# Generalized oscillator strength for the $3p^6 \rightarrow 3p^5(4s, 4s')$ transitions in argon

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Absolute values for the generalized oscillator strength (GOS) for the $3p^6 \rightarrow 3p^5(4s, 4s')$ transitions in argon ($^1S_0$) have been determined by 1-keV electron scattering. The relative differential cross section, measured in the angular range of 1.0°–15°, has been made absolute by comparison with known absolute values for the elastic differential cross section, and the results converted to GOS. The GOS's have also been calculated, using both the first Born and Glauber approximations, and the results are compared to the experimental values.

## I. INTRODUCTION

The minima and maxima in the generalized oscillator strength (GOS) for the transition from the ground state ($3p^6, {}^1S_0$) to the $3p^5(4s, 4s'), ({}^1P_1\,{}^3P_0, {}^3P_2\,{}^3P_1)$ excited states in the argon atom have been theoretically studied by several authors, using the first Born approximation.

Bonham[1] was the first to predict their existence, using numerical one-electron Hartree-Fock wave functions. This work was extended by Shimamura,[2] who used hydrogenlike wave functions to obtain analytical forms for the generalized oscillator strengths for some electronic excitations in Ar and other atoms. Ganas and Green,[3] using a simplified model, calculated generalized oscillator strengths for a single-particle excitations in Ne, Ar, Kr, and Xe. Their results, normalized to available optical oscillator strengths, showed a complex nodal structure in the generalized oscillator strengths when plotted as a function of the momentum transfer, $K$.

In the experimental side, the only known measurement of the generalized oscillator strength for the aforementioned transition in Ar covering the region of the first maximum was made by Wong *et al.*[4] at 25-keV incident electron energy. This work was primarily devoted to the determination of the positions of the extrema and only relative results were presented. It was also shown that the intensity value at the minima was not negligible as predicted by the first Born approximation.

Very recently, Li *et al.*,[5] working at 500-eV incident energy and with an improved energy resolution ($<0.1$ eV), measured the GOS for the $4s$ and $4s'$ transitions, but their measurements were restricted to a small angular range and did not cover the $K$ region where the first maximum and minimum appear.

Because of the intrinsic importance of maxima and minima in the GOS (Ref. 6) (which are related, for instance, to the nodal properties of the radial parts of the atomic and molecular orbital wave functions associated with the initial and final states), we have decided to determine absolute values for the GOS in a large-$K$ region, both experimentally and theoretically. For the latter, calculations have been made using the first Born and Glauber approximations and the results will be presented below, along with our experimental data.

## II. APPARATUS AND PROCEDURE

The apparatus used for the present measurements, shown schematically in Fig. 1, has been described before.[7] It consists basically of a rotatable electron gun, a neutral gas beam, and a Möllenstedt velocity analyzer fixed on the vacuum chamber wall. A crossed-beam geometry is used for the electron-gas collision. The electron beam, produced by a triode-type electron gun, has a full width at half maximum (fwhm) of approximately 0.6 eV and is used without prior energy selection. A typical beam current is 10 $\mu$A and the beam diameter, measured at the scattering region, is approximately 0.5 mm. The gas beam is produced by the expansion of a given sample inside the scattering chamber through a hypodermic needle with a 0.2-mm internal diameter and an aspect ratio (length/diameter) of 50.

The scattered electrons are velocity analyzed by the Möllensted analyzer and detected by an electron multiplier (Spiraltron, Galileo Electro Optics). The energy resolution of this system was set to 0.6 eV, as determined by the FWHM of the elastic peak.

A new version of the Möllenstedt analyzer has been

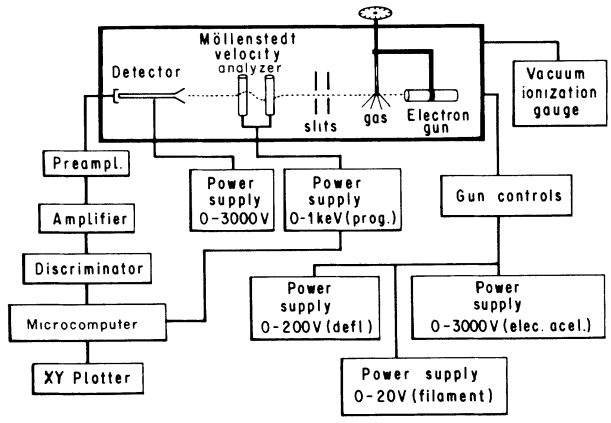


FIG. 1. Schematic arrangement of the electron energy-loss spectrometer.

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employed in the present work. In this new version the position of the two high voltage rods can be continuously and precisely adjusted from the outside by well-isolated micrometers. This allows for the selection, *in vacuo,* of different caustic positions (and hence for different energy resolutions). This analyzer, which also employs larger steelmade rods, has been utilized in the experimental study of the characteristics of the Möllenstedt analyzer.[8]

The analyzer viewing cone is determined by two parallel circular apertures of 200 and 50 $\mu$m diameter, located, respectively, at 120 and 150 mm from the scattering center. The apertures have been selected so that the acceptance cone includes as little in excess of the scattering volume as possible.

The stray magnetic fields in the plane of the measurements have been reduced to less than 10 mG in all directions by three pairs of square orthogonal Helmholtz coils.

The energy-loss spectra were measured in a signal-average mode, using an eight-bit microcomputer, which scanned a digitally programmable power supply (BERTAN, model 205-01R) and stored the acquired data.

The scattering angle zero was determined by measuring the scattering distribution over a 15° range on both the right- and left-hand side of the primary beam.

A very precise positioning mechanism allows the determination of the scattered angle with an accuracy of 0.02°. Nevertheless, the angular resolution, defined by the set of apertures in front of the Möllenstedt analyzer, is 0.2° in our experiment.

The vacuum was $1\times10^{-6}$ torr, without a gas beam and $4.5\times10^{-5}$ torr (maximum) when Ar was injected. Interatomic double scattering, viz., elastic scattering by a large angle, preceeded or followed by inelastic scattering by a small angle, has been shown to be a serious potential source of experimental uncertainty for large angle spectra,[9] and we have accordingly taken great care to ensure that our data was free from its contribution. Every spectrum has been measured at least at two different pressures. Only above 10° the ratio between the areas of the elastic and inelastic peaks changed significantly (20%), on passing from 3 to $5\times10^{-5}$ torr. The large angle data used in this work has been obtained at an average pressure of $3\times10^{-5}$ torr and is considered to be free of double (or multiple) scattering.

For each angle the residual gas contribution to the energy-loss spectrum was determined by repeating the experiment a second time. In this case the gas was admitted to the same working pressure ($3\times10^{-5}$ torr) but from a side flange located far from the scattering center. This background, which was always limited to less than 2% of the scattering signal, was then subtracted from the basic data.

The observed count rate was never allowed to exceed 20 kHz, in order to eliminate the need for dead time correction.

A further correction was necessary owing to the field-sweeping mode of the analyzer (Kollath correction);[10] the intensity of the spectrum was multiplied by $E_0/(E_0-E)$ ($E_0$, the primary beam energy and $E$, the excitation energy), a correction of 5% at the highest energy loss encountered.

The energy calibration was checked by measuring the energy-loss spectrum for helium. An excitation energy of 21.2 eV was found for the $1s\rightarrow2p$ line, in good agreement with the known value.

The uncertainties are estimated in the following manner. The maximum statistical uncertainty was 3%, as at least 1300 counts were accumulated at the maximum of the inelastic profile for each scattering angle. Fluctuations in the primary beam current and in the sample pressure were of the order of 1% and 0.5%, respectively. The main source of error in the determination of the scattering intensity was the limited angular resolution (0.2%), which contributes with an uncertainty of approximately 20% below 4.5° and 14% above, due to the strong angular dependence of the cross section. The area of the $4s,4s'$ peak, determined by a Gaussian fitting, is subject to an uncertainty which we estimate to be of the order of 10% for scattering angles above 4.5°.

An additional source of error comes from the quoted uncertainty (6.5%) in the absolute elastic cross section which was used in the normalized procedure, described in Sec. III.

The overall uncertainty $\delta$ is defined as

$$\delta=\left[\sum_i \delta_i^2\right]^{1/2},$$

and is equal to approximately 21% below 4.5° and to 18% above this angle

## III. RESULTS

Energy-loss spectra have been obtained in the 1.0°–15° angular range and submitted to background subtraction and to the Kollath correction. Figure 2 shows the resulting data for $\theta=1.0°$ and 4.0°. Features I and III refer to dipole-allowed transitions from the ground state mainly to the $4s,4s'$ (11.8 eV) and to the $5s,5s'$ and $3d$ (14.2 eV) excited states. Feature II is associated to nonallowed transitions to the $4p,4p'$ (13.4 eV) final states.[11] Several autoionizing structures can be clearly seen around 27

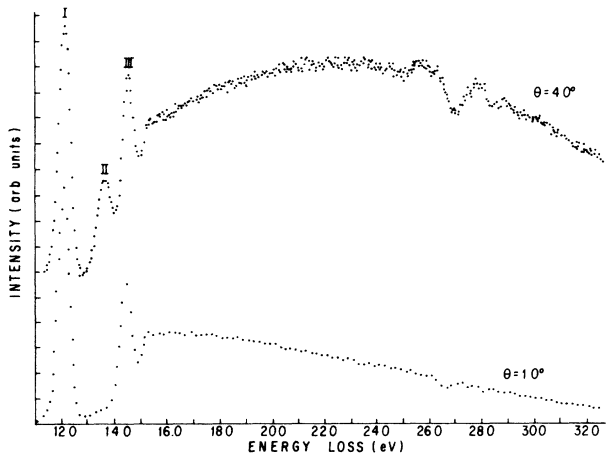


FIG. 2. Energy-loss spectra of argon, measured at 1-keV incident energy and $\theta=1.0°$ and 4.0°.

eV.[12,13] Although peak I is well separated from the other peaks in our spectra, the experimental energy resolution is not enough to distinguish between the $4s$ and $4s'$ contributions. The ratio between the areas of the $4s,4s'$ and elastic peaks was obtained by fitting the two peaks with Gaussian functions; the intensity of the former peak was then normalized to the known absolute values for the elastic differential cross section.[14]

The inelastic differential cross sections were converted into generalized oscillator strength by the well-known formula[15]

$$f_{on}(K)=\frac{k_0}{k_n}\frac{E}{2}K^2\frac{d\sigma_{on}}{d\Omega}\ , \tag{1}$$

where $k_0$ and $k_n$ are the modules of the momenta associated with the incident and scattered electron, $E$ is the excitation energy, $d\sigma_{on}/d\Omega$ represents the differential cross section for the excitation from the state $o$ to state $n$ and $K$ is the module of the momentum transfer vector, $\mathbf{K}=\mathbf{k}_0-\mathbf{k}_n$.

The absolute GOS's are shown in Fig. 3, along with the experimental results of Wong *et al.*[4] (normalized to our results at $K^2=1.25$), Li *et al.*,[5] and the first Born approximation (FBA) results. Our experimental results are also compared to the theoretical results obtained through the Glauber approximation in Fig. 4.

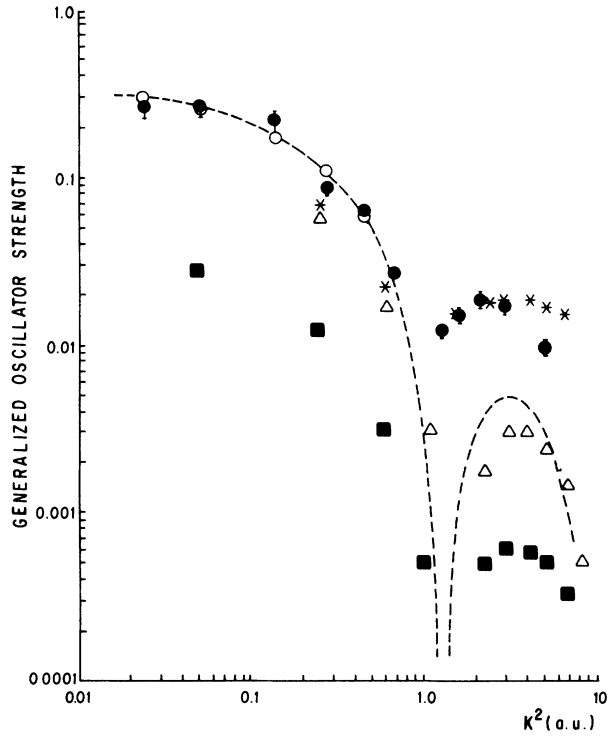


FIG. 3. Absolute GOS for the $3p^6\rightarrow 3p^5(4s,4s')$ transition in the argon atom. ●, experimental this work; ○, experimental Li *et al.* (Ref. 5); *, experimental, Wong *et al.* (Ref. 4); — — —, FBA, this work; ■, FBA, Bonham (Ref. 1); △, FBA, Shimamura (Ref. 2).

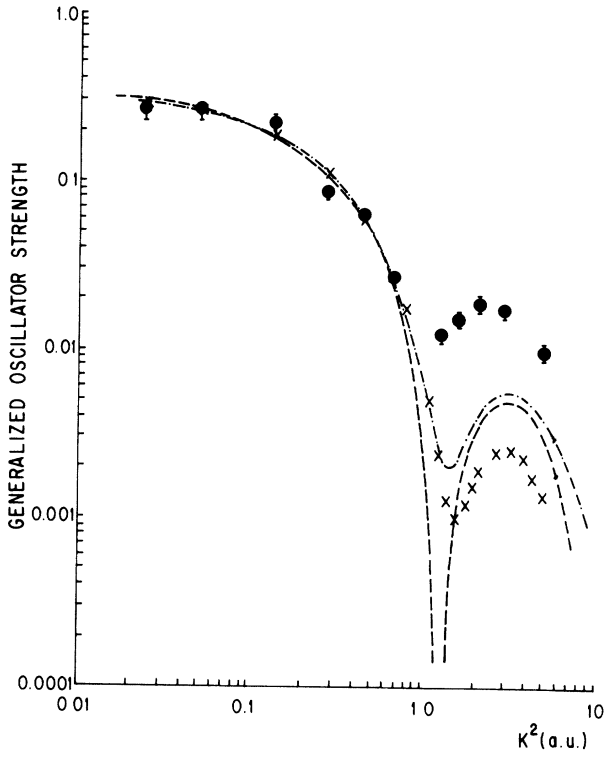


FIG. 4. Absolute GOS for the $3p^6\rightarrow 3p^5(4s,4s')$ transitions in the argon atom (all values from this work). ●, experimental; —·—·—·, Glauber YFC; x, Glauber IFC; — — —, FBA.

## IV. DISCUSSION

The renormalized experimental results of Wong *et al.*[4] are seen to be in reasonable agreement with the present results only above $K^2=1$. The former work aimed mainly on the determination of the position of maxima and minima; even the contribution of background scattering, which was not subtracted from their raw data, could not affect significantly the intensity values and explain the observed discrepancy. Both data show, though, a reasonable agreement on the position of maximum and minimum and on the non-neglectible intensity for the latter. The recent experimental results of Li *et al.*[5] shows a good overall agreement with the present results.

For the sake of clarity we will now discuss separately the first Born and Glauber approximation results.

### A. First Born approximation calculations

Self-consistent wave functions were constructed from Gaussian orbitals adopting the $L$-$S$ coupling scheme. No exchange terms between the incident and atomic electrons were considered, but their contributions are considered to be small at the impact energy and $K^2$ values range considered in this work. Indeed, for $K^2=0.8$ a.u. the Bonham-Ochkur approximation predicts that the exchange term will contribute with less than 1% to the scattered intensity, as compared to the direct term. For

$K^2=10$ a.u. this contribution raised to about 13%. Considering that the experimental data deviates from the theoretical results by more than one order of magnitude for large $K^2$ values, it seems safe to neglect the exchange-term contribution in the present analysis.

It is known[16] that spin-orbit effects are important in the description of these excited states, coupling the states of total angular momentum $J=1$. This coupling can be considered through the semiempirical method of Cowan[17] and Andrew and Cowan,[18] in which the wave functions belonging to the $4s'[\frac{1}{2}]^0_1$ and $4s[\frac{3}{2}]^0_1$ levels are obtained as linear combinations of the wave functions of the $[(3p)^5 4s]4\,^1P_1$ and $[(3p)^5 4s]4\,^3P_1$ configurations:

$$|\,4s'[\tfrac{1}{2}]^0_1;M_j\rangle = b\,|\,[(3p)^5 4s]4\,^1P_1;M_j\rangle + a\,|\,[(3p)^5 4s]4\,^3P_1;M_j\rangle\ , \tag{2a}$$

$$|\,4s[\tfrac{3}{2}]^0_1;M_j\rangle = -a\,|\,[(3p)^5 4s]4\,^1P_1;M_j\rangle + b\,|\,[(3p)^5 4s]4\,^3P_1;M_j\rangle\ , \tag{2b}$$

where $a=-0.450$ and $b=0.893$, these coefficients being determined from the experimentally observed levels of $4s$ and $4s'$ states.

Neglecting exchange between the incident and the atomic electrons, the contribution to the GOS from the triplet states in the above expression is null, that is, only the first term in the right-hand side of the above expressions contribute to the GOS.

Within our experimental resolution these levels are not resolved and so in our calculations we summed up the contributions of the two $J=1$ levels. This is equivalent to considering just the $[(3p)^5 4s]4\,^1P_1$ configuration when one neglects the exchange between the incident and the atomic electrons.

At this point an interesting consideration can be made based on the recent GOS results of Li *et al.*[5] Their experimental results separate, for a restricted $K^2$ region, the contributions of the $4s'$ and $4s$ levels, related, respectively, to the $^3P_0,{}^1P_1$ and $^3P_2,{}^3P_1$ states. If the assumption that the exchange between the incident and atomic electrons is neglectible is correct, only the first term in the right-hand side of expression (2) would contribute to the GOS, (2a) related to the $4s'$ peak and (2b) to the $4s$ peak. In this case, using expression (2) and neglecting exchange, the relation between the GOS for the $4s'$ and $4s$ peaks should be equal to $b^2/a^2=3.93$. Indeed from their figure a constant relation between the GOS for the $4s'$ and $4s$ peaks of about 3.9 is found.

Coming back to the calculations, we have found that the addition of three diffuse $s$-type functions to the $13s/10p$ basis of Gianolia *et al.*[19] was necessary in order to get good values for the excitation energy and for the optical oscillator strength.

The agreement with the results of Ganas and Green,[3] normalized to the experimental GOS, is excellent, but both results disagree with the experimental values above $K^2=1.0$. The results of Shimamura,[2] based on hydrogenlike wave functions and the results of Bonham,[1] based on numerical Hartree-Fock wave functions, show only a qualitative agreement with the experimental values in the whole measured $K^2$ range.

A similar behavior has been previously obtained by Kim *et al.*[6] on their work on the GOS, for the electronic excitation of the Xe atom from the ground to the $5p^5 6s(^2P_{3/2})$ state; their FBA results also fall much lower than the experimental data for large $K^2$ values.

The poor quantitative agreement observed between the theoretical (FBA) and our experimental results above $K^2=1.0$ could be due to the following reasons.

(i) The first Born approximation does not take into account the interaction between the incident electron and the atomic nucleus.

(ii) All the calculations assume a "frozen" core, which implies that the interaction of the incident electron with the atomic electrons which do not participate directly in the excitation process is not taken into account.

(iii) Contributions from higher-order terms in the Born series may be important to fully describe the scattering process.

These three factors are thought to have a growing importance as $K$ increases; this can be rationalized considering that classically this situation corresponds to a smaller impact parameter and hence to a stronger interaction between the incident electron and the atom as a whole.

### B. Glauber approximation calculations

The calculations have been done in two levels.[20] Firstly, the incident electron was assumed to interact only with the atomic electron that participates directly in the excitation process and with one nuclear charge ["inert frozen core" (IFC) calculation].

The second level of calculation ["Yukawa frozen core" (YFC) calculation] also takes into account the interaction of the incident electron with the $(N-1)$ nuclear charges and with the atomic electrons which do not participate directly in the excitation process. This is done by taking a Yukawa potential[21] whose parameters were obtained by a fitting to a numerical potential, determined through a Hartree-Fock-$X\alpha$ method. In order to generate this numerical potential, an atomic calculation of the $(3p)^5 4s\,^1P_1$ state was initially performed. The potential was then obtained from the orbitals associated with the $1s^2 2s^2 2p^6 3s^2 3p^5$ electronic configuration, and 17 nuclear charges.

In both calculations "single-zeta" (SZ) and "double-zeta" (DZ) basis sets were determined for the excited state by a Hartree-Fock-Roothaan method, using Slater-type orbitals. For the ground state, SZ, DZ, and other expanded basis sets determined by Clementi and Roette[22] have been used.

The $L$-$S$ coupling scheme has been assumed to describe the atomic wave functions and, as in the FBA calculation, no corrections have been included for exchange processes. The GOS was determined from the differential cross section, through expression (1).

We can see in Fig. 3 that the Glauber and Born results show a similar behavior. They both present a good quantitative agreement with the experimental results for small-$K^2$ values ($K^2<0.5$ a.u.). For larger-$K$ values, the agreement is only qualitative.

The first minimum of the GOS is more correctly described by the Glauber calculations as compared to the FBA results, showing that the former offers a better description of the collision process.

Comparing now the Glauber YFC to the IFC results one sees, consistently, that the interaction of the incident electrons with the core electron affects significantly the results only for larger-$K$ values. But even the Glauber YFC results do not agree quantitatively with the experimental results for larger-$K$ values. This cannot be attributed to the small scattering angle restriction associated with the Glauber approximation,[23,24] since our measurements have been restricted to a small scattering angle range (1.0°–15°).

## V. CONCLUSIONS

Absolute values for the generalized oscillator strength for the $3p^6 \rightarrow 3p^5(4s,4s')$ transitions in the argon atom ($^1S_0$) have been determined, both experimentally and theoretically.

The experimental results have been obtained from relative inelastic differential cross sections, normalized to known absolute values for the elastic differential cross section.

The theoretical results have been obtained using both the first Born and Glauber approximations. In the first case, Gaussian atomic orbitals were used; a value for the optical oscillator strength in good agreement with the known experimental value was obtained, and a good agreement was found with the experimental GOS values for small values of the square of the momentum transfer, $K^2$ ($K^2 < 1.0$).

The Glauber approximation results were obtained both considering the interaction of the incident electrons with the atomic electrons not directly involved in the excitation process and the $(N-1)$ nuclear charges (YFC calculations) and by neglecting this interaction (IFC calculations). In both models a good value for the optical oscillator strength was found as well as a good agreement with the experimental GOS for small-$K^2$ values.

The first Born and Glauber approximations correctly predict the positions of the observed maximum and minimum in the GOS curve as a function of $K^2$.

The Glauber approximation calculations also predict a non-negligible intensity for the minimum, as experimentally observed.

Both approximations fail to quantitatively reproduce the experimental results for $K^2 > 1$, showing that in this range a more realistic description of the collision process must be taken into account.

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