai et al. [10] measured the CP of some inert gases using the same spectrometer. They compared the results of the experiments with Dirac-Hartree-Fock calculations. The most interesting result in this comparison was the mismatch between the increasing atomic number and the experiment and theory. This study highlights the importance of considering electron correlations in relativistic calculations. Ma et al. [11] conducted CP measurements of NO and C_2H_2 gases with stimulating photons of 20 keV in the center of BL15U1 Shanghai Synchrotron Radiation Facility (SSRF). This study revealed that Synchrotron Radiation sources should be used in experimental CP measurements and the results should be supported by stronger theoretical models such as DFT instead of HF.

In the present study, Compton profiles of argon and oxygen gases are presented using 59.54 keV gamma rays emitted from ^{241}Am radioactive source. The results obtained were compared with the theoretical results obtained with the HF method, and the causes of harmonization and incompatibilities in different momentum transfer regions were tried to be explained.

2. MEASUREMENTS AND DATA ANALYSIS PROCEDURE

Compton profile measurements were performed using a well-designed Compton spectrometer shown in Figure 1. In this experimental geometry, the measured experimental mean scattering angle is 165°. The properties of the spectrometer and detector used in the experiment were explained in our previous study [3]. Total resolution of the experiment including geometrical broadening was 0.65 a.u. In order to decrease the statistical errors, the live time of the experiment was set to 4.4x10^{5}s. The measured spectrums of the Ar and O_2 gases are given in Fig 2 and 3.

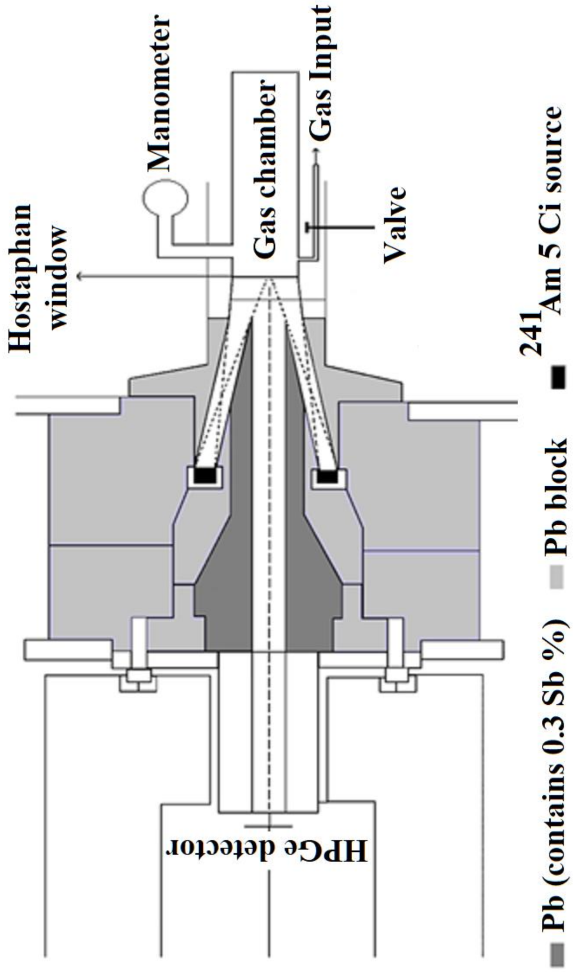

Figure 1. Experimental setup

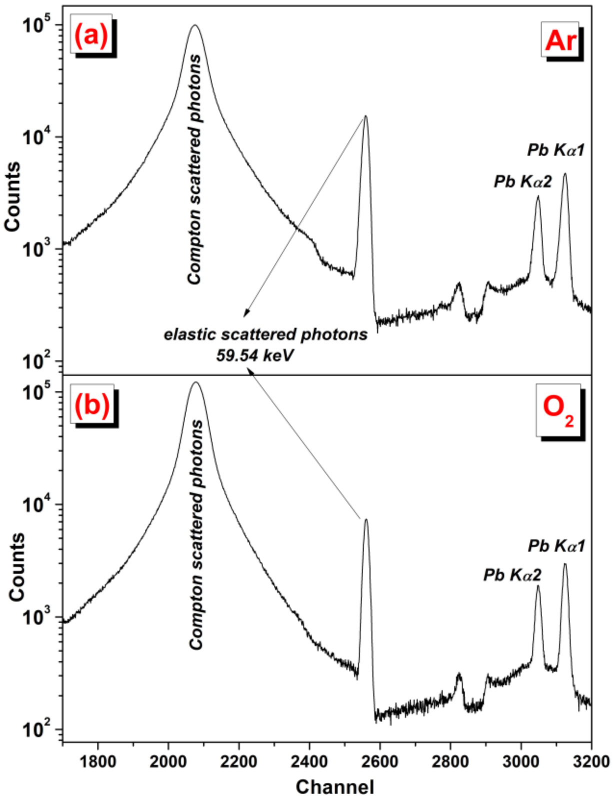

Figure 2. Compton spectrums of the Ar (a) and O_2 (b)

The gas chamber used in this study was first vacuumed and the air inside was evacuated. The gases were then taken into the sample chamber with the help of the tube connected to the gas cell. In this process, the pressure of the sample room was checked at regular time intervals and the ambient temperature was measured regularly using a GOERZ type thermoscript.

Various systematic corrections should be applied to the measured raw spectrum ($M(w)$) to obtain CP of the investigated samples. These corrections are given by equation 3.

$$C(w) = G(w)X(w)A(w)[M(w) - B(w)] \quad (3)$$

where $B(w), G(w), A(w)$ *and* $X(w)$ are the background, efficiency, sample self-absorption and relativistic cross-section corrections, respectively [12].

The self-absorption and relativistic Compton cross section corrections can be calculate using equations 4 and 5, respectively.

$$A(w) = \frac{[\mu(w_2)\mathrm{csc}n_1 + \mu(w_1)\mathrm{csc}n_2]t}{1 - \exp\{-t[\mu(w_2)\mathrm{csc}n_1 + \mu(w_1)\mathrm{csc}n_2]\}} \quad (4)$$

where t is the sample thickness, μ is the linear absorption coefficient (LAC), and n_1 and n_2 are the angle of incident and scattered in reference to the sample surface, respectively. The LAC values were taken from WinXCOM [13] based Phy-X/PSD [14] software.

$$X(w) = \frac{1}{2c^2}\frac{w_2}{w_1}[c\sqrt{w_1^2 + w_1^2 - 2w_1w_2\cos\varphi} + q(w_1 - w_2)]^{-1}.B \quad (5)$$

$$B = \frac{w_1(1+q/c)}{w_2(1-q/c)} + \frac{w_2(1-q/c)}{w_1(1+q/c)} + 2c^2[\frac{1}{w_1(1+q/c)} - \frac{1}{w_2(1-q/c)}] + c^4[\frac{1}{w_1(1+q/c)} - \frac{1}{w_2(1-q/c)}]^2 \quad (6)$$

Detailed descriptions of well-known terms used in equations 5 and 6 are available in Williams 1977 [2]. Finally, the corrected data $C(w)$ were deconvoluted with detector resolution function using equation 7 and, all spectrums normalized relative to the number of electrons of the samples.

$$\int_{-\infty}^{+\infty} J(P_z)d\,z = \frac{1}{T}\int_{-\infty}^{+\infty} (C(w) * R(w))dw\frac{dq}{dw} = N\ electrons \quad (7)$$

In the experimental data analysis, the software prepared by Warwick University Compton research group was used [15].

3. RESULTS AND DISCUSSION

The Compton profiles of the experimentally measured Ar and O_2 gases and their theoretical results obtained using the HF method are shown in Figures 3 and 4, respectively. The X axis (p_z values) in these profiles is directly related to the positions of electrons in the orbitals. It is known that electrons in inner orbitals move at relatively higher speeds than electrons in outer orbitals. This allows the inner orbital electrons to have higher momentum values. For these reasons explained, regions with low p_z values in Compton profile spectra are generally created by outer orbitals (valence) electrons, while large p_z values are created by core electrons. Also, unlike valence electrons, the core electrons do not play a role in chemical bonding process, and are also less affected by external influences such as pressure and temperature. Therefore, the wave functions of the core electrons are largely similar to the free electron wave functions. Thus, the Hartree-Fock Compton profiles obtained from free electron wave functions generally comply with experimental data in regions controlled by core electrons (large p_z values). This is clearly seen in the Compton profiles of the Ar and O_2 gases given in Fig 3 and 4. In these figures, in general, the Compton profiles of the measured both gases are in agreement with the HF theoretical results calculated using the free electron wave functions. In particular, this harmony reached better levels in high momentum regions ($p_z > 3$ a.u.) in both gases. However, in the lowest momentum regions (from p_z=0 a.u. to p_z=2 a.u.) there is some incompatibility due to both the known deficiencies of the HF model and the various difficulties of CP measurements for gases. Because gas molecules are in constant motion, their Compton profiles are difficult to measure precisely, and their precise CP values can only be obtained using very powerful radioactive sources like synchrotron.

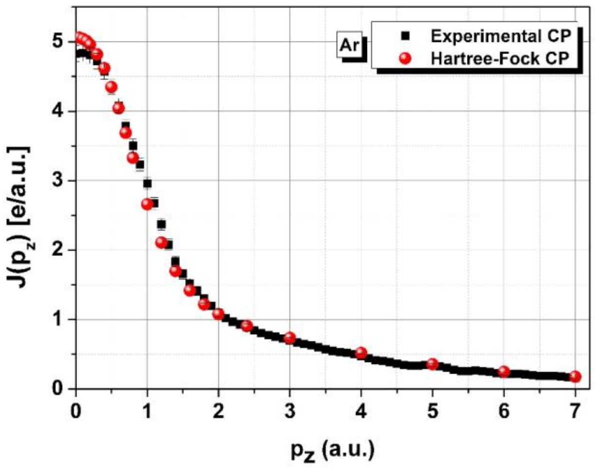

Figure 3. Experimental and theoretical Compton profile of the Ar

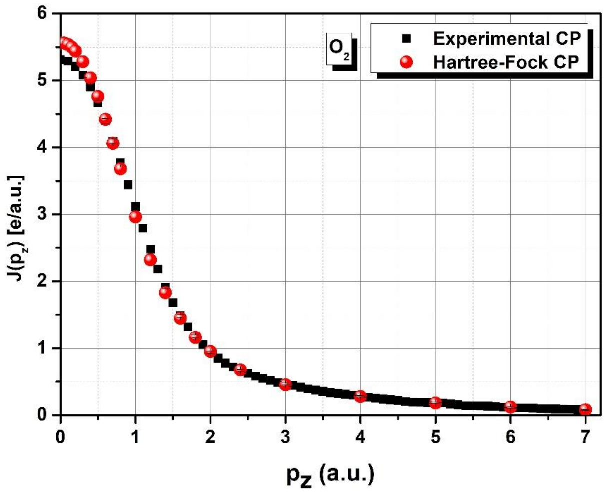

Figure 4. Experimental and theoretical Compton profile of the O_2

The relative comparisons of the experimental and theoretical results obtained were calculated with the following equation and are shown in Figs 5 and 6 for Ar and O_2 gases, respectively:

$$\Delta J(p_z) = \frac{J_x(p_z) - J(p_z)}{J(0)} \times 100 \qquad (8)$$

When Fig 5 is examined, at p_z = 0 a.u., there is a 4.51% difference between the experimental and theoretical CP values of Ar gas. It is clearly seen from this graph that this difference decreases gradually in parallel with increasing electron momentum values. As explained earlier, as pz increases, the contribution of core electrons in the CP spectrum increases, and since real wave functions of this electrons are very similar to free electron wave functions, their Compton profiles greatly match the results obtained with the HF method. Similar situation is valid for O_2 gas. However, the concordance of the experimental and theoretical results of the O_2 gas are in better than the results obtained for the Ar gas. As a matter of fact, after p_z = 1.6 a.u. for O_2 gas, the difference between theory and experiment decreased to less than 1%. The main reason that oxygen gives more harmonious results than argon gas can be thought to be its atomic number lower than Ar. Because, in the article Sakurai (2015) [10], it is mentioned that the effects of electron correlations effects should be taken into consideration in the relativistic calculations made for atoms with large atomic numbers or molecules containing these atoms.

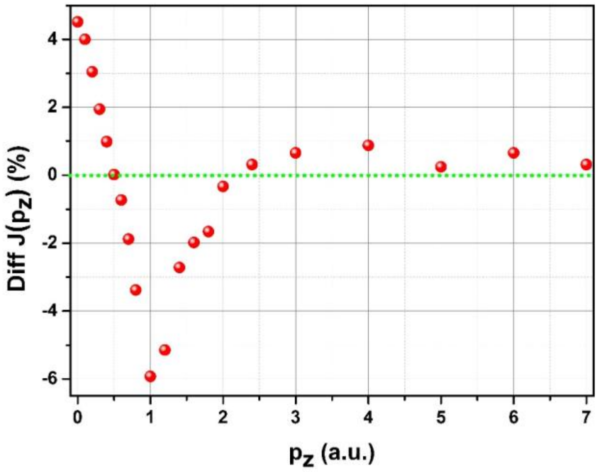

Figure 5. The relative diferences between experimental and theoretical Compton profile results of the Ar

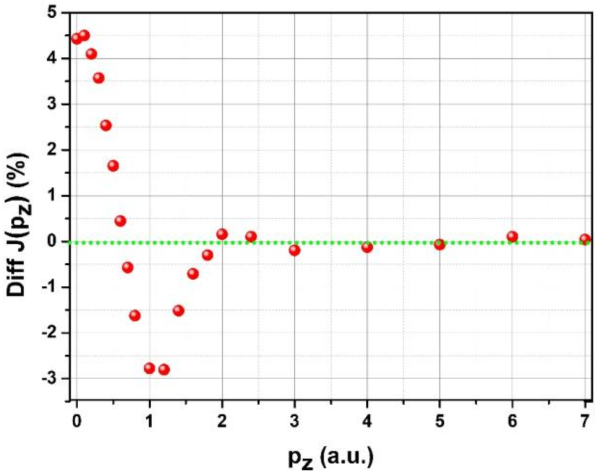

Figure 6. The relative diferences between experimental and theoretical Compton profile results of the O_2

It is known that accurate CP results can only be obtained using powerful radioactive sources in well-designed Compton spectrometers. The researchers lacking a synchrotron source perform their experiments at long measurement times to reach accurate experimental data in solid samples. However, the prolonged measurement time in gas samples increases the already existing stability problem. For this reason, the use of synchrotron sources is absolutely necessary in the Compton profile measurements of gas samples. Moreover, the experimental data obtained must be compared with the theoretical results calculated by DFT (density functional theory) methods, which include more detailed effects such as electron correlation, exchange and crystallization.

4. CONCLUSION

Compton profiles of Ar and O_2 gases were measured experimentally using 59.54 keV gamma photons emitted from the ^{241}Am source. A well-designed Compton spectrometer was used in experimental measurements. The obtained results are compared with the theoretical results obtained with the HF method. It has been determined that the experimental CP measurements of Ar and O_2 gases are in good agreement with the theoretical results calculated with HF, and the results of the O_2 gas are closer to the theoretical results. Finally, it has been mentioned that the importance of using synchrotron sources in CP experiments related gases and the necessity of making theoretical calculations with new and more accurate methods such as DFT.

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