Selectivity in valence excitation processes of noble atoms studied by fast electron impact

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The experimental and theoretical ratios of the generalized oscillator strength (GOS) for valence excitations of Ne, Ar, and Kr are presented. The excitation selectivity for fast electron impact is elucidated: the *LS* coupling singlet nature of the ground state selects the *LS* coupling singlet components from the intermediate coupling wave function of the excited state. According to this rule, the GOS's for some unresolved transitions were determined. Furthermore, this work provides an experimental method to determine the intermediate coupling coefficients of singlet components.

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I. INTRODUCTION

As noble atoms get heavier, it is well known that *LS* coupling would be transferred to intermediate coupling to describe the energy levels. This is because of the enhancement of the relativistic effect. However, Snell *et al.* [1] suggested that a four-parameter model, which depends on the validity of *LS* coupling, is sufficient to describe the Xe 4*d* photoionization process. Thus we pose this question: "Why can the photoionization process for such a heavy noble atom be described by *LS* coupling, and how about the excitation process?" This is a fundamental question of atomic physics. In the present work, we explore the valence shell transition behavior of heavy noble atoms for fast electron impact by analyzing the generalized oscillator strength (GOS) ratio, and we try to answer this question.

The GOS can be used to evaluate the theoretical methods, determine correct spectral assignments [2], and explore the excitation dynamics [3]. In addition, the electron impact excitation mechanism for noble atoms has been extensively used in producing x-ray lasers [4,5]. The differential cross-section (DCS) ratios were used to probe the scattering behavior of noble atoms by Bartschat and Madison [6], Khakoo *et al.* [7–11], and Guo *et al.* [12–14]. However, these works were carried out with slow electron impact, and the collision problem is considered as a combined system of the incident particle and the target, in which both of them lost their mechanical individuality. On the contrary, 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. Therefore, the cross section can be factorized into two factors: one dealing with the incident particle and the other (GOS) dealing with the target [15]. So the GOS ratio may provide new insight into the atomic excitation process.

II. EXPERIMENTAL METHOD

The angle-resolved electron-energy-loss spectrometer (AREELS) used in this experiment has been described in detail in Refs. [16–18]. Briefly, it consists of an electron gun, a hemispherical electrostatic monochromator made of aluminum, a rotatable energy analyzer of the same type, an interaction chamber, a number of cylindrical electrostatic lenses, and a one-dimensional position sensitive detector to detect the scattered electrons. All of these components are enclosed in four separate vacuum chambers made of stainless steel. In the present experiment, the impact energy was 2.5 keV and the energy resolution was 65 meV [full width at half maximum (FWHM)]. The background pressure in the vacuum chamber was 5×10^{-5} Pa. The electron-energy-loss spectra in the region of 9–13 eV of Kr and in the region of 16–21 eV of Ne were measured for scattering angles from 1.0° to 8.5°. The effects of double scattering and angular resolution were corrected, as described in Refs. [17,19]. The GOS ratios for Ar were determined from our previous work [20]. The intermediate coupling coefficients for relevant levels of Ne, Ar, and Kr were calculated using the Cowan code [21].

III. RESULTS AND DISCUSSION

A typical electron-energy-loss spectrum of Kr is shown in Fig. 1, and the assignments are listed in Table I. Since the spin-forbidden transition is negligibly small for fast electron impact [15] and the initial ground state is singlet, the contribution of excitation to triplet component to GOS can be neglected. The states of $5s[3/2]_2$, $5s'[1/2]_0, 5p[5/2]_3, 4d[1/2]_0, 4d[7/2]_4$, and $6s[3/2]_2$ of Kr, which are dominated by the triplet components according to the intermediate coupling coefficients, do not appear in our measured spectra. It is noticed that the excitations to $5p[1/2]_1, 5p[3/2]_1, 5p'[3/2]_1$, and $5p'[1/2]_1$, which correspond to the magnetic dipole transitions based on their singlet components, do not appear in our measured spectra. This might be interpreted as that the magnetic dipole excitation generally occurs only between the electronic states with the same configuration [23], e.g., $2p^2\ ^3P_1 \rightarrow 2p^2\ ^3P_2$. However, for the present excitations, the final configuration $4p^5 5p$ is different from the initial one, so that transition probability is negligibly small. And the magnetic quadrupole excitations to $4d[3/2]_2$ and $4d[5/2]_2$ should be even weaker. Thus the singlet components of the observed excited states of Kr consist of

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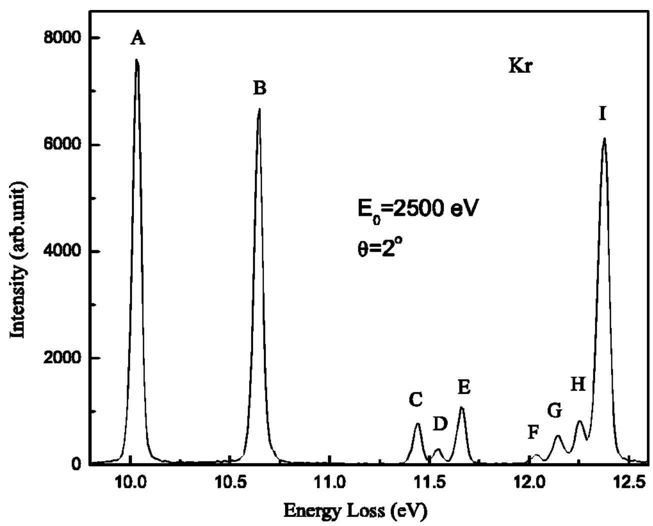

FIG. 1. An electron-energy-loss spectrum of krypton.

$4p^5(5p,6p)^1S_0, 4p^5(4d,5s,5d,6s)^1P_1, 4p^55p\ ^1D_2$, and $4p^54d^1F_3$, whose corresponding transitions are the electric monopole, electric dipole, electric quadrupole, and electric octupole excitations, respectively. Here we present the significant intermediate coupling coefficients and the transition multipolarity for the peaks from *A* to *I* of Kr in Table I.

In Fig. 2(a), the experimental GOS ratios between the dipole-allowed excitations to $5s[3/2]_1$ and $5s'[1/2]_1$ of Kr are presented. In order to compare the intermediate coupling coefficients with the GOS ratios, here we define the "component coefficient square ratio (CCSR)" as $E_1\alpha_1^2/E_2\alpha_2^2$ (E_1 and E_2 are the excitation energies, and α_1 and α_2 are the intermediate coupling coefficients of the same singlet components). According to the Bethe theory [15,24], the GOS ratio between excitations to $5s$ and $5s'$ of Kr can be expressed as

$$\delta = \frac{f(5s[3/2]_1)}{f(5s'[1/2]_1)} = \frac{E_1}{E_2}\frac{K_2^2}{K_1^2}\frac{\left|\langle\Psi_{5s[3/2]_1}|\sum_{j=1}^{N}\exp(i\mathbf{K_1}\cdot\mathbf{r_j})|\Psi_{^1S_0}\rangle\right|^2}{\left|\langle\Psi_{5s'[1/2]_1}|\sum_{j=1}^{N}\exp(i\mathbf{K_2}\cdot\mathbf{r_j})|\Psi_{^1S_0}\rangle\right|^2} \simeq \frac{E_1}{E_2}\frac{0.713^2}{0.700^2} = 0.98, \quad (1)$$

TABLE I. The assignments of peaks from *A* to *I*. The energy values are taken from Moore [22]. $E0, E1, E2$, and $E3$ represent electric monopole, electric dipole, electric quadrupole, and electric octupole transitions from the ground state ($4p^6\ ^1S_0$), respectively.

Peak	*JL* designation	Intermediate coupling		Transition energy (eV)
	$4p^6$	$0.9973(4p^6\ ^1S_0)$		0
A	$4p^55s[3/2]_1$	$-0.6969(4p^55s\ ^3P_1)+0.7132(4p^55s\ ^1P_1)$	$E1$	10.033
B	$4p^55s'[1/2]_1$	$0.7059(4p^55s\ ^3P_1)+0.6995(4p^55s\ ^1P_1)$	$E1$	10.644
C	$4p^55p[5/2]_2$	$0.7143(4p^55p\ ^3D_2)+0.1642(4p^55p\ ^3P_2)$ $-0.6801(4p^55p\ ^1D_2)$	$E2$	11.445
D	$4p^55p[3/2]_2$	$0.2251(4p^55p\ ^3D_2)+0.8661(4p^55p\ ^3P_2)$ $+0.4455(4p^55p\ ^1D_2)$	$E2$	11.546
E	$4p^55p[1/2]_0$	$-0.6859(4p^55p\ ^3P_0)+0.7126(4p^55p\ ^1S_0)$ $-0.1157(4p^56p\ ^1S_0)$	$E0$	11.666
F	$4p^54d[1/2]_1$	$0.9411(4p^54d\ ^3P_1)+0.1029(4p^54d\ ^1P_1)$ $+0.2471(4p^54d\ ^3D_1)-0.1185(4p^55s\ ^3P_1)$ $+0.1088(4p^55d\ ^3P_1)$	$E1$	12.037
G	$4p^55p'[3/2]_2$	$0.6618(4p^55p\ ^3D_2)-0.4709(4p^55p\ ^3P_2)$ $+0.5817(4p^55p\ ^1D_2)$	$E2$	12.144
	$4p^54d[7/2]_3$	$0.7837(4p^54d\ ^3F_3)+0.1861(4p^54d\ ^3D_3)$ $-0.5906(4p^54d\ ^1F_3)$	$E3$	12.179
H	$4p^55p'[1/2]_0$	$0.7261(4p^55p\ ^3P_0)+0.6601(4p^55p\ ^1S_0)$ $-0.1687(4p^56p\ ^1S_0)$	$E0$	12.257
	$4p^54d[5/2]_3$	$0.2226(4p^54d\ ^3F_3)+0.8031(4p^54d\ ^3D_3)$ $+0.5494(4p^54d\ ^1F_3)$	$E3$	12.284
I	$4p^54d[3/2]_1$	$0.6338(4p^54d\ ^3D_1)-0.6057(4p^54d\ ^1P_1)$ $+0.2037(4p^55d\ ^1P_1)+0.3250(4p^56s\ ^1P_1)$ $-0.2402(4p^56s\ ^3P_1)$	$E1$	12.354
	$4p^56s[3/2]_1$	$0.2863(4p^54d\ ^1P_1)+0.1220(4p^54d\ ^3P_1)$ $-0.2495(4p^54d\ ^3D_1)-0.5437(4p^56s\ ^3P_1)$ $+0.7278(4p^56s\ ^1P_1)$	$E1$	12.385

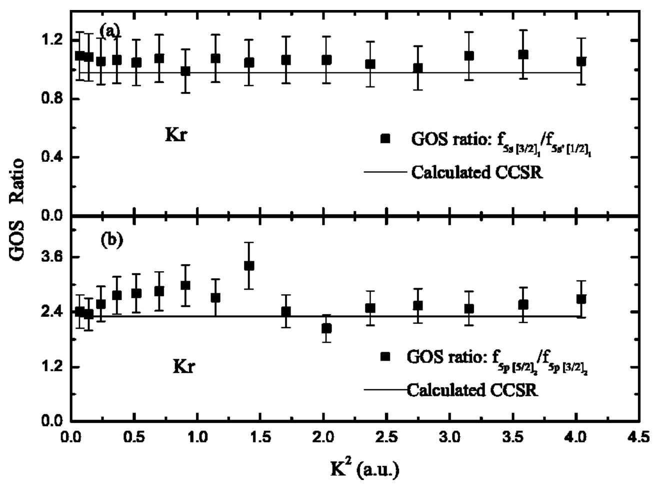

FIG. 2. The experimental GOS ratios and calculated CCSR's: (a) between the excitations to $5s[3/2]_1$ and $5s'[1/2]_1$ of Kr, (b) between the excitations to $5p[5/2]_2$ and $5p[3/2]_2$ of Kr.

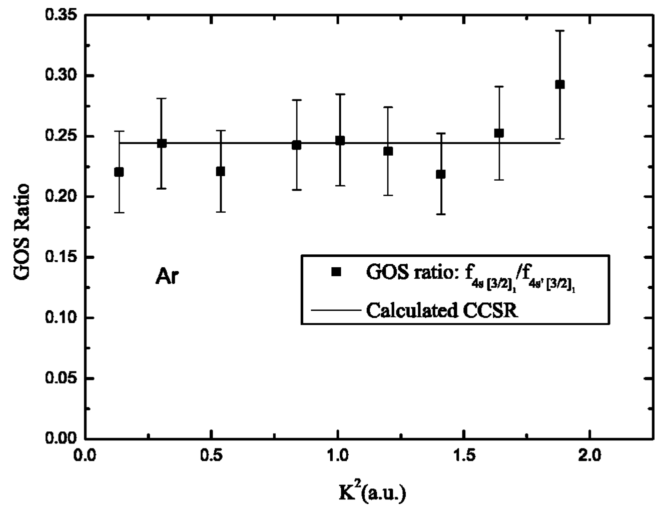

FIG. 4. Same as Fig. 2, but between the excitations to $4s[3/2]_1$ and $4s'[1/2]_1$ of Ar.

where f is GOS, and K is the momentum transfer ($K_1 \simeq K_2$). Thus the excellent agreement between the GOS ratios and the calculated CCSR between excitations to $5s$ and $5s'$ can be well explained, and the ratios are independent of K^2. The GOS ratios between the electric quadrupole excitations to $5p[5/2]_2$ and $5p[3/2]_2$ of Kr are shown in Fig. 2(b). Although the difference is somewhat larger around $K^2 = 1.5$ a.u., where the minimum of the GOS is located, the GOS ratios agree well with the corresponding CCSR in general. Furthermore, the measured GOS ratios and the calculated CCSR's between excitations to $ns[3/2]_1$ and $ns'[1/2]_1$ for Ne($n=3$) and Ar($n=4$) are shown in Fig. 3 and Fig. 4, respectively. It can be seen that the experimental GOS ratios are in excellent agreement with the corresponding CCSR's.

All of these results indicate that 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 components are enough to describe the excitation process from the ground state for fast electron impact. Since the photoexcitation and photoionization are the electric dipole processes, which correspond to the excitation and ionization for fast electron impact at the optical limit ($K^2 \to 0$), the four-parameter model suggested by Snell *et al.* [1] is sufficient to describe the Xe $4d$ photoionization process.

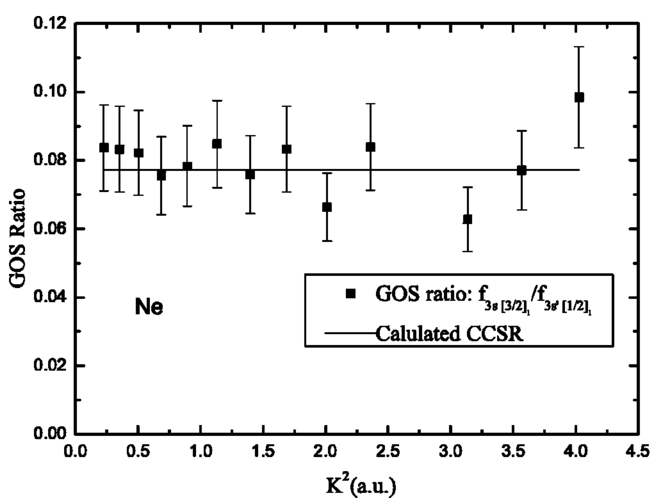

FIG. 3. Same as Fig. 2, but between excitations to $3s[3/2]_1$ and $3s'[1/2]_1$ of Ne.

In addition, using the mean value of the experimental GOS ratios and the normalization character of the intermediate coupling coefficients, we determined the intermediate coupling coefficients of the singlet components for the states of $ns[3/2]_1$ and $ns'[1/2]_1$ of Ne ($n=3$), Ar ($n=4$), and Kr ($n=5$) (here the configuration interaction can be neglected), and the results are shown in Table II. It can be seen that the experimental values are in good agreement with our calculated ones. So the GOS ratios or the derived experimental intermediate coupling coefficients can be used as a stringent test of the wave function as well as the interaction model.

The present results show that the GOS for an excitation with only one significant singlet component is proportional to the square of the corresponding intermediate coupling coefficient. Thus if the GOS for such an excitation is known, the GOS for another excitation with the same singlet component can be determined based on this rule.

The GOS for peak G of Kr, which shows a wide profile and approaches zero as $K^2 \to 0$, is presented in Fig. 5(a). From Table I, we notice that the peak G corresponds to two unresolved transitions of the electric quadrupole ($4p^6\,{}^1S_0$

TABLE II. The experimental (E) and theoretical (T) intermediate coupling coefficients of singlet components for the states of $ns[3/2]_1$ and $ns'[1/2]_1$ of Ne ($n=3$), Ar ($n=4$), and Kr ($n=5$).

	Ne		Ar		Kr	
State	E	T	E	T	E	T
$ns[3/2]_1$	0.274	0.269	0.444	0.446	0.728	0.713
$ns'[1/2]_1$	0.962	0.963	0.896	0.894	0.686	0.700

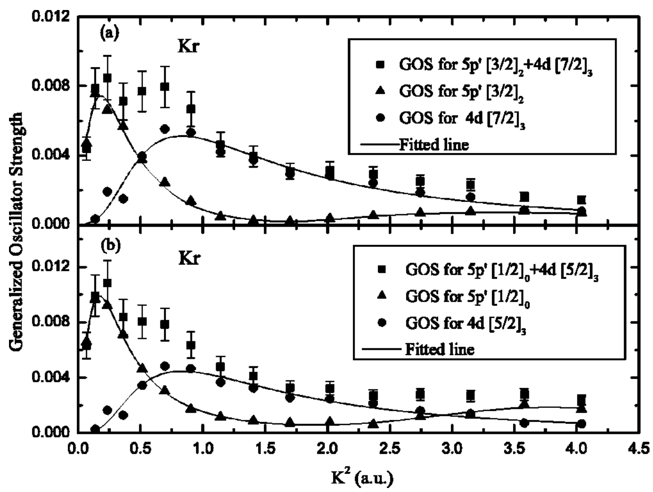

FIG. 5. The GOS's for the peak G (a) and H (b).

$\rightarrow 4p^5 5p\ ^1D_2$) and the electric octupole ($4p^6\ ^1S_0 \rightarrow 4p^5 4d\ ^1F_3$). Since the GOS for the excitations to the $4p^5 5p\ ^1D_2$ peak C has been obtained in our previous work [25], the GOS for the electric quadrupole excitation in peak G can be determined with the help of the corresponding CCSR. Then by subtracting the contribution of the electric quadrupole component from the GOS of peak G, we can obtain the GOS for the electric octupole excitation to $4p^5 4d\ ^1F_3$. Thus the GOS's for the excitations to $5p'[3/2]_2$ and $4d[7/2]_3$ are all obtained. It should be noticed that the GOS profile for the electric octupole transition is much broader than one for electric monopole and electric quadrupole transitions, and its maximum is located at about $K^2 = 0.8 \pm 0.1$ a.u. A similar behavior for Xe was also observed by Suzuki *et al.* [26], therein the GOS for the electric octupole excitation to $5p^5 5d[7/2]_3$ was measured at the incident energy of 400 eV. This behavior may be the characteristic of the electric octupole transitions.

The GOS for peak H of Kr is presented in Fig. 5(b). From Table I we can find that peak H corresponds to electric monopole excitations to $4p^5(5p,6p)^1S_0$ and electric octupole excitation to $4p^5 4d\ ^1F_3$. Since the GOS for electric octupole excitation to $4p^5 4d\ ^1F_3$ in peak G has been obtained, using the same method as in peak G, the GOS's for the electric octupole and electric monopole excitations in peak H can be separated, as shown in Fig. 5(b).

IV. SUMMARY AND CONCLUSION

In summary, by analyzing the experimental GOS ratios and the calculated CCSR's for the dipole-allowed and dipole-forbidden excitations of noble atoms, it is found that the *LS* coupling singlet nature of the ground state selects the *LS* coupling singlet components from the excited wave function, i.e., the contributions from excitations to singlet components are dominant the GOS's. According to the intermediate coupling coefficients, the valence excitations of Kr can be classified as electric monopole, electric dipole, electric quadrupole, and electric octupole transitions. An experimental method to determine the coefficients of singlet components in the intermediate coupling scheme is provided when the configuration interaction is neglected. Thus the quantitative information of the excited wave functions can be determined directly from experiment, and the GOS ratio can serve as a rigorous test of the wave functions as well as the interaction model. For an excited state including only one singlet component in the intermediate coupling scheme, the GOS is proportional to the square of the intermediate coupling coefficient of the singlet component. Furthermore, the GOS's for excitations to $5p'[3/2]_2, 4d[7/2]_3, 5p'[1/2]_0$, and $4d[5/2]_3$ of Kr are obtained, although they cannot be resolved in the present experiment.

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