Relativistic effects on the $4p$-$5s$ excitations of krypton

Qi-Cun Shi,[1,2] Ke-Zun Xu,[1] Zhang-Jin Chen,[1] Hyuck Cho,[2] and Jia-Ming Li[3]
[1]*Department of Modern Physics, University of Science and Technology of China, Hefei 230026, China*
[2]*Department of Physics, Chungnam National University, Taejon 305-764, Korea and Center for Molecular Science, Taejon 305-701, Korea*
[3]*Institute of Physics, Chinese Academy of Science, Beijing 100080, China*
(Received 25 June 1997; revised manuscript received 17 November 1997)

Generalized oscillator strengths of krypton for $4p^6(^1S_0)\rightarrow 4p^5(^2P_{3/2,1/2})5s$ are calculated in the first Born approximation at a self-consistent-field level with Dirac-Fock, Dirac-Slater, Hartree-Fock, and Hartree-Slater wave functions. Strong relativistic effects are noticed. The estimate of relaxation on a single-electron matrix element suggests the necessity of a Dirac-Fock multiconfiguration calculation for the excitations. [S1050-2947(98)05506-1]

PACS number(s): 34.80.Dp

Recently, absolute generalized oscillator strengths (GOS's) for the excitation of krypton $4p^6(^1S_0)$ to $4p^5(^2P_{1/2})5s$ and $4p^5(^2P_{3/2})5s$ states, or represented by $4p$-$5s$ in a nonrelativistic approximation, were measured by Takayanagi *et al.* [1] using an electron-energy-loss spectroscopy. The experimental data were utilized by Padma and Deshmukh [2] to check a recently developed relativistic local density potential method, which was designated as RCΞ. A large GOS difference between the RCΞ approach and the Hartree-Slater (HS) calculation was shown and attributed to, with respect to the experiment, three sources: different exchange potential models, different excitation energies, and relativistic effects.

More recently, Msezane and Sakmar [3] gave a simpler formula than the polynomial of Lassettre *et al.* [4] to predict the $4p^5(^2P_{3/2})5s$ optically allowed GOS for the target at small squared momentum transfer (K^2), particularly in the nonphysical region where experimentalists could not reach but had to enter when they normalized relatively measured data using the optical limitation ($K^2=0$). The work of Msezane and Sakmar and the comments of Williams and Stelbovics [5] showed the importance of kinematic and dynamic effects at small K^2. To the best of our knowledge, so far there has been no complete theory that includes all the dynamic effects they listed. We present here the strong relativistic effects at small K^2 in electron-krypton excitation and a significant relaxation estimation before the dynamic effects are investigated one by one.

In the present relativistic calculation in the Coulomb gauge an incident electron at intermediate energies 10^2-10^3 eV is studied, so the relativistic contribution of traverse electromagnetic field is neglected [6] and the first Born approximation is thought to be valid. The details of first Born GOS are well known [3,5,6]. In this paper atomic units (a.u.) are used unless specified otherwise.

The single-electron matrix element in the first Born approximation is studied at the self-consistent-field (SCF) level. The corresponding single-electron orbitals, $4p$ and $5s$ of krypton, are calculated using Dirac-Fock (DF) and Dirac-Slater (DS) approaches. In the DS calculation the exchange potential is approximated with the Slater model (the Slater coefficient α is one) and the $4p$ and $5s$ radial wave functions are obtained based on the same SCF potential. This is the usual way to calculate the matrix element in which the orbitals are orthogonal. In order to estimate the relaxation effect during the excitation, the $4p(^2P_{1/2,3/2})$ orbitals are calculated in the ground-state SCF potential while the $5s(^2S_{1/2})$ orbital is calculated in the excited-state SCF potential, although the overlap between the nonorthogonal ''passive'' electrons is ignored.

It should be noted that at the present SCF level, the deficiency of the DS method is the approximate exchange potential, while that of DF theory in the present krypton excitation is from the average-configuration-state (ACS) approximation. A combined approach of the DF and DS theories is therefore given in this work. In addition, problems related to the electron excitation of closed atoms such as krypton and alkaline metal atoms such as sodium will be discussed together elsewhere [7]. Here the ACS influence on $4p$ and $5s$ orbitals is very small.

The relativistic effects are totally represented by the difference between the sum of the relativistic GOS's for $4p(^2P_{1/2,3/2})$-$5s(^2S)$ and the $4p$-$5s$ GOS of the HS and HF calculations. The HS and HF single-electron orbitals are calculated by enlarging the light speed c in the fine-structure constant $1/c$ by a factor of 1000 in our relativistic programs. The approximate orbitals are tested through nonrelativistic GOS results at $K^2=0$: (i) the present value 0.051 for the $2p$-$3s$ excitation of sodium (not shown here), which agrees with the value 0.050 of Chen and Msezane [8] using the Clementi-Roetti [9] wave functions, and (ii) the lithiumlike GOS's, which are also in agreement with the calculations of Qu *et al.* [6].

The present nonrelativistic GOS's shown in Fig. 1 with solid, dashed, dotted, and dot-dashed curves corresponding to four cases: the usual HS calculation, the result involving the relaxation effect, the result involving the correlation effect, and the result involving these two effects, respectively. Here the correlation effect is estimated with a local density potential model of Vijayakumar *et al.* [10] for the ''Coulomb hole'' [11] as the exchange effect is approximated with the Slater model [12] for the ''Fermi hole.'' In the present calculation the local density correlation potential for the spin-down electrons takes the form of the spin-up electrons, al-

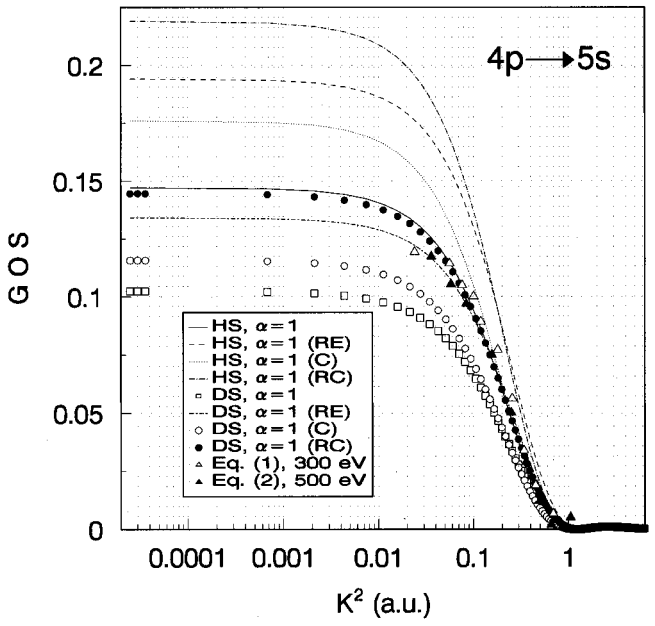

FIG. 1. GOS calculations for the $4p$-$5s$ excitation of krypton. Closed and open triangles denote the GOS sums (obtained as given in the text) of the experimental results of Takayanagi *et al.* [1] at 300 eV and 500 eV electron-impact energies, respectively. The curves and other symbols denote the following: the present calculations of the Dirac-Slater (DS) (Slater coefficient $\alpha=1$) and corresponding Hartree-Slater (HS) approaches (squares and solid curve), and the DS and HS calculations involving the relaxation (RE) effect (long-dash–short-dashed, and dashed curves), the correlation (C) effect (circles and dotted curve), and the mixture of both effects (closed circles and dot-dashed curve).

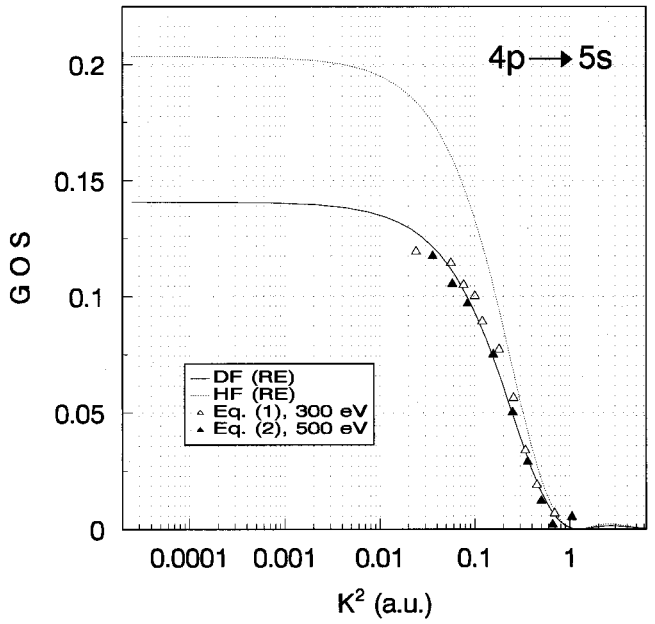

FIG. 2. Same as in Fig. 1, but the solid curve represents the Dirac-Fock result involving the relaxation effect, while the dotted curve represents Hartree-Fock result involving the relaxation effect.

though this technique certainly leads to a small error in the calculation of the spin-down orbital of $4p$. It is noted that in Fig. 1 there is a plateau at K^2 less than 0.01 for each curve. Actually, for krypton K^2 smaller or greater than 0.006 at an electron energy 300 eV defines approximately the nonphysical and physical regions if the experimental excitation energy $w\approx 0.369$ [2] for the $4p^5(^2P_{3/2})5s$ excitation is considered. The corresponding K^2 value at 500 eV is 0.004. A comparison to the relativistic results in Fig. 1 represented as squares, a long-dash–short-dashed curve, circles, and solid circles, respectively, shows large differences at $K^2\leqslant 1$ that cover the nonphysical region and a part of the physical region. The difference at $K^2=0$ reaches nearly 40–100 % depending on the interactions involved: the exchange potential or the exchange potential plus the relaxation effect, or plus the correlation effect, or plus both effects (see Fig. 1).

The existing experiment was given by Takayanagi *et al.* [1]. Because the K^2 values of their $^2P_{3/2}$ data are different from those of their $^2P_{1/2}$ data, we cannot simply sum the points from their figures. The parabolic fitting is then used for the $^2P_{3/2}$ data at 300 eV from their figures and one obtains

$$f_{3/2}=0.161\ 611K^4-0.206\ 794K^2 +0.069\ 269\ 7\ \ \text{at 300 eV}. \tag{1}$$

The experimental data of $^2P_{1/2}$ at a given K^2 value and the corresponding fitted GOS's for $^2P_{3/2}$ are added to yield the approximate value (triangle) shown in Fig. 1. Similarly, the estimation at 500 eV is

$$f_{3/2}=0.106\ 177K^4-0.166\ 747K^2 +0.062\ 722\ 8\ \ \text{at 500 eV}. \tag{2}$$

Figure 1 shows that the approximation from the results of Takayanagi *et al.* [1] supports the present relativistic calculations if the relaxation and correlation effects, particularly the former, are considered, although the results involving both effects are somewhat large in the nonphysical region. The surprisingly distinct relativistic effects by the DS curves over the present small K^2 region are noticed with respect to the HS curves. One should pay special attention to the usual HS result ($\alpha=1$, solid curve), which is very close to the experiment and one of the present relativistic results (solid circles) at $K^2=0.04$–1 of the physical region, but deviates from the two experimental points clearly at $K^2=0.02$–0.04 corresponding to 1.4°–2.3° at 300 eV, although there may be a relatively large experimental error at these points.

In Fig. 2 we identify, furthermore, the relativistic effects with the DF and HF calculations. The present relaxation effect must be involved for the exact consideration of the nonlocal exchange terms, each of which is related to all the single-electron orbitals in one SCF potential. For the $4p$-$5s$ excitation of krypton we cannot separate the relaxation from the exchange potential in order to find the GOS in which only the nonlocal exchange terms are involved. The necessity of the present DS calculation and the advantage of the combined approach of the DF and DS theories at the SCF level are therefore shown clearly. In the $2p$-$3s$ (inner-shell) excitation of sodium [7], however, this separation in the DF approach has been realized and strong relativistic effects

over a wider K^2 region (0–2.0) are noticed. Finally, since the present relaxation effect is estimated approximately, a complete calculation of the multiconfiguration interaction is strongly recommended to verify the present GOS results, particularly, at $K^2=0.02$. At the same time, more detailed measurements are needed.

Financial support for this work was provided in part by the National Natural Science Foundation of China, the National Education Committee of China, and the Center for Molecular Science at Korea Advanced Institute of Science and Technology. Q.S. would like to express his thanks to Professor Yang-Soo Chung.

[1] 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).
[2] Padma and P.C. Deshmukh, Phys. Rev. A **46**, 2513 (1992).
[3] A. Z. Msezane and I. A. Sakmar, Phys. Rev. A **49**, 2405 (1994).
[4] E. N. Lassettre, A. Skerbele, and M. A. Dillon, J. Chem. Phys. **50**, 1829 (1969).
[5] J. F. Williams and Andris T. Stelbovics, J. Phys. B **30**, 439 (1997).
[6] Y. Qu, X. Tong, and J. Li, Acta Phys. Sin. **44**, 1720 (1995).
[7] Q. Shi, H. Cho, K. Xu, and J. Li (unpublished).
[8] Z. Chen and A. Z. Msezane, Phys. Rev. A **50**, 3517 (1994).
[9] E. Clementi and C. Roetti, At. Data Nucl. Data Tables **14**, 177 (1974).
[10] M. Vijayakumar, N. Vaidehi, and M. S. Gopinathan, Phys. Rev. A **40**, 6834 (1989).
[11] T. Kato, Commun. Pure Appl. Math. **10**, 151 (1957).
[12] J. C. Slater, Phys. Rev. **81**, 385 (1951).