Generalized oscillator strengths for the 3*d* electrons of Cs, Ba, and Xe: Effects of spin-orbit-activated interchannel coupling

M. Ya. Amusia,[1,2] L. V. Chernysheva,[2] Z. Felfli,[3] and A. Z. Msezane[3]
[1]*Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel*
[2]*A. F. Ioffe Physical- Technical Institute, St. Petersburg 194021, Russia*
[3]*Center for Theoretical Studies of Physical Systems, Clark Atlanta University, Atlanta, Georgia 30314, USA*
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This paper investigates for the first time ever the use of the spin-orbit interaction to probe correlation effects in the generalized oscillator strength (GOS) of the 3*d* subshells of Cs, Ba, and Xe, viz. we study how the GOSs of the Cs, Ba, and Xe $3d_{3/2}$ and $3d_{5/2}$ levels are affected by the intradoublet correlations. The calculations are carried out using the Hartree-Fock approximation and within the framework of a modified version of the spin-polarized random phase approximation with exchange, which takes into account multielectron correlation effects. The effects of relaxation of the excited electrons due to the creation of the 3*d* vacancy are also accounted for. Our GOS results for Cs, Ba, and Xe, obtained for values of the momentum transfer q from 0 to 4 a.u. and energy transfer ω from 0.1 to 8 Ry, demonstrate the strong interaction between the components of the spin-orbit doublet of the 3*d* electrons in Cs, Ba, and Xe. This leads to the appearance of an additional maximum in the GOS for the $3d_{5/2}$ subshell, due to the action of the $3d_{3/2}$ electrons. Additionally, for the dipole transition in Cs and Ba, the $3d_{5/2}$ subshell perturbs the $3d_{3/2}$ strongly for q=4 a.u. only, while in Xe, the perturbation is at the ω value of approximately 58 Ry for all considered q values. It is concluded that the intradoublet correlations are generally very important in the dipole, monopole, and quadrupole transitions, but particularly in the dipole transition.

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I. INTRODUCTION

Multielectron correlation effects play an important role in atomic collisions; therefore, the appropriate theoretical methods must include electron-electron correlations for realistic predictions. These many-body effects include correlation in the initial and final ionic discrete states and in the final continuum state, often referred to as interchannel coupling [1]. Stimulated by the observation of a structure in the Xe $3d_{5/2}$ experimental investigation of the photoionization cross section just above the $3d_{3/2}$ threshold [2], an aspect of interchannel coupling has been realized [3], known as spin-orbit-activated interchannel coupling; it results only through the spin-orbit splitting of inner-shell thresholds.

In this paper, we investigate the generalized oscillator strengths (GOSs) for the 3*d* electrons of Cs, Ba, and Xe using nonrelativistic one-electron wave functions and including multielectron correlations within the framework of the random phase approximation with exchange (RPAE) and some of its generalized versions, e.g., generalized RPAE (GRPAE) [4]. Different calculations have established that multielectron correlations can be accounted for with sufficient accuracy within the RPAE and GRPAE. The nonrelativistic approximation has proven to be quite sufficient, even in such a heavy atom as Xe [3,5]; thereby demonstrating the adequacy of a nonrelativistic approach which is used here.

The GOS was introduced by Bethe [6] in the context of high-energy electron impact scattering. The importance of the GOS in the study of electron impact phenomena was reviewed by Inokuti [7]. The GOS is important in radiation physics, plasma research, laser development, determining the correct spectral assignments [8], as well as in the normalization of the measured relative and understanding of electron differential cross sections (DCSs), particularly at small scattering angles [9]. It is also useful for obtaining integral cross sections and optical oscillator strengths (OOSs), and for probing the intricate nature of the valence-shell and inner-shell electron excitation [10]. The GOS, as a function of the momentum transfer squared (q^2), is characterized by a complex structure of minima [11], which essentially manifest the properties of the radial parts of the initial and final state wave functions. The difficulty in reliably measuring the electron DCSs at small scattering angles, including zero and the characteristic multiple minima, as well as of extrapolating the measured DCSs to $\theta=0°$ and 180° to obtain the integral cross sections, still continues to plague experimenters [12,13].

The role of electron correlations in the GOSs was previously investigated by Amusia and collaborators [14–17] by using the RPAE. In [14], they demonstrated the importance of intrasubshell correlations, while in [15,16] the significance of intersubshell correlation was revealed.

Recently, there has been a growing interest in the study of the GOSs from various theoretical perspectives, including correlation effects [18], coupling effects [19], inter-shell correlations [20], inter-shell transitions between two open shells of atoms [21], and minima and their approach to the high-energy limit [11]. Fan and Leung [10] measured *inter alia* the lowest nondipole discrete transition in Ar, which was subsequently attributed, contrary to the experimental interpretation, to the combined monopole, the dominant component, and quadrupole contributions [22]. The measured GOS for the Na $2p^6 3s\ ^2S \rightarrow 2p^5 3s^2\ ^2P^o$ transition [23] has been contrasted with results obtained in a second-quantization formalism used together with the spin-polarized techniques of the RPAE spin-polarized RPAE (SPRPAE) [4] to investigate correlation effects [20]. The calculation [20] has also obtained good agreement with measurement [24] for the multiplet oscillator strengths for the C $2s^22p^2\ ^3P \rightarrow 2s2p^3\ ^3P^o$ and $2s^22p^2\ ^3P \rightarrow 2s2p^3\ ^3D^o$innershell transitions. Experimental absolute GOSs for the $5s$, $5s'$, and $5p$ transitions of Kr have been determined [25]. When the angular resolution and pressure effects were considered, the measured position of the GOS minimum for the $5s+5s'$ transitions was found to agree excellently with the theoretical determination [26].

Recent experimental and theoretical investigations have revealed that multiple minima characterize the GOSs of the noble gas atoms and the alkali-metal atoms in the region $q^2 \rightarrow 0$. Thus, these minima may create problems in the extrapolation of the GOS function [27,28] to the optical limit, $q^2=0$, in the determination of precise dipole oscillator strengths.

Here, we study the action of the $3d_{3/2}$ electrons upon those of the $3d_{5/2}$ in Cs, Ba, and Xe through the GOS, viz. the effect of the spin-orbit interchannel coupling. Section II presents the theory briefly, while Sections III and IV give the results and the summary and conclusion, respectively.

II. THEORY

The inelastic scattering cross sections of fast electrons, or other charged particles incident upon atoms or molecules, are expressed via the GOS $G(\omega,q)$ [6,29], which is a function of the energy ω and the momentum transferred q to the target in the collision process. The GOS is defined as [6] (atomic units are used throughout this paper)

$$G_{fi}(\omega,q) = \frac{2\omega}{q^2}\left|\sum_{j=1}^{N} \int \psi_f^*(\vec{r}_1, \ldots, \vec{r}_N) \times \exp(i\vec{q}\cdot\vec{r}_j)\psi_i(\vec{r}_1, \ldots, \vec{r}_N)d\vec{r}_1 \ldots d\vec{r}_N\right|^2 \quad (1)$$

where N is the number of atomic electrons and $\psi_{i,f}$ are the atomic wave functions in the initial and final states with energies E_i and E_f, respectively, and $\omega=E_f-E_i$. Because the projectile is assumed to be fast, its wave functions are plane waves and its mass, M, enters the GOS indirectly, namely via the energy and momentum conservation law:

$$\frac{p^2}{2M} - \frac{(\vec{p}-\vec{q})^2}{2M} = \omega. \quad (2)$$

Here $\vec{p}$ is the momentum of the projectile. It follows from the GOS definition [Eq. (1)] that when $q=0$, the GOS coincides with the OOS or is simply proportional to the photoionization cross section (see for example [29]), depending upon whether the final state is a discrete excitation or belongs to the continuous spectrum. The energy ω enters the GOS either via a factor in Eq. (1) or indirectly, via the energy E_f of the final state $|f\rangle$.

In the one-electron Hartree-Fock (HF) approximation Eq. (1) simplifies considerably, reducing to

$$g_{fi}^L(q,\omega_{fi}) = \frac{2\omega_{fi}}{q^2}\left|\int \phi_f^*(\vec{r})j_L(qr)P_L(\cos\vartheta)\phi_i(\vec{r})d\vec{r}\right|^2 = \frac{2\omega_{fi}}{q^2}|\langle f|j_L(qr)|i\rangle|^2 \quad (3)$$

where $\phi_{f,i}(\vec{r})=R_{n_{f,i}}Y_{l_{f,i},m_{f,i}}(\theta_{\vec{r}},\varphi_{\vec{r}})\chi_{s_{f,i}}$ are the HF one-electron wave functions with their radial, angular, and spin parts, respectively, $j_L(qr)$ is the spherical Bessel function, $P_L(\cos\vartheta)$ is the Legendre polynomial and $\cos\vartheta=\vec{q}\cdot\vec{r}/qr$. The excitation energy of the $i-f$ transition is denoted as ω_{fi}. The principal quantum number, the angular momentum, its projection, and spin quantum numbers of the initial i and final f states are denoted by $n_{f,i}$, $l_{f,i}$, $m_{f,i}$, and $s_{f,i}$, respectively.

To take into account the many-electron correlations in the RPAE, the following system of equations was solved:

$$\langle f|A_L(q,\omega_{fi}^R)|i\rangle = \langle f|j_L(qr)|i\rangle + \left(\sum\int_{n'\le F,k'>F} - \sum\int_{n'<F,k'\le F}\right) \times \frac{\langle k'|A_L(q,\omega_{fi}^R)|n'\rangle\langle n'f|U|k'i\rangle_L}{\omega_{fi}^R - \epsilon_{k'} + \epsilon_{n'} + i\eta(1-2n_{k'})} \quad (4)$$

Here, $\le F(>F)$ denotes occupied (vacant) HF states, ϵ_n are the one-electron HF energies, $\eta \rightarrow +0$ and $n_k=l(0)$ for $k \le F(>F)$; $\langle nf|U|ki\rangle_L=\langle nf|V|ki\rangle_L-\langle nf|V|ik\rangle_L$ is the L component of the matrix elements of the Coulomb interelectron interaction V. It is seen that the system of equations for each total angular momentum of an excitation L is separate. The procedure of solving this equation is described in detail in [4,30]. Note that the excitation energy of the $i-f$ transition in RPAE ω_{fi}^R is different from the HF value $\omega_{fi}=\epsilon_f-\epsilon_i$. The procedure of calculating ω_{fi}^R is also described in [4,30].

A relation similar to Eq. (3) determines the GOSs in RPAE $G_{fi}^L(q,\omega_{fi}^R)$:

$$G_{fi}^L(q,\omega_{fi}^R) = \frac{2\omega_{fi}^R}{q^2}|\langle f|A_L(q,\omega_{fi}^R)|i\rangle|^2, \quad (5)$$

Here, $\langle f|$ and $|i\rangle$ are the final and initial HF states, respectively. Using these formulas, the GOSs were calculated for dipole $L=1$, monopole $L=0$ and quadrupole $L=2$ components.

The operator of the interaction between fast charged particles and atomic electrons can also be represented in another form than $\hat{A}(q)=\hat{A}^r(q)\equiv\exp(i\vec{q}\cdot\vec{r})$. This is anologous to the case of photoionization and can be called *length* form. The other one is similar to the velocity form in photoionization and looks like [30]

$$\hat{A}^v(\omega,q) = [\exp(i\mathbf{q}\cdot\mathbf{r})(\mathbf{q}\cdot\boldsymbol{\nabla} - \mathbf{q}\cdot\overleftarrow{\boldsymbol{\nabla}})\exp(i\mathbf{q}\cdot\mathbf{r})], \quad (6)$$

where the upper arrow in $\overleftarrow{\boldsymbol{\nabla}}$ in Eq. (6) implies that the function being operated on is standing to the left-hand side.

III. RESULTS

Figure 1 shows the dipole GOSs for Cs 3*d* calculated using the SPRPAE and HF approximations for values of $q = 0.1$, 2.0, and 4.0 a.u. Near their thresholds, the GOSs for both $3d_{5/2}$ and the $3d_{3/2}$ are characterized by maxima for all q values studied in both SPRPAE and HF. Intradoublet correlations have dramatic effects upon the GOSs of the Cs $3d_{5/2}$, reducing the heights of the HF maxima as well as introducing additional maxima under the $3d_{3/2}$ peaks. For the GOSs of Cs $3d_{3/2}$ the spin-orbit interaction, apart from reducing the magnitude of the GOS maxima calculated in HF, distorts only the peak at $q=4$ a.u. and shifts its position to a higher value of ω.

The dipole GOSs for Ba 3*d* calculated in SPRPAE and HF are displayed in Fig. 2. Intradoublet correlations have even more dramatic effects upon the 3*d* subshell of Ba. Comparing the two results (SPRPAE and HF), we see that the spin-orbit interaction generates strong maxima in the GOSs of the $3d_{5/2}$ subshell, whose positions move toward threshold, as q increases from 0.1 through 4.0 a.u. Furthermore, as in Cs, the GOS for the $3d_{3/2}$ gains an additional maximum only at $q=4$ a.u., due to the spin-orbit interaction.

Interestingly, the HF GOSs for both the $3d_{5/2}$ and $3d_{3/2}$ exhibit no structure, clearly showing the importance of RPAE effects in Ba 3*d*.

For the Xe $3d_{5/2}$ subshell (results are given in Fig. 3), the intradoublet correlations manifest themselves as secondary

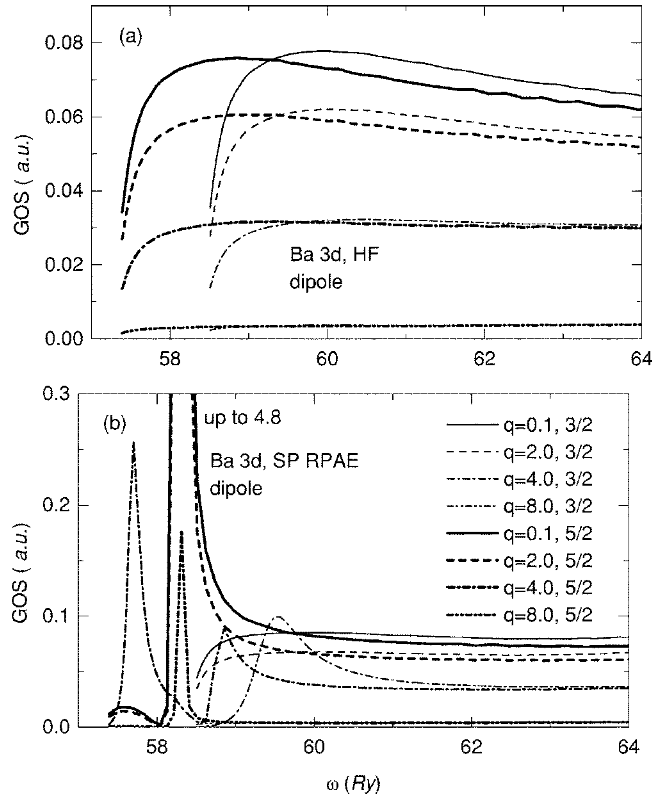

FIG. 2. Dipole generalized oscillator strengths for Ba 3*d* subshell: (a) HF approximation and (b) SPRPAE results.

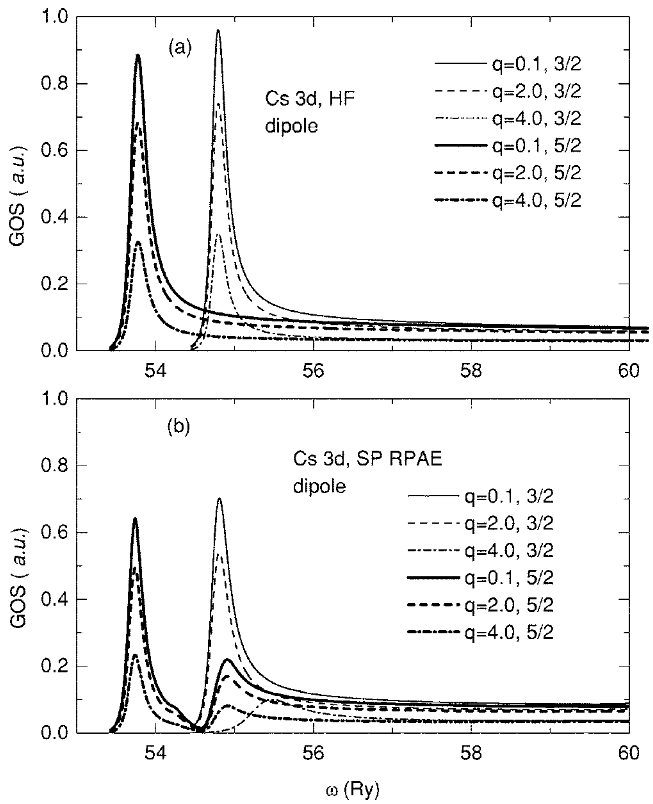

FIG. 1. Dipole generalized oscillator strengths for Cs 3*d* subshell: (a) HF approximation and (b) SPRPAE results.

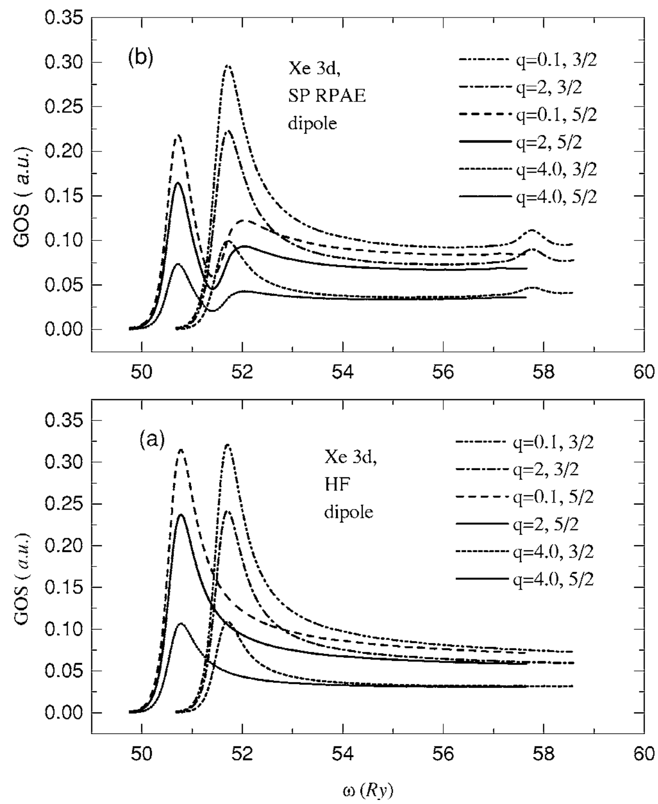

FIG. 3. Dipole generalized oscillator strengths for Xe 3*d* subshell: (a) HF approximation and (b) SPRPAE results.

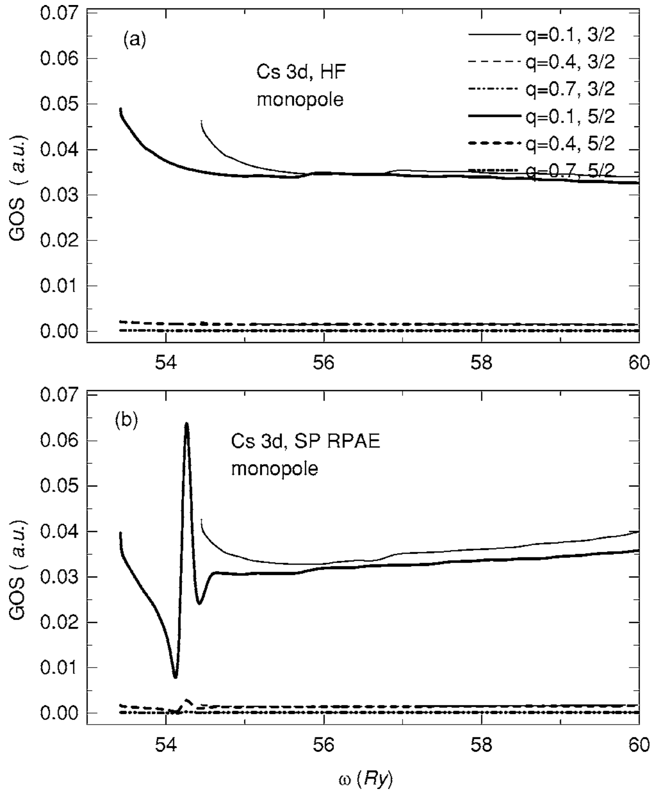

FIG. 4. Monopole generalized oscillator strengths for Cs $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

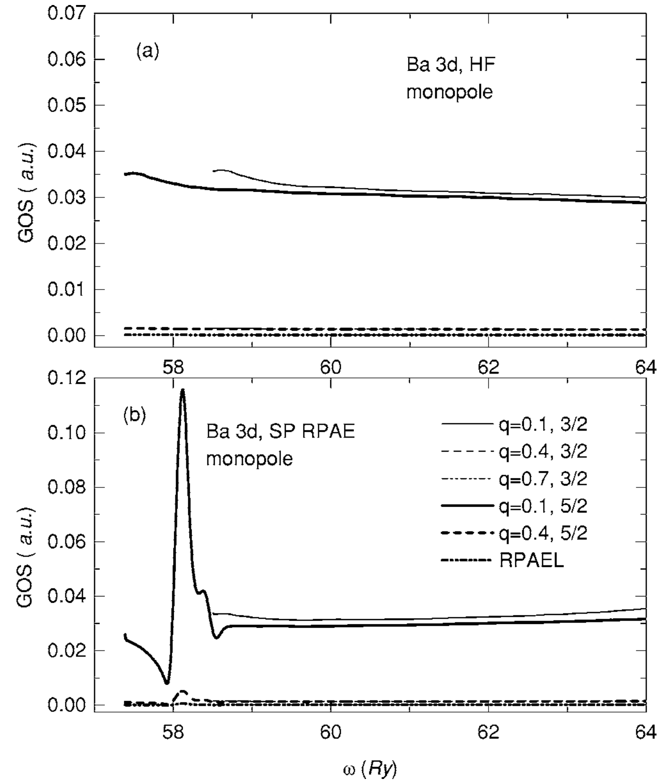

FIG. 5. Monopole generalized oscillator strengths for Ba $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

peaks under the Xe $3d_{3/2}$ maxima and are as dramatic as found in Cs. These second maxima are induced by the spin-orbit interaction, causing the Xe $3d_{3/2}$ to perturb the $3d_{5/2}$ resulting in the second maxima at roughly the positions of the Xe $3d_{3/2}$ subshell maxima for all the values of q considered, viz. 0.1, 2, and 4. Contrary to the cases of Cs and Ba, the $3d_{5/2}$ perturbation of the $3d_{3/2}$ through the spin-orbit interaction is observable even at relatively high values of ω. These effects show up as maxima in the GOSs at around $\omega = 58$ Ry for all the values of q examined here.

The monopole GOSs for Cs $3d$ calculated in SPRPAE and HF are shown in Fig. 4. For the Cs $3d$ monopole transition, intradoublet correlation effects are significant only at small q values, viz. $q=0.1$ a.u. They introduce two extra minima near threshold only in the $q=0.1$ a.u. curve; there is also a small maximum in the $q=0.7$ a.u. curve. For all other values of q correlation effects are insignificant. The monopole GOSs for the Ba $3d$ calculated in SPRPAE and HF are displayed in Fig. 5. Here, the effects of intradoublet correlations are seen to be similar to those in Cs $3d$.

In the case of the Xe monopole transition, contrary to the Cs and Ba results, the intradoublet correlations cause the development of a less dramatic but pronounced structure, affecting only the $3d_{5/2}$ subshell at $q=0.1$ a.u. The results are shown in Fig. 6. As in previous cases, the HF GOSs for both the $3d_{5/2}$ and the $3d_{3/2}$ of Xe show no structure, again demonstrating the importance of the RPAE effects, but this time at only small q values.

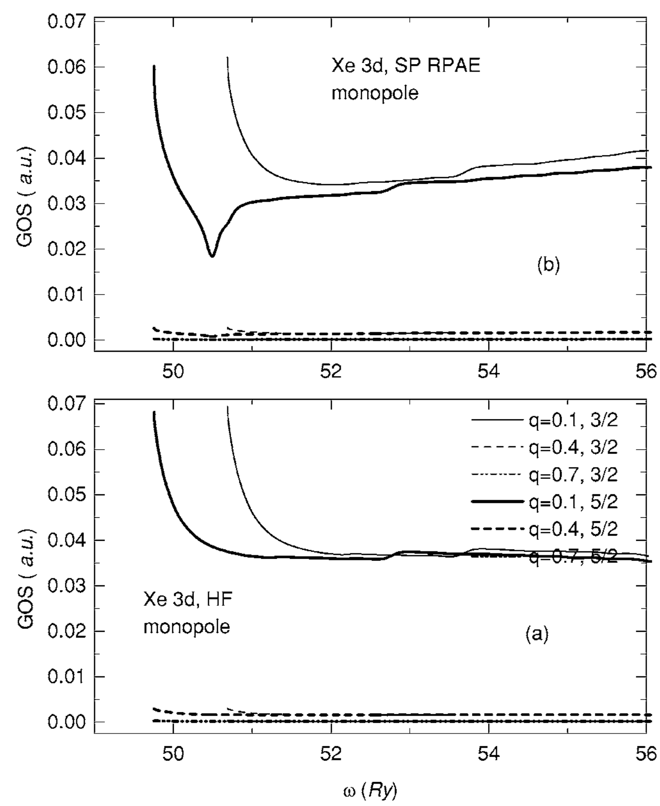

FIG. 6. Monopole generalized oscillator strengths for Xe $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

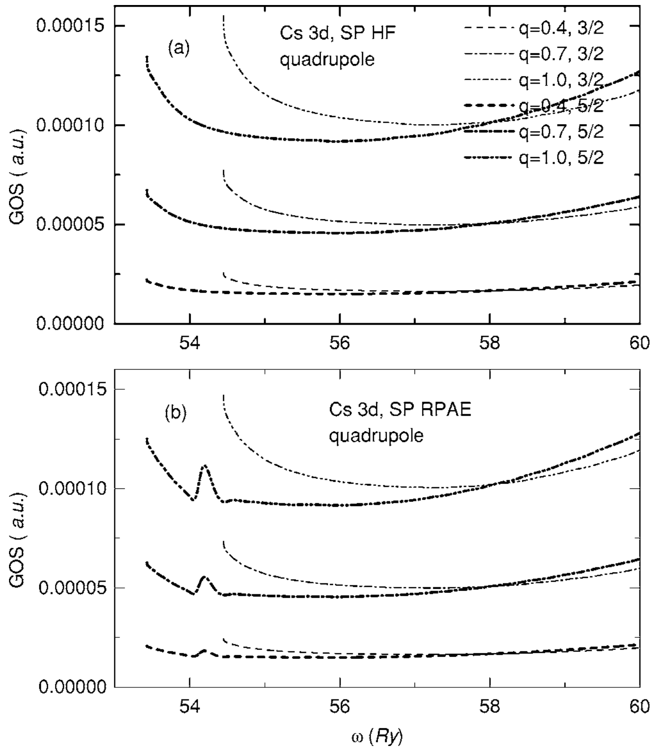

FIG. 7. Quadrupole generalized oscillator strengths for Cs $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

Figure 7 shows quadrupole GOSs for Cs $3d$ evaluated in SPRPAE and HF. For the quadrupole transition in Cs $3d$, the spin-orbit interaction affects only the Cs $3d_{5/2}$ GOSs; their effects increase with increasing momentum transfer q. This interaction shows in the form of a maximum in the GOS near threshold. The intradoublet correlations have a similar effect on the quadrupole GOSs for Ba $3d$ as in Cs $3d$; the results are shown in Fig. 8. However, for the Xe $3d$, these effects are almost insignificant, as seen in Fig. 9.

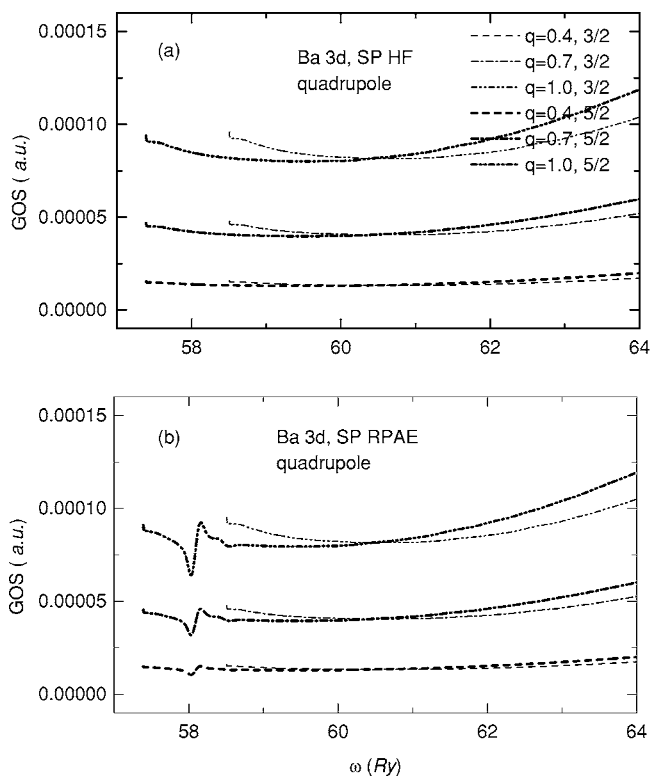

FIG. 8. Quadrupole generalized oscillator strengths for Ba $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

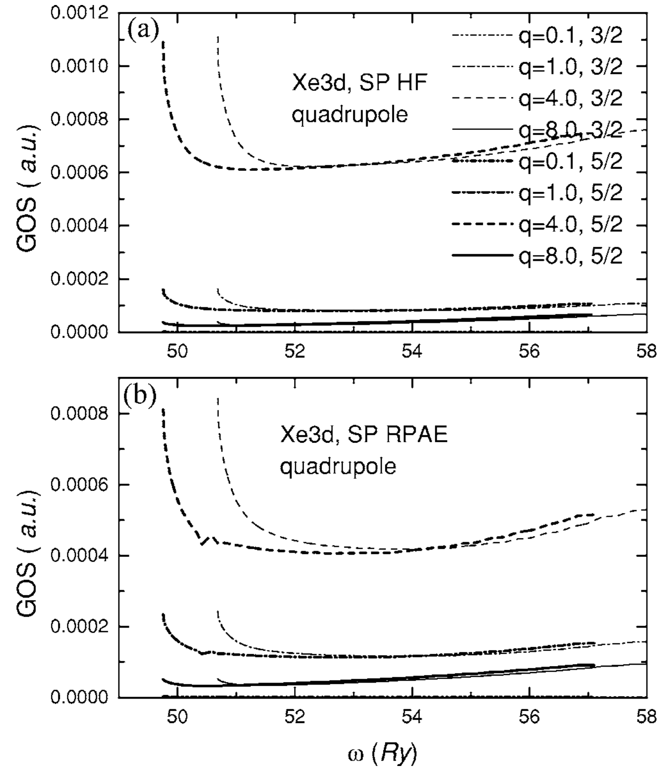

FIG. 9. Quadrupole generalized oscillator strengths for Xe $3d$ subshell: (a) HF approximation and (b) SPRPAE results.

IV. SUMMARY AND CONCLUSION

Our GOS results for the $3d$ subshells of Cs, Ba, and Xe—obtained for values of the momentum transfer q from 0 to 4.0 *a.u.* and energy transfer ω from 0.1 to 8 Ry above the ionization threshold—demonstrate the importance of the intradoublet correlations viz. the strong interaction between the components of the spin-orbit doublet of the $3d$ electrons in Cs, Ba, and Xe. This leads to the appearance of an additional maximum in the GOS for the $3d_{5/2}$ subshell due to the action of the $3d_{3/2}$ electrons.

The dipole GOSs for both the $3d_{5/2}$ and $3d_{3/2}$ subshells of Cs, Ba, and Xe are characterized by maxima near their respective thresholds, and intradoublet correlations have dramatic effects upon the GOSs for the $3d_{5/2}$; the largest peak corresponding to the smallest q-value. Intradoublet correlations induce significant structures in the GOSs for the monopole transitions in both Cs and Ba $3d_{5/2}$ only at $q=0.1$ a.u. For the quadrupole transitions in Cs and Ba, correlation effects cause the appearance of a peak in the GOSs for the $3d_{5/2}$, but hardly any in the $3d_{3/2}$ and Xe $3d$. The strength of the correlations increases with q and becomes most important at $q=1.0$ a.u.

In conclusion, we have used HF and SPRPAE to demonstrate the importance of the spin-orbit activated intrachannel coupling in the GOSs of the $3d$ subshells of Cs, Ba, and Xe. The intradoublet correlations are generally very important in the dipole, monopole and quadrupole transitions near the threshold region, but particularly in the dipole transitions. When the spin-orbit interaction is activated, it provides a mechanism for modifying the $3d_{5/2}$ HF GOSs by splitting them into two distinct components. The Cs $3d_{3/2}$ HF dipole GOS is broadened and shifted in position only at $q=4.0$ a.u.; while for the Ba $3d_{3/2}$, the spin-orbit interaction defines a distinct GOS peak. For the monopole and quadrupole transitions in both Cs and Ba $3d$, correlation effects are important near threshold, resulting in the appearance of distinct peaks in the $3d_{5/2}$ at $q=0.1$ *a.u.* only.

Experimentally, it is difficult to observe contributions of monopole and quadrupole transitions, since they are much smaller than the dipole contribution. However, Fan and Leung [10] did measure the combined contribution from both the monopole and quadrupole transitions to the GOS. So, it is expected that similar measurements will be carried out in the future. Nevertheless, the GOSs for the dipole transitions are, by themselves, sufficiently interesting to be a subject of experimental investigation.

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[1] U. Fano, Phys. Rev. **178**, 131 (1969)
[2] A. Kivimäki, U. Hergenhahn, B. Kempgens, R. Hentges, M. N. Piancastelli, K. Maier, A. Rudel, J. J. Tulkki, and A. M. Bradshaw, Phys. Rev. A **63**, 012716 (2000).
[3] M. Ya. Amusia, L. V. Chernysheva, S. T. Manson, A. M. Msezane, and V. Radojević, Phys. Rev. Lett. **88**, 093002 (2002).
[4] M. Ya. Amusia, *Atomic Photoeffect* (Plenum, New York, London, 1990).
[5] B. Schmidtke, M. Drescher, N. A. Cherepkov, and U. Heinzmann, J. Phys. B **33**, 2451 (2000)
[6] H. A. Bethe, Ann. Phys. **5**, 325 (1930).
[7] M. Inokuti, Rev. Mod. Phys. **43**, 297 (1971).
[8] C. C. Turci, J. T. Francis, T. Tyliszczak, G. G. de Souza, and A. P. Hitchcock, Phys. Rev. A **52**, 4678 (1995).
[9] Z. Felfli, A. Z. Msezane, and D. Bessis, Phys. Rev. Lett. **81**, 963 (1998).
[10] X. W. Fan and K. T. Leung, Phys. Rev. A **62**, 062703 (2000).
[11] N. B. Avdonina, D. Fursa, A. Z. Msezane, and R. H. Pratt, Phys. Rev. A **71**, 062711 (2005)
[12] B. Predojevic, D. Sevic, V. Pejcev, B. P. Marinkovic, and D. M. Filipovic, J. Phys. B **38**, 3489 (2005), and references therein.
[13] B. Predojevic, D. Sevic, V. Pejcev, B. P. Marinkovic, and D. M. Filipovic, J. Phys. B **38**, 1329 (2005), and references therein.
[14] M. Ya. Amusia, N. A. Cherepkov, L. V. Chernysheva, and S. I. Sheftel, Phys. Lett. **40A**, 1 (1972).
[15] M. Ya. Amusia, N. B. Berezina, and L. V. Chernysheva, Phys. Lett. **51A**, 101 (1975).
[16] M. Ya. Amusia, V. K. Ivanov, and S. A. Sheinerman, J. Phys. B **9**, 1537 (1976).
[17] M. Ya. Amusia, L. V. Chernysheva, Z. Felfli, and A. Z. Msezane, Phys. Rev. A **64**, 032711 (2001) and references therein.
[18] Z. Chen and A. Z. Msezane, J. Phys. B **35**, 815 (2002).
[19] Z. Chen and A. Z. Msezane, J. Phys. B **33**, 2135 (2000).
[20] Z. Chen and A. Z. Msezane, Phys. Rev. A **70**, 032714 (2004) and references therein.
[21] Z. Chen and A. Z. Msezane, Phys. Rev. A **72**, 050702(R) (2005).
[22] A. Z. Msezane, Z. Felfli, M. Ya. Amusia, Z. Chen, and L. V. Chernysheva, Phys. Rev. A **65**, 054701 (2002).
[23] C. E. Bielschowsky, C. A. Lucas, G. G. B. de Souza, and J. C. Nogueira, Phys. Rev. A **43**, 5975 (1991).
[24] W. L. Wiese, J. R. Fuhr, and T. M. Deters, *Atomic Transition Probabilities of Carbon, Nitrogen, and Oxygen. A Critical Data Compilation. Journal of Physical and Chemical Reference Data Monograph No. 7* (American Chemical Society and American Institute of Physics for the National Institute of Standards and Technology, Gaithersburg, MD, 1996).
[25] W. B. Li, L. F. Zhu, X. J. Liu, Z. S. Yuan, J. M. Sun, H. D. Cheng, Z. P. Zhong, and K. Z. Xu, Phys. Rev. A **67**, 062708 (2003).
[26] Z. Chen and A. Z. Msezane, J. Phys. B **33**, 5397 (2000).
[27] T. Ester and J. Kessler, J. Phys. B **27**, 4295 (1994).
[28] T. Y. Suzuki, H. Suzuki, S. Ohtani, B. S. Min, T. Takayanagi, and K. Wakiya, Phys. Rev. A **49**, 4578 (1994); *ibid.* **53**, 3081 (1996).
[29] L. D. Landau and E. M. Lifschitz, *Quantum Mechanics, Non-Relativistic Theory* (Pergamon, Oxford 1962).
[30] M. Ya. Amusia and L. V. Chernysheva, *Computation of Atomic Processes* (IOP, Bristol, 1997).