The valence shell excitations for some noble atoms studied by fast electron impact

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Abstract.
The differential cross sections and apparent generalized oscillator strengths for the valence-shell excitations of the noble atoms of He, Ne, Ar, and Kr are reviewed. The asymptotical behaviors of the apparent generalized oscillator strengths show that the first Born approximation is more easily reached for the dipole-allowed transition than the dipole-forbidden one of \( ^1S_0 \rightarrow ^1S_0 \) type, in which the final state has the same term symbol of the initial state. Furthermore, the experiments reveal that the generalized oscillator strengths for the transitions, which are formed by the same electronic configuration through the intrachannel interaction such as \((n-1)p^5ns[3/2]_1\) and \((n-1)p^5ns'[1/2]_1\) of Ne (n=3), Ar (n=4), and Kr (n=5), are nearly parallel. This phenomenon is elucidated by the fact that the \(LS\) coupling singlet nature of the ground state selects the \(LS\) coupling singlet component from the excited wave function, i.e., the contribution from the singlet component in the excitation is dominant in the generalized oscillator strength.

1. Introduction
The cross sections of electron impact excitation are important in the diverse fields such as radiation physics, plasma physics, atmospheric physics, and astrophysics, and there are ever-increasing needs for such cross-section data. For fast electron impact, the first Born approximation (FBA) is applicable if the incident electron energy is high enough but still nonrelativistic. The differential cross section (DCS) can be factorized into two factors: one dealing with the incident particle only, which is a trivial factor; the other dealing with the target only, which is the so-called generalized oscillator strength (GOS) [1, 2]. The GOS is a fundamental quantity for atoms and molecules since it relates to various physical constants and is very sensitive to the structure of the target. Experimentally, the GOS can be obtained directly from the DCS measured by the electron scattering experiment when the FBA condition is satisfied. However, for the slow electron impact, the FBA condition is not satisfied, then the GOS can not be derived from the experimental DCS and the influence of the incident electron should be considered. Therefore, the investigations of the GOS, as well as the condition of the FBA, is an important content for atomic and molecular physics [2].

In this paper the DCSs and apparent GOSs for the valence-shell excitations of He, Ne, Ar, and Kr measured at the higher impact energies (\( \geq 100\text{eV} \)) are reviewed. For helium, it is the simplest multi-electron system, and its experimental DCSs and GOSs can test the theoretical method and the theoretical code stringently. Therefore, its DCSs and GOSs were
extensively investigated both experimentally [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13] and theoretically [14, 15, 16]. Among them, Trajmar and his collaborators [3, 4] have measured the DCSs of electron-impact excitation of helium at the incident electron energies lower than 100 eV, while Dillon and Lassettre [5, 6] and Suzuki and his coworkers [7, 8, 9] measured the DCSs of the $^1S_0$, $^1P_1$, and $^3S_1$ states at moderate kinetic energies of 100-700 eV. In our group, we measured the DCSs and GOSs of the excitations of helium at much higher impact energies of 1500 eV and 2500 eV [10, 11, 12, 13]. Recently, the GOSs of the $^1S_0$ and $^1P_1$ states of helium were measured by me and my collaborators using the inelastic X-ray scattering (IXS) [17]. The theoretical calculations were carried out by the R-matrix theory [14], the first Born approximation [15], and the convergent close-coupling theory [16]. Furthermore, it should be mentioned that the DCSs and GOSs for the excitations of helium have been measured and calculated with a high accuracy [12, 14, 15, 18], so the reliable DCSs and GOSs of He are often used as the standard data to normalize the DCSs and GOSs for other atoms and molecules [18, 19].

For the noble atoms of Ne, Ar and Kr, their DCSs and GOSs were measured by Refs. [9, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30]. Among them, Khakoo and his collaborators [20, 21] reported the DCSs of electron-impact excitation of Ne and Ar at the incident electron energies lower than 100 eV, while Suzuki and his coworkers [9, 22, 23, 24] measured the DCSs of the valence-shell excitations of Ne, Ar and Kr at moderate kinetic energies of 100-500 eV. In our group, we measured the DCSs and GOSs for the valence-shell excitations of Ne, Ar and Kr at the high impact energies of 1500 eV and 2500 eV [19, 25, 26, 27, 29, 30]. As for the theoretical studies, the DCSs and GOSs were calculated by the random phase approximation with exchange (RPAE) methods [31, 32, 33]. Since the excited energy level structures of Ne, Ar and Kr are well described by the intermediate coupling, while their ground states are closed-shell $^1S_0$ states of LS coupling, these noble atoms are the typical systems to study the valence-shell excitation mechanism and dynamics relating to two different coupling scheme, which were explored by our group recently [19, 26, 34].

In this paper, the DCSs and GOSs for electron impact excitation of the valence-shell states of He, Ne, Ar and Kr at the moderate and high impact energies are reviewed. In the following sections, the theoretical background will be given in Sec. 2. Then, the apparent GOSs of helium will be reviewed in Sec. 3, and the momentum transfer dependence behaviors for Ne, Ar and Kr will be discussed in Sec. 4. Finally some conclusions will be included in Sec. 5.

2. Theoretical background

When the incident electron is sufficiently fast, i.e., the first Born approximation is valid and the incident and scattered electrons can be treated as plan waves, the differential cross section $d\sigma_n^e/d\Omega$ of a state $n$ of an atom or molecule excited by the fast electron impact can be described by [1, 2] (in atomic unit):

$$
\frac{d\sigma_n^e}{d\Omega} = \frac{2}{E_n p_0 K^2} f(E_n, K^2),
$$

(1)

where $f(E_n, K^2)$ is the generalized oscillator strength defined as

$$
f(E_n, K^2) = \frac{2E_n}{K^2} \left| \langle \Psi_n | \sum_{j=1}^N e^{iK \cdot r_j} | \Psi_0 \rangle \right|^2.
$$

(2)

Here $E_n$ and $K$ are the excitation energy and the momentum transfer, while $p_0$ and $p_a$ stand for the incident and scattered electron momenta, respectively. $\Psi_0$ and $\Psi_n$ are $N$ electrons wave functions for the initial and final states, respectively. $r_j$ is the position vector of the $j$th electron. It can be seen that for sufficiently fast electron impact, the influence of the incident
particle upon an atom or molecule can be regarded as a sudden and small external perturbation, and the exchange effect is negligibly small [1, 2]. Differently, for the slow electron impact, the collision problem is considered as a combined system of the incident particle and the target, in which both of them lose their mechanical individuality. Even so, the DCSs measured at the intermediate impact energies can be converted into the oscillator strengths, i.e., the so-called apparent generalized oscillator strength (AGOS) \( f_A(E_0, E_n, K^2) \), with a similar form as Eq. (1):

\[
f^A(E_0, E_n, K^2) = \frac{E_n p_0}{2 p_a} K^2 \frac{d\sigma^p_\omega}{d\Omega}.
\]  

With the increasing of the incident electron energies, the measured apparent GOSs should approach to the same curve, which means that the FBA is valid and the measured apparent GOSs approach to GOS:

\[
f^A(E_0, E_n, K^2)|_{E_0 \to \infty} = f(E_n, K^2).
\]  

Then the measured GOS of \( f(E_n, K^2) \), which is free of the influence of the incident electron, reveals the momentum distribution character related to the wave functions of the initial and final states (\( |\Psi_0\rangle \) and \( |\Psi_n\rangle \)), and it provides the benchmark to testify the theoretically calculated wave functions [2, 14].

Another experimental method to explore the character of the wave functions of the initial and final states is the Compton excitation, in which the DCSs of the excited states are measured through the inelastic X-ray scattering or the so-called nonresonant X-ray Raman scattering [35, 36, 37]. The differential cross section \( d\sigma^r_n/d\Omega \) of the Compton excitation can be described by:

\[
d\sigma^r_n/d\Omega = r_0^2 \frac{\omega_f}{\omega_i} |\vec{e}_i \cdot \vec{e}_f^*|^2 S(\vec{q}, \omega),
\]  

where \( S(\vec{q}, \omega) \) is the dynamical structure factor defined as

\[
S(\vec{q}, \omega) = \left| \langle \Psi_n | \sum_{j=1}^{N} e^{i\vec{q} \cdot \vec{r}_j} |\Psi_0\rangle \right|^2.
\]  

Here \( r_0 \) is the classic electron radius, while \( \hbar \omega_i \) and \( \hbar \omega_f \) are the energies of incident and scattered X-ray, respectively. \( \hbar \omega = \hbar \omega_i - \hbar \omega_f \) is the energy loss, and \( \vec{q} \) is the momentum transfer (which is called \( \vec{K} \) in the electron impact experiment). \( \vec{e}_i \) and \( \vec{e}_f \) stand for the polarization directions for the incident and scattered photons.

Considering \( E_n = \hbar \omega \) and \( \vec{K} = \vec{q} \), the GOS defined in Eq. (2) can be rewritten as (in atomic unit):

\[
f(\omega, q^2) = \frac{2\omega}{q^2} S(\vec{q}, \omega) = \frac{1}{r_0^2} \frac{\omega_i}{\omega_f} \frac{2\omega}{q^2} \frac{1}{|\vec{e}_i \cdot \vec{e}_f|^2} \frac{d\sigma^r_n}{d\Omega}.
\]  

It can be seen from Eqs. (1-2) and (5-7) that there is an intrinsic connection between the fast electron scattering and the inelastic X-ray scattering. Therefore, we can convert the DCS measured by IXS into GOS, which provide a new and independent method to determine the GOS, and the GOSs measured by these two methods can provide independent cross-check. The merit of the IXS method is that the FBA is always valid, which avoids the tedious procedures to judge the validity of FBA on a trial-and-error basis in the electron impact method. The demerit of the IXS method is the low cross section of the Compton excitation.
3. Generalized oscillator strengths for He

The DCSs for the excitations of $2^1S_0$ and $2^1P_1$ of He were extensively investigated both experimentally [3, 5, 7, 9, 10, 11, 12] and theoretically [14, 15]. Herein only the recent apparent GOSs of $2^1S_0$ and $2^1P_1$ measured at the incident electron energies from 100 eV to 2500 eV are shown in Fig. 1 and 2 along with the theoretical GOSs calculated within the FBA [15], which are in a good agreement with other calculations [14]. It can be seen from Fig. 1 and 2 that the apparent GOSs for the both excitations approach to the GOSs with the increasing of the impact energies, which is the typical behaviors of apparent GOS. It can also be seen that for the dipole-allowed transition of $2^1P_1$ the apparent GOSs in the low $K^2$ region are very close to the GOSs even for the impact energy as low as 200 eV, while for the dipole-forbidden transition of $2^1S_0$ the apparent GOSs are systematically lower than the GOSs even for the impact energy as high as 1500 eV. These results mean that the FBA is more easily reached for the dipole-allowed transition than the dipole-forbidden one of $1S_0 \rightarrow S_0$ type. The above phenomenon can be elucidated by that the higher order Born amplitudes give the main contribution to the apparent GOS for the transition in which the terms of the initial and final states are the same, as the case of the $1S_0 \rightarrow S_0$ transition [7, 9].

Fig. 2 also shows the GOSs measured by IXS [17]. Because the FBA is always valid for IXS method, the results measured by IXS can serve for the standard GOS data. The situation is true because the GOSs measured by IXS show a very good agreement with the theoretical calculations [14, 15] and the results measured by a high energy electron impact [10]. Therefore the work of Xie et al [17] shows that the IXS is a new and independent method to determine the GOS at a third generation synchrotron. The independent cross-check shows that the theoretical calculations [14, 15] are reliable and the FBA holds for the measured $K^2$ region of 0-4.4 a.u. of the $2^1P_1$ transition at an impact energy of 1500 eV.
The apparent GOSs measured at different incident electron energies are in general agreement, while the small GOSs for the excitations of $3\,^2\!p$ energies of 300, 400 and 500 eV measured by Suzuki that the FBA is more difficult to reach for the dipole-forbidden excitation of $3\,^2\!p$. The experimental results were taken from Cheng et al [19] and Suzuki et al [22].

The apparent GOSs for the valence-shell excitations of Ne were measured by Refs. [19, 20, 22] and calculated by Refs. [31, 32, 33]. Fig. 3 shows the measured apparent GOSs for the dipole-forbidden excitation of Ne. It can be seen that the apparent GOSs measured at a much higher impact energy of 2500 eV [19] are systemically higher than the ones of Suzuki et al [22] measured at the impact energies of 300, 400 and 500 eV. The discrepancies may be due to the different normalization procedures: Suzuki et al [22] normalized their data on the elastic DCs of Ne while Cheng et al [19] normalized their data on the absolute GOSs of $2\,^4\!P_1$ of He calculated by Cann and Thakkar [15], which are in good agreement of the experimental results as shown in Section 3. Furthermore, the theoretical calculation by the random phase approximation with exchange (RPAE) methods [32, 33] supports the results measured at 2500 eV. Whereas, if we exclude the possible normalization problems, the FBA is valid at the lower $K^2$ region when the impact energy is larger than 300 eV since the apparent GOSs at the impact energies of 300, 400 and 500 eV measured by Suzuki et al [22] are in the same curve. The apparent GOSs for the dipole-forbidden excitation of $3\,^2\!p[1/2]_0$ of Ne are shown in Fig. 4, and it can be seen that there are apparent discrepancies including the shapes and the extremal positions between the apparent GOSs measured at the different impact energies, which means that the FBA is more difficult to reach for the dipole-forbidden excitation of $3\,^2\!p[1/2]_0$ than the dipole-allowed ones of $2\,^5\!s[3/2]$, and $2\,^5\!s'[1/2]$. This phenomenon is similar with the one of He discussed above, since the $2\,^5\!p[1/2]_0$ state in Ne is well represented by the LS term $^1S_0$, which is the same as the term $^1S_0$ of the ground state of $2\,^5\!p$ [9, 19]. The RPAE calculation [31] is in general agreement with the experimental one measured at 2500 eV. Therefore, we have the reason to believe that the FBA holds at an impact energy of 2500 eV for the valence excitations of Ne.

The apparent GOSs for the valence-shell excitations of Ar were measured by Refs. [21, 23, 25, 26, 27, 28] and calculated by Refs. [31, 32]. Fig. 5 shows the measured apparent GOSs for the excitations of $3\,^5\!d[3/2]_1$ and $3\,^5\!d'[1/2]_1$ of Ar [23, 25, 26]. The apparent GOSs measured at different incident electron energies are in general agreement, while the small
The experimental results were taken from Zhu et al [26], Ji et al [25] and Li et al [23].

differences for the large $K^2$ region may be attributed to the fact that the FBA does not hold in this $K^2$ region. The total apparent GOSs of $3p^64s[3/2]_1$ and $3p^64s'[1/2]_1$ measured by Zhu et al [26] show a good agreement with the ones of Fan and Leung [28], in which the different normalization procedure of the TRK sum rule is used. The shape and extremal positions of the GOSs calculated by RPAE [32] are in a good agreement with the experimental results of Zhu et al, so we have the confidence to believe that the FBA is satisfied at an impact energy of 2500 eV in the measured $K^2$ region.

The apparent GOSs for the valence-shell excitations of Kr were measured by Refs. [9, 24, 29] and calculated with RPAE by Refs. [31, 32]. Fig. 6 shows the measured apparent GOSs for the excitations of $4p^55s[3/2]_1$ and $4p^55s'[1/2]_1$ of Kr [29, 24]. Although the apparent GOSs measured at an impact energy of 2500 eV [29] are apparently different from the ones determined at 500 eV [24], the former is more reliable and satisfies the FBA since it is measured at a much higher impact energy and perfectly matches the theoretical calculation based on RPAE [29, 32, 33].

4.2. Generalized oscillator strength ratios of Ne, Ar and Kr

It can be seen from Fig. 3, 5 and 6 that the GOS curves of $(n-1)p^5ns[3/2]_1$ and $(n-1)p^5ns'[1/2]_1$ of Ne (n=3), Ar (n=4), and Kr (n=5) measured at 2500 eV are nearly parallel. Considering the simpleness of the physical picture of the FBA for the fast electron impact excitation, this common phenomenon for Ne, Ar and Kr is strange because the wave functions for the two excited states of every atom are different although its wave function for the ground state is same. In order to elucidate the phenomenon, the wave functions of the relevant states of Ne, Ar and Kr were calculated using Cowan code [19, 26, 34]. Then the intermediate coupling wave functions for the excited states can be expanded by the linear combination of the basis of the $LS$ coupling wave function:

$$\Psi_{ns[3/2]_1} = \alpha_n \Psi_{ns^3P_1} + \beta_n \Psi_{ns^1P_1}, \quad (8)$$

$$\Psi_{ns'[1/2]_1} = -\beta_n \Psi_{ns^3P_1} + \alpha_n \Psi_{ns^1P_1}. \quad (9)$$

Here $\Psi_{ns[3/2]_1}$ and $\Psi_{ns'[1/2]_1}$ are the wave functions of the excited states of $ns[3/2]_1$ and $ns'[1/2]_1$ in the intermediate coupling scheme, and $\Psi_{ns^3P_1}$ and $\Psi_{ns^1P_1}$ are the wave function basis in the $LS$ coupling scheme while $\alpha_n$ and $\beta_n$ are the corresponding intermediate coupling coefficients.
from the intermediate coupling wave functions of the excited states, and the
Ar (n=4), and Kr (n=5) shown in Figs. 3, 5 and 6 can be easily elucidated: the
Now the parallel character of the GOSs of (\(n=3\)) \[19\]; (b) Ar (n=4) \[26\]; and (c) Kr
\(n\) electron of \(f\)

The experimental and calculated GOS ratios between the excitations of \((n - 1)p^{2}ns[3/2]_1\) and \((n - 1)p^{2}ns'[1/2]_1\) of (a) Ne
\(n=3\) \[19\]; (b) Ar (n=4) \[26\]; and (c) Kr
\(n\) [34].

Since the spin-forbidden transition is negligibly small for fast electron impact \[2\] and the initial
ground state is singlet, the contribution of the triplet component in the excitation to GOS is
negligibly small, i.e., \(< \Psi_{ns^3P_1} | \sum_{j=1}^{N} e^{iK\cdot r_j} | \Psi_{(n-1)p^{6}S_0} > = 0\). Since the momentum transfers at
the same scattering angle are nearly equal for the two transitions, the GOS ratio \(\delta\) can be
obtained according to the definition of GOS:

\[
\delta = \frac{f_{ns[3/2]_1}}{f_{ns'[1/2]_1}} = \frac{E_1}{E_2} \left| \frac{< \beta_n \Psi_{ns^3P_1} | \sum_{j=1}^{N} e^{iK\cdot r_j} | \Psi_{(n-1)p^{6}S_0} >}{< \alpha_n \Psi_{ns^3P_1} | \sum_{j=1}^{N} e^{iK\cdot r_j} | \Psi_{(n-1)p^{6}S_0} >} \right|^2
\]

\[
= \frac{E_1}{E_2} \frac{\beta_n^2}{\alpha_n^2}, \tag{10}
\]

where \(f_{ns[3/2]_1}, E_1\) and \(p_1\) are the GOS, excitation energy and the momentum of the scattered
electron of \(ns[3/2]_1\), while \(f_{ns'[1/2]_1}, E_2\) and \(p_2\) are the corresponding quantities of \(ns[3/2]_1\),
respectively.

From Eq. (10) it can be seen that the GOS ratio for the pair of transitions, which are formed
by the same electronic configuration through the intrachannel interaction, should be a constant.
Now the parallel character of the GOSs of \((n - 1)p^{2}ns[3/2]_1\) and \((n - 1)p^{2}ns'[1/2]_1\) of Ne \((n=3)\),
Ar \((n=4)\), and Kr \((n=5)\) shown in Figs. 3, 5 and 6 can be easily elucidated: the \(LS\) coupling
singlet nature of the ground states of noble atoms selects the \(LS\) coupling singlet components
from the intermediate coupling wave functions of the excited states, and the \(LS\) coupling singlet

Figure 8. The experimental GOS ratios between the inner excitations of \(1s2s(^3S_1)2p\)
\(2P\) and \(1s2s(^1S_0)2p\) \(2P\) of lithium [38].
components are enough to describe the excitation process from the ground state for fast electron impact. The quantitative GOS ratios for \((n - 1)p^5ns\[3/2\]_1\) and \((n - 1)p^5ns'[1/2]_1\) of Ne \((n=3)\), Ar \((n=4)\), and Kr \((n=5)\) are shown in Fig. 7, and it can be seen that the agreement between the experimental results and the theoretical ones are satisfactory.

Recently, the GOS ratios for the higher dipole-allowed excitations of Ne [19], the dipole-forbidden excitations of Kr [34], as well as the inner shell excitations of Li [38] shown in Fig.8, were measured. All these results [19, 34, 38] show that the GOSs ratios for the pair of transitions formed by the same electronic configuration through the intrachannel interaction are constant, which is consistent with the above theoretical analysis. Furthermore, since the photoexcitation and photoionization are the electric dipole processes, which correspond to the excitation and ionization for fast electron impact at optical limit \((K^2 \rightarrow 0)\), the 4-parameter model, which depends on the validity of \(LS\) coupling, suggested by Snell et al [39] is sufficient to describe the Xe 4d photoionization process.

5. Conclusion
The DCSs and apparent GOSs for the valence-shell excitations of the noble atoms of He, Ne, Ar, and Kr measured by the intermediate and high energy impact are reviewed, and the conditions that the FBA holds for the excitations are discussed. The asymptotical behaviors of the apparent GOSs of He and Ne show that the FBA is more easily reached for the dipole-allowed transition than the dipole-forbidden one of \(^1S_0 \rightarrow ^1S_0\) type, in which the final state has the same term symbol of the initial state. From the measured GOS curves of \((n - 1)p^5ns\[3/2\]_1\) and \((n - 1)p^5ns'[1/2]_1\) of Ne \((n=3)\), Ar \((n=4)\), and Kr \((n=5)\), the parallel character of the pair transitions was observed. With the help of the calculated intermediate coupling coefficients, this phenomenon is elucidated by the fact that the \(LS\) coupling singlet nature of the ground state selects the \(LS\) coupling singlet component from the excited wave function, i.e., the contribution from the singlet component in the excitation is dominant in the generalized oscillator strength. Furthermore, the similar dynamic behavior was observed not only for the valence-shell excitations but also for the inner-shell excitations and the dipole-forbidden excitations.

Acknowledgments
The author acknowledges the supports by the National Nature Science Foundation of China (Grants Nos. 10734040 and 10874168), CAS Knowledge Promotion Project (Grants No. KJCS1-YW-N30) and National Basic Research Program of China (Grant No. 2010CB923301).

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