Generalized oscillator strength for Na $3s - 3p$ transition

Zhifan Chen and Alfred Z. Msezane

Center for Theoretical Studies of Physical Systems, and Department of Physics
Clark Atlanta University, Atlanta, Georgia 30314, U. S. A.

ABSTRACT

Generalized oscillator strengths (GOS’s) for the Na $3s - 3p$ transition have been investigated using the spin-polarized technique of the random phase approximation with exchange (RPAE) and the first Born approximation (FBA), focussing our attention on the position of the minimum. Intershell correlations are found to influence the position of the minimum significantly, but hardly that of the maximum. The RPAE calculation predicts for the first time the positions of the minimum and maximum at momentum transfer, $K$ values of 1.258 a.u. and 1.61 a.u., respectively. The former value is within the range of values extracted from experimental measurements, $K = 1.0 - 1.67$ a.u.. We recommend careful experimental search for the minimum around the predicted value for confirmation.

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1 Introduction

The generalized oscillator strength (GOS) is an important property of the atom, since Bethe introduced it [1]. To study this property, the sodium atom has been chosen as the subject in many experimental and theoretical investigations because its electronic configuration has an inert core and a single valence electron which is similar to that of the hydrogen atom. The differential cross sections (DCS’s) and GOS’s for the Na 3s – 3p transition were measured by Shuttleworth et al [2] using a high-resolution electron spectrometer over the angular range of 1°-20° at the incident electron energies of 54.4, 100, 150 and 250 eV. The measurement observed a GOS minimum at the momentum transfer value of \( K = 0.67 \) a.u. (or \( K^2 = 0.45 \) a.u.). Buckman and Teubner [3] measured data for the same transition using a modulated crossed-beam technique at the incident energies of 54.4, 100, 150 and 217.7 eV, covering the extended angular range of 2° to 145°. Their data are in good agreement with those of Shuttleworth et al at small angles but did not show a GOS minimum around \( K^2 = 0.45 \) a.u..

The Shuttleworth’s et al minimum was also not confirmed by the experiment of Srivastava and Vuskovic [4], which utilized a crossed-electron-beam-metal-atom-beam scattering technique and performed measurements at incident energies of 10, 20, 40, and 54.4 eV. The data of Srivastava and Vuskovic are in poor agreement with those of Buckman and Teubner at large
angles. Srivastava and Vuskovic implied that the conflict was caused by the improper geometrical correction factor in Buckman and Teubner’s experiment. To resolve the discrepancy Teubner et al [5] remeasured the Na 3s – 3p transition at 22.1 and 54.4 eV and found a probable source of systematic error in the measurement of Srivastava and Vuskovic. All the measurements did not observe the GOS minimum around \( K = 0.67 \) a.u. as predicted by Shuttleworth et al. Some recent measurements on the Na 3s – 3p transition by Bielschowsky et al [6] at impact energy of 1 keV and Marinkovic et al [7] at 10, 20 and 54.4 eV did not report GOS minima in the momentum transfer regions they considered.

Theoretically, Shimamura [8] predicted GOS minima to appear between \( K^2 = 0.72 \) and 0.93 a.u., depending on the choice of the exponent in the Slater orbitals. Miller [9] calculated the GOS minimum in the Na 3s – 3p transition within the FBA, employing hydrogenlike orbitals with effective nuclear charge and predicted a minimum at \( K^2 = 0.71 \) a.u.. The FBA calculation from Bielschowsky et al showed a minimum around \( K^2 = 2.0 \) a.u.. In summary the GOS minimum for the Na 3s – 3p transition observed in Shuttleworth’s et al experiment was not confirmed by other measurements. Also, the wide range of positions of the minimum predicted by the theoretical calculations at \( K^2 = 0.71, 0.72, 0.93 \) and 2.0 a.u. are not observed by the experiments. Obviously, the GOS and the position of its minimum for the Na
3s – 3p transition is still an unresolved and interesting problem.

In this paper we have used the spin-polarized technique of the random phase approximation with exchange (RPAE) to investigate the GOS for the Na 3s – 3p transition. The major objective of our calculation has been the unambiguous identification and location of the positions of the minimum and maximum. As a result, values of $K$ were carefully selected. We found for the first time the positions of the minimum and the maximum to be at around $K = 1.258$ a.u. (or $K^2 = 1.582$ a.u.) and $K = 1.61$ a.u. (or $K^2 = 2.59$ a.u.), respectively. After careful analysis of the experimental data, we found the GOS minimum at $K = 1.258$ a.u. is supported by some previous measurements. The intershell correlations are found to have significant effect to the position of the minimum.

2 THEORY

In the FBA the generalized oscillator strength, $f$, for dipole allowed transitions in the length form can be calculated [10] as

$$f = \frac{2(2l + 1)N_l w}{(2l_i + 1)K^2}|d_\alpha|^2$$  \hspace{1cm} (1)

where $N_l$ is the number of electrons in the excited state, $l_i$ is the initial orbital angular momentum of the excited electron, $l$ is the total angular momentum of the electron-hole pair, which satisfies triangle rule $|l_f + l_i| > l > |l_i - l_f|$, and $w$ is the width of the initial state.
$w$ is the excitation energy (a.u.). The dipole matrix element, $d_\alpha$, can be calculated from

$$<\phi_f|d_\alpha|\phi_i> = \sqrt{(2l_i+1)(2l_f+1)} \left( \begin{array}{ccc} l_f & l & l_i \\ 0 & 0 & 0 \end{array} \right) \int_0^\infty P_i(r)P_f(r)j_l(Kr)dr \quad (2)$$

where $P_i(r), P_f(r)$ are the radial wave functions of the initial and final states, respectively, $j_l(Kr)$ is the spherical Bessel function. For the dipole allowed transition, the calculations performed in this paper are with $l = 1$ and $P_i(r), P_f(r)$ represented by Hartree-Fock wave functions. Each channel, such as $s-p$ transition, includes three discrete excited states, 3$p$, 4$p$, 5$p$, and seventeen continuum states. The radial part of the wave function for each state was represented by 700 points.

According to the semiempirical Hund rule, the total spin of a shell in the ground state reaches the largest value permitted by the Pauli principle. Therefore, in the semiclosed shell all the electron spin vectors are collinear, and their projections on to an arbitrary fixed direction are equal. Therefore, every shell can be devided into two spin subshells, each having a certain spin direction, $\uparrow$ or $\downarrow$. Because of this the wave functions double. Electrons in the subshell interact with other electrons in two different ways, with or without exchange. Since the Coulomb interaction will not change the spin direction, only the electrons having the same spin direction can interact with exchange.

The equation for the dipole matrix element in the spin-polarized technique
of the RPAE [11] is

\[
(D_{\alpha}^+, D_{\alpha}^-) = (d_{\alpha}^+, d_{\alpha}^-) + \sum_{\alpha'} (D_{\alpha'}^+, \chi_{\alpha'}^+) \left( \begin{array}{cc}
u_{\alpha'\alpha}^+ & u_{\alpha'\alpha}^+ \\ u_{\alpha'\alpha}^- & \nu_{\alpha'\alpha}^- \end{array} \right)
\]

(3)

where \( u_{\alpha'\alpha} \) is the Coulomb inter-electron potential, \( d_{\alpha}^\pm \) represents the amplitude for the direct excitation of \( 3s^\mp - 3p^\mp \) and \( \chi_{\alpha'} \) is the electron-vacancy propagator. If the states of \( \uparrow \) and \( \downarrow \) electron are equivalent, the dipole matrix element \( D_{\alpha}^\uparrow \) in the RPAE for the \( 3s^\uparrow - 3p^\uparrow \) transition can be obtained from

\[
< \epsilon_f | D_{\alpha}^\uparrow | \epsilon_i > = < \epsilon_f | d_{\alpha}^\uparrow | \epsilon_i > + \left( \sum_{\epsilon_3 \leq F, \epsilon_4 > F} - \sum_{\epsilon_3 > F, \epsilon_4 \leq F} \right) \frac{< \epsilon_4 | D_{\alpha}^\uparrow | \epsilon_3 > < \epsilon_3 \epsilon_f | u_{\alpha'\alpha}^+, \nu_{\alpha'\alpha}^+ | \epsilon_4 \epsilon_i >}{w - \epsilon_4 + \epsilon_3 + i\eta(1 - 2n_4)}
\]

(4)

where \( \epsilon_3 \) and \( \epsilon_4 \) represent the virtual excitation states, \( i\eta \) gives the direction of tracing the pole while integrating over the energy, \( \eta \to +0 \), \( F \) is the Fermi energy of the atom, and \( n_4 \) is the Fermi step: \( n_4 = 1, \epsilon_4 \leq F; n_4 = 0, \epsilon_4 > F \). The symbol \( \sum \) denotes summation over discrete and integration over continuous states. A similar equation for the \( D_{\alpha}^\downarrow \) of the \( 3s^\downarrow - 3p^\downarrow \) transition can be obtained. It is important to remember that only the states with the same spin direction can have exchange interaction in the sum of Eq. (4).

Finally, the GOS for Na \( 3s - 3p \) transition can be written as

\[
f = \frac{2(2l + 1)N_i w}{(2l_i + 1)K^2} (D_{\alpha}^{\uparrow \uparrow} + D_{\alpha}^{\downarrow \downarrow})
\]

(5)

Eqs. (1),(2),(4) and (5) are the basic equations used in this paper to calculate the GOS’s in FBA and RPAE.
3 RESULTS AND DISCUSSION

The results of our calculation are given in Fig. 1, Fig. 2 and Table 1. Table 1 lists the positions of the minimum and maximum for the Na $3s - 3p$ transition obtained by different authors. The results of our FBA calculation are in excellent agreement with that of Bielschowsky et al. The data of Shuttleworth et al showed a GOS minimum around $K = 0.67$ a.u.. However, this minimum was not observed in all other measurements. After analyzing the experimental data obtained by Shuttleworth et al [2], Buckman and Teubner [3], Srivastava and Vuskovic [4], Teubner et al [5], and Marinkovic et al [7], we found that the GOS minima from the experimental data are around $K = 1.0 - 1.67$ a.u. (or $K^2 = 1.0 - 2.80$ a.u.) at impact energies of 20 and 54.4 eV. These minima were not originally noticed by the experimentlists.
Fig. 1 shows the GOS’s versus $K^2$ for the Na $3s - 3p$ transition at 20 eV. The circles are from Srivastava and Vuskovic, black dots are from Marinkovic.
et al, and squares are from Teubner et al. The solid and dotted lines represent our RPAE and FBA calculations, respectively. All three measurements show the GOS minimum around $K = 1.0 - 1.18$ a.u. (or $K^2 = 1.0 - 1.39$ a.u.) which are close to our results.
Fig. 2 is the same as Fig. 1, except that the data are at 54.4 eV and the crosses are from Buckman and Teubner and the triangles are from Shut-
tleworth et al. The black dots and circles show the GOS minimum around
\( K = 1.67 \) a.u. (or \( K^2 = 2.80 \) a.u.), while the crosses give a minimum at
\( K = 1.51 \) a.u. (or \( K^2 = 2.29 \) a.u.). Since the position of the GOS min-
imum for the Na 3s – 3p transition occurs at large momentum transfer,
\( K = 1.0 - 1.67 \) a.u., which corresponds to large angles in the measurements;
for example \( \theta = 37^\circ \) at the impact energy of 54.4 eV (\( K = 1.258 \) a.u.). There-
fore it is easy to miss the GOS minimum in the measurement by taking large
angular steps at large angles as is the usuall practice. From our calculation
it is suggested to reperform the experiment for Na 3s – 3p transition and pay
particular attention to the position of the minimum we indicated in Table 1.
We believe more experimental data will be obtained to confirm our results.

It is interesting to compare the intershell correlations for the transitions
between the Na 3s – 3p and the Ar 3p – 4s. The position of the GOS min-
imum for the later is influenced insignificantly by correlations. The difference
between positions of the minimum from the RPAE and FBA is less than
0.7\%. However, in the Na 3s – 3p transition the difference is more than 11\%.
This is because the Na 3p level is only 2.1 eV above the 3s ground state;
therefore Na has an enormous dipole polarizability (23.6 × 10^{−24} cm^3). In
RPAE the many-electron correlation effects are essentially due to polariza-
tion of the electron shell by the external field. Results from RPAE will show
a significant difference from that of FBA if the atoms have large polarizabil-
ity. Therefore, we can expect that Li, K and Rb atoms will yield results that are similar to those of Na, while Ne, Kr and Xe atoms will exhibit results similar to those of Ar if we compare the positions of GOS minimum from RPAE and FBA.

Correlations between intershell electrons are strongly affected by the separation of the subshells. The $3s \uparrow - 3p \uparrow$ excitation energy will be 0.07267 (a.u.), 0.07255 (a.u.), 0.07190 (a.u.), and 0.07175 (a.u.) if channels $(s^\uparrow - p^\uparrow)$, $(s^\uparrow - p^\uparrow + p^\downarrow - s^\downarrow + p^\downarrow - s^\downarrow)$, $(s^\uparrow - p^\uparrow + p^\downarrow - d^\downarrow + p^\downarrow - d^\downarrow)$ and $(s^\uparrow - p^\uparrow + p^\downarrow - d^\downarrow + p^\downarrow - d^\downarrow + p^\uparrow - s^\downarrow + p^\downarrow - s^\downarrow)$ are included in the RPAE calculations. This indicates that the influence of the $3d^{\uparrow \downarrow}$ electrons upon the $3p \uparrow$ electron is larger than that from the $4s^{\uparrow \downarrow}$ electrons.

In conclusion, the positions of the minimum and the maximum for the Na $3s - 3p$ transition have been calculated and found for the first time from RPAE at the momentum transfer values of $K = 1.258$ a.u. and 1.61 a.u., respectively. Furthermore, the many-electron correlations are found to play an important role in the determination of the position of the minimum. We recommend that experiments search carefully for predicted minimum, using the value obtain here as a guide.
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Figure Captions

Fig. 1. GOS’s versus $K^2$ for the Na 3$s$ – 3$p$ transition at 20 eV. Circles are from Srivastava and Vuskovic, black circles are from Marinkovic et al., and squares are from Teubner et al. Solid and dotted lines represent our RPAE and FBA calculations, respectively.

Fig. 2. GOS’s versus $K^2$ for the Na 3$s$ – 3$p$ transition at 54.4 eV. All the symbols have the same meaning as in Fig. 1, except that crosses are from Buckman and Teubner and triangles are from Shuttleworth et al.
Table 1: Positions of the GOS minimum and the maximum for the excitation of Na $3s - 3p$

| Atom          | Authors                    | $K$ (a.u.) for minimum | $K$ (a.u.) for maximum |
|---------------|----------------------------|------------------------|-------------------------|
|               | Shuttleworth *et al* [2]   | 0.67                   |                         |
|               | Marinkovic *et al* [7]     | 1.18-1.67              |                         |
|               | Buckman and Teubner [3]    | 1.51                   |                         |
|               | Srivastava and Vuskovic [4]| 1.0-1.7                |                         |
|               | Teubner *et al* [5]        | 1.15                   |                         |
|               | Shimamura                 | 0.847-0.965            |                         |
| Na            | Miller                    | 0.84                   |                         |
|               | Bielschowsky *et al*       | 1.41                   |                         |
|               | Present RPAE              | 1.258                  | 1.61                    |
|               | Present H-F               | 1.401                  | 1.69                    |