

## Generalized oscillator strengths for open-shell and closed-shell atoms

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(Received 19 April 2002; published 6 February 2003)

A formula, which is a useful improvement over existing ones, has been obtained to calculate the generalized oscillator strengths (GOS's) for the discrete transitions of both open-shell and closed-shell atoms. We illustrate our formula by calculating the GOS for the oxygen  $2p^4(^3P)-2p^3(^4S)3s(^3S)$  transition and obtain overall good agreement with the experimental data. Our calculated GOS reveals a minimum at around the momentum transfer,  $K=1.26$  a.u. We recommend a careful experimental search for the position of the minimum around the predicted value for confirmation.

DOI: 10.1103/PhysRevA.67.024701

PACS number(s): 34.10.+x, 34.50.Fa, 31.50.Df

The generalized oscillator strength (GOS), introduced by Bethe [1] and discussed further by Inokuti [2], is an important scattering quantity which is closely related to the differential cross sections (DCS's) in the Born approximation and is also useful for obtaining integral cross sections and optical oscillator strengths (OOS's) [3,4].

After many years of studies of the GOS, it is still fair to state that extensive theoretical investigations of the GOS's have been carried out only for the noble-gas atoms and a few other closed-shell systems [5–17]. This is because of the availability of detailed experimental results and the relative ease of dealing theoretically with spherically symmetric targets. Hydrogenlike wave functions with an effective nuclear charge [5,9], the analytical independent particle model [7], Hartree-Fock wave functions [8,11,12], the Glauber approximation [13], the random phase approximation with exchange [15,16], etc., are the most general methods used in the calculations. In this paper we first define the GOS and derive a formula to calculate it for the discrete transitions of both open-shell and closed-shell atoms. Then we use the formula to study the oxygen  $2p^4(^3P)-2p^3(^4S)3s(^3S)$  transition and compare our results with those of measurements.

The GOS is defined as [1]

$$gf(K, w) = \frac{2w}{K^2} \left| \sum_{i=1}^{i=N} \int \psi_f(\vec{r}_1 \cdots \vec{r}_N) \times \exp(i\vec{K} \cdot \vec{r}_i) \psi_i(\vec{r}_1 \cdots \vec{r}_N) d\vec{r}_1 \cdots d\vec{r}_N \right|^2, \quad (1)$$

where  $\vec{r}_i$  is the position vector of the  $i$ th electron,  $w = \epsilon_f - \epsilon_i$  is the excitation energy,  $K$  is the momentum transfer to the atom,  $\psi_i(\vec{r}_1 \cdots \vec{r}_N)$  and  $\psi_f(\vec{r}_1 \cdots \vec{r}_N)$  are the wave functions representing the initial and final states of the target atom, respectively, and  $g$  is the  $g$  factor [18]. The summation in Eq. (1) is performed over all  $N$  atomic electrons.

Let us consider the atomic transition  $|l_2^N L S M_L M_S\rangle \rightarrow |(l_2^{N-1} L'_2 S'_2, l_3) L' S' M'_L M'_S\rangle$  and expand  $\exp(i\vec{K} \cdot \vec{r}_i)$  in terms of the spherical Bessel function  $j_l(Kr_i)$  and spherical harmonics  $Y_{l0}(\theta_i)$ :

$$\exp(i\vec{K} \cdot \vec{r}_i) = \sqrt{4\pi} \sum_l i^l \sqrt{2l+1} j_l(Kr_i) Y_{l0}(\theta_i). \quad (2)$$

Equation (1) for the GOS then becomes

$$gf(K, w) = \frac{4w}{K^2} \sum_l (2l+1) |T_l|^2. \quad (3)$$

The matrix elements  $T_l$  are given by

$$T_l = \sqrt{2\pi} i^l \sum_i \langle (l_2^{N-1} L'_2 S'_2, l_3) L' S' M'_L M'_S \| Y_{l0}(\theta_i) j_l(Kr_i) \| l_2^N L S M_L M_S \rangle, \quad (4)$$

where  $l_2$  is the initial orbital angular momentum of the electron, and  $L$  and  $S$  are the total orbital angular momentum and spin of the atom, respectively.  $M_L$  and  $M_S$  are the projections of  $L$  and  $S$  on the  $z$  axis,  $L'$ ,  $S'$ ,  $M'_L$ , and  $M'_S$  have the same meaning as  $L$ ,  $S$ ,  $M_L$ , and  $M_S$ , and  $l_3$  is the orbital angular momentum of the excited electron in the final state.  $L'_2$  and  $S'_2$  are the total angular momentum and spin of the atom in the intermediate state with  $l_2^{N-1}$  electrons.

Using the Wigner-Eckart theorem [19], the matrix elements  $T_l$  can be reduced to

$$T_l = \sqrt{2\pi} i^l (-1)^{L'-M'} \delta_{SS'} \delta_{M_S M'_S} \begin{pmatrix} L' & l & L \\ -M'_L & 0 & M_L \end{pmatrix} \sum_i \langle (l_2^{N-1} L'_2 S'_2, l_3) L' S' \| Y_l(\theta_i) j_l(Kr_i) \| l_2^N L S \rangle. \quad (5)$$

The  $3j$  symbol in Eq. (5) shows that  $M_L$  is conserved, viz.,  $M_L = M'_L$ . We can sum over  $M_L$  of the squared  $3j$  symbol and obtain  $1/(2l+1)$ .

We further expand the initial state in terms of  $|l_2^{n-1}\alpha'L_2'S_2'\rangle$  and the final state according to Eq. (15.33) of Sobel'man [19] and use Eqs. (16.21) of [19] and (15.7) of [20] to reduce the matrix elements  $T_l$  to

$$T_l = (-1)^{L'+l_3} i^l C_{L_2'S_2',l_3L'S'}^{l_2LSl} \begin{pmatrix} l_3 & l & l_2 \\ 0 & 0 & 0 \end{pmatrix} [l_2, l_3]^{1/2} \int_0^\infty R_{n_3l_3}(r) R_{n_2l_2}(r) j_l(Kr) r^2 dr, \quad (6)$$

where the coefficient  $C_{L_2'S_2',l_3L'S'}^{l_2LSl}$  is given by

$$C_{L_2'S_2',l_3L'S'}^{l_2LSl} = \sqrt{\frac{n}{2}} [L, L'] (-1)^{L'+L_2'+l_2+l} \left\{ \begin{matrix} L_2' & l_3 & L' \\ l & L & l_2 \end{matrix} \right\} (l_2^{n-1} L_2' S_2') \{ l_2^n L S \}. \quad (7)$$

The  $(l_2^{n-1} L_2' S_2') \{ l_2^n L S \}$  is the fractional parentage coefficient,  $n$  is the electron number,  $n \leq 2(2l_2 + 1)$ , and  $\{ \}$  is the 6j symbol. Two triangle rules for  $l$  are obtained:  $|l_2 + l_3| \geq l \geq |l_2 - l_3|$  and  $|L + L'| \geq l \geq |L - L'|$ .

It should be noted that for the discrete transitions of the rare-gas atoms such as Ar  $3p-4s$ , the  $C_{L_2'S_2',l_3L'S'}^{l_2LSl}$  equals 1.

Therefore, the main difference between the open-shell and closed-shell atoms in the GOS evaluation is Eq. (7).

For the transitions involving open-shell atoms we have to specify the atomic terms in the initial, final, and intermediate states. However, for the closed-shell atoms we only need to evaluate the integral in Eq. (6). After setting the  $C_{L_2'S_2',l_3L'S'}^{l_2LSl}$  and  $g$  equal to 1 we can compare Eq. (3) with the previous closed-shell equations [21]. We note that the previous equations [5,7,9,21] used the triangle rule  $|l_2 + l_3| \geq l \geq |l_2 - l_3|$ . Miller [5] and Ganas and Green [7] evaluated the GOS with the summation over  $m$ . As we discussed before, the summation over  $m$  can be done analytically and the equations will be much simpler, particularly for the electron transitions  $p-p$  and  $p-d$ . For the electron  $p-p$  transition,  $l$  can be 0 or 2, and for  $p-d$ ,  $l$  can be 1 or 3. Experience has taught us that different wave functions for  $l=0$  and  $l=2$  will need an additional computer code to evaluate the integrals. Equation (5a) of [9] is the same as Eq. (10.8) of [21]. By comparing Eq. (10.8) of [21], the closed-shell equation, and our Eq. (3) we found that the main difference is the factors  $2N_i w(2l+1)/(2l_i+1)K^2$  and  $4w(2l+1)/K^2$  where  $N_i$  in Eq. (10.8) of [21] represents the electron number in the initial state, and  $l_i$  is the initial orbital angular momentum of the excited electron. Since the two factors are equal to each other, the results from Eq. (10.8) of [21] and our Eq. (3) should be the same. We note that our Eq. (3) is simpler than Eq. (10.8) of [21]. Also, the difference between our approach and those of Davis and Sinanoglu [22] and Wells and Miller [23] lies in that, contrary to our method, in both of these calculations the summation over the magnetic quantum number  $m$  has not been effected.

The GOS for the oxygen  $2p^4(^3P) \rightarrow 2p^3(^4S)3s(^3S)$  transition has been calculated using Eqs. (3), (6), and (7). The wave functions for the ground state,  $2p^4(^3P)$ , and excited state,  $2p^3(^4S)3s(^3S)$ , were evaluated, respectively, in the Hartree-Fock and Hartree-Fock frozen core approximations. The radial part of the wave function for each state was represented by 1000 points. Our GOS results are plotted with

the experimental data in Fig. 1 as a solid line. As the differential cross sections and the GOS's are related through Eq. (3.1) of [2], the DCS's from the measurements of Kanik *et al.* [24] at an impact energy of 100 eV and Vaughan and Doering [25] at 200 and 150 eV have been transformed to GOS's and are indicated in Fig. 1 by the black circles and circles and crosses, respectively. By carefully choosing the momentum transferred  $K$ , a minimum of the GOS was found around  $K = 1.26$  a.u. From the data of Kanik *et al.* we found that at  $K = 1.15, 1.37,$  and  $1.81$  a.u. the GOS = 0.002 09, 0.001 48, and 0.001 51, respectively. Although the extrapolated experimental data above show a minimum at  $K = 1.37$  a.u., we recommend more experimental investigation of the position of the minimum around the predicted value for confirmation. The significance of the minima of the generalized oscillator strengths has been discussed by Inokuti [2] and Inokuti *et al.* [26]. Briefly, a zero-value minimum of the GOS occurs when the relevant dipole matrix element as a function of  $K^2$  changes its sign. A nonzero minimum of the GOS may signal a failure of the first Born approximation, thereby manifesting the importance of multielectron correlations. The position of the minimum is related to the nodes of

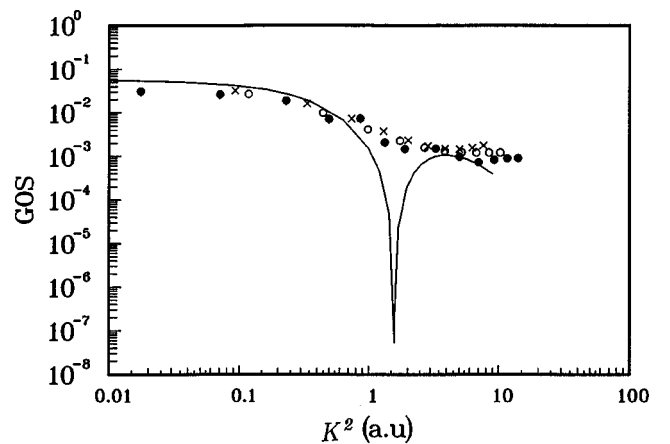


FIG. 1. GOS's vs  $K^2$  for the oxygen  $2p^4(^3P) \rightarrow 2p^3(^4S)3s(^3S)$  transition. The solid line is our results using Eqs. (3), (6), and (7); black circles are from the measurements of Kanik *et al.* [24] at an impact energy of 100 eV; and the circles and crosses are the experimental results of Vaughan and Doering [25] at 200 and 150 eV, respectively.

TABLE I. Optical oscillator strengths for the oxygen  $2p^4(^3P)-2p^3(^4S)3s(^3S)$  transition.

| Authors                   | Expt.             | Theory                    |
|---------------------------|-------------------|---------------------------|
| Bell and Hibbert [27]     |                   | 0.0496 ( <i>R</i> matrix) |
| Bell and Hibbert [27]     |                   | 0.0511 (CI)               |
| Fischer [28]              |                   | 0.0419                    |
| Ganas [29]                |                   | 0.0560                    |
| Reader <i>et al.</i> [30] | 0.050             |                           |
| Jenkins [31]              | $0.053 \pm 0.006$ |                           |
| Present <i>HF</i>         |                   | 0.057                     |

the radial parts of the orbitals of the wave functions of the active electron involved in the transition. Consequently, the minimum provides a stringent test of the accuracy of the calculated wave functions when the results of calculations are compared with those of measurements. Recently, multi-

electron correlation effects on the position of the minimum have been investigated in various atomic transitions [15,16].

When  $K^2 \rightarrow 0$ , the GOS converges to the optical oscillator strength. The result of our calculation is given in Table I. The calculation used the experimental excitation energy [24],  $w = 9.51$  eV, in Eq. (3). Table I lists the OOS's obtained by different authors. Our OOS is in reasonable agreement with those from measurements [30,31] and other calculated results [27–29].

In conclusion, we have obtained a formula to calculate the GOS's for the transitions of both open-shell and closed-shell atoms. The GOS for the oxygen  $2p^4(^3P) \rightarrow 2p^3(^4S)3s(^3S)$  transition has been investigated, and a minimum of the GOS has been found at around the momentum transfer  $K = 1.26$  a.u. More measurements are recommended to confirm the predicted position of the minimum.

This research was supported by U.S. DOE, Division of Chemical Sciences, Office of Basic Energy Sciences, Office of Energy Research (A.Z.M.) and NSF.

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- [1] H. A. Bethe, *Ann. Phys. (Leipzig)* **5**, 325 (1930).  
 [2] M. Inokuti, *Rev. Mod. Phys.* **43**, 297 (1971).  
 [3] G. P. Li, T. Takayanagi, K. Wakiya, H. Suzuki, T. Ajiro, S. Yagi, S. S. Kano, and H. Takuma, *Phys. Rev. A* **38**, 1240 (1988).  
 [4] T. Takayanagi, G. P. Li, K. Wakiya, H. Suzuki, T. Ajiro, T. Inaba, S. S. Kano, and H. Takuma, *Phys. Rev. A* **41**, 5948 (1990).  
 [5] K. J. Miller, *J. Chem. Phys.* **59**, 5639 (1973).  
 [6] J. M. Peek and M. M. Madsen, *Phys. Rev. A* **43**, 147 (1991).  
 [7] P. S. Ganas and A. E. S. Green, *Phys. Rev. A* **4**, 182 (1971).  
 [8] R. A. Bonham, *J. Chem. Phys.* **36**, 3260 (1962).  
 [9] I. I. Shimamura, *J. Phys. Soc. Jpn.* **30**, 824 (1971).  
 [10] I. R. Peterson and J. E. Allen, Jr., *J. Chem. Phys.* **56**, 6068 (1972).  
 [11] T. C. Wong, J. S. Lee, and R. A. Bonham, *Phys. Rev. A* **11**, 1963 (1975).  
 [12] Yong-Ki Kim, M. Inokuti, G. E. Chamberlain, and S. R. Mielczarek, *Phys. Rev. Lett.* **21**, 1146 (1968).  
 [13] C. E. Bielschowsky, G. G. B. de Souza, C. A. Lucas, and H. M. Boecheat Roberty, *Phys. Rev. A* **38**, 3405 (1988).  
 [14] C. E. Bielschowsky, C. A. Lucas, and G. G. B. de Souza, *Phys. Rev. A* **43**, 5975 (1991).  
 [15] Zhifan Chen, A. Z. Msezane, and M. Ya. Amusia, *Phys. Rev. A* **60**, 5115 (1999); Zhifan Chen and A. Z. Msezane, *ibid.* **61**, 30703(R) (2000).  
 [16] Zhifan Chen and A. Z. Msezane, *J. Phys. B* **33**, 2135 (2000); **33**, 5397 (2000); **35**, 815 (2002).  
 [17] Yong-Ki Kim and P. S. Bagus, *Phys. Rev. A* **8**, 1739 (1973).  
 [18] A. Hibbert, *Comput. Phys. Commun.* **9**, 141 (1975).  
 [19] I. I. Sobel'man, *Introduction to the Theory of Atomic Spectra* (Pergamon, Oxford, 1972).  
 [20] U. Fano and G. Racah, *Irreducible Tensorial Sets* (Academic, New York, 1959).  
 [21] M. Y. Amusia and L. V. Chernysheva, *Computation of Atomic Processes* (Institute of Physics, Bristol, 1997).  
 [22] S. L. Davis and O. Sinanoglu, *J. Chem. Phys.* **62**, 3664 (1975).  
 [23] C. A. Wells and K. J. Miller, *Phys. Rev. A* **12**, 17 (1975).  
 [24] I. Kanik, P. V. Johnson, M. B. Das, M. A. Khakoo, and S. S. Tayal, *J. Phys. B* **34**, 2647 (2001).  
 [25] S. O. Vaughan and J. P. Doering, *J. Geophys. Res.* **92**, 7749 (1987).  
 [26] M. Inokuti, Y. Itikawa, and J. E. Turner, *Rev. Mod. Phys.* **50**, 23 (1978).  
 [27] K. L. Bell and A. Hibbert, *J. Phys. B* **23**, 2673 (1990).  
 [28] C. Froese Fischer, *J. Phys. B* **20**, 1193 (1987).  
 [29] P. S. Ganas, *Mol. Phys.* **39**, 1513 (1980).  
 [30] J. Reader, C. H. Corliss, W. L. Wiese, and G. A. Martin, *1980 Atomic Transition Probabilities*, Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 68 (U.S. GPO, Washington, D.C., 1980).  
 [31] D. B. Jenkins, *J. Quant. Spectrosc. Radiat. Transf.* **34**, 55 (1985).