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# Addendum to "Ab initio treatment of final-state spin-orbit interactions: Photoionization of the 6s electron in cesium"

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# Addendum to "*Ab initio* treatment of final-state spin-orbit interactions: Photoionization of the 6*s* electron in cesium"

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A previous calculation of the cesium $6s \to \epsilon p$ photoionization cross section is extended to include interchannel coupling to the $5p \to \epsilon d$ photoionization channels. For photon energies in the range 0.35 a.u. $\lesssim \hbar\omega \lesssim$ 1.12 a.u., i.e., just above the near-threshold cross-section minimum, the 6s photoionization cross section is found to be dominated by $5p \to 5d$ resonance transitions. No experimental data exist for this energy region.

The 6s photoionization cross section of atomic cesium has been studied extensively by both experimentalists[1-4] and theorists[5-13] near threshold, in the vicinity of the cross-section minimum. However, there appear to be no experimental data and only a single other *ab initio* theoretical calculation[12] of the photoionization cross section beyond about 3 eV above threshold. We report here an extension of our previous *ab initio* theoretical calculations (Ref. 13, hereafter referred to as I) to include interchannel interactions over the energy region from threshold up to 1 a.u. (27.21 eV) above. We find that above the near-threshold minimum the cross section is dominated by $5p \to 5d$ resonance transitions.

In I we presented *ab initio* theoretical calculations of the cross section, photoelectron angular distribution, and photoelectron spin polarization resulting from photoionization of the 6s electron in cesium. We started from a basis of nonrelativistic Hartree-Fock wave functions and treated final-state spin-orbit interactions in the Breit-Pauli approximation[14] exactly. Only $6s \to \epsilon p$ transitions were considered over the photoelectron energy range $0 \leq \epsilon \leq 1$ a.u. Our results agreed with experimental measurements just above threshold, but our cross sections beyond 2 eV above threshold failed to rise as rapidly as the experimental measurements,[3,4] some semiempirical calculations,[8,9] and the nonrelativistic random-phase approximation (RPA) calculation of Amusia and Cherepkov[12] (which did not treat relativistic interactions, but which included interchannel coupling between the 6s and 5p subshells).

We report here *ab initio* calculations of the 6s photoionization cross section in cesium which include final-state interchannel coupling between the 6s and 5p subshell channels as well as the most important final-state spin-orbit interactions. Specifically, we have included the following six channels in our calculations:

$$\mathrm{Cs}\,5p^6 6s(^2S_{1/2}) + \hbar\omega \to \mathrm{Cs}^+\,5p^6(^1S_0)\epsilon p(^2P_{3/2,1/2}) \tag{1}$$

$$\to \mathrm{Cs}^+\,5p^5 6s(^3P)\epsilon d(^2P_{3/2,1/2}) \tag{2}$$

$$\to \mathrm{Cs}^+\,5p^5 6s(^1P)\epsilon d(^2P_{3/2,1/2})\,. \tag{3}$$

The final-state $J=\frac{3}{2}$ and $\frac{1}{2}$ channels, of course, do not interact. For each value of $J$, the final-state interactions that we have included in our calculations are indicated in Table I. Here the spin-orbit interactions are the same as were included in I. The Coulomb interactions are those induced by the electrostatic interaction operator $\sum_{i>j} r_{ij}^{-1}$. There are no Coulomb interactions along the diagonals in Table I since we use a basis of Hartree-Fock continuum wave functions calculated in the field of the appropriate ion, whose orbital wave functions are those of the neutral cesium atom (i.e., we have used the so-called $V^{N-1}$ potential in the "frozen-core" approximation[15]). The binding energies we have used are the Hartree-Fock orbital eigenenergies.

Our results for the 6s photoionization cross section over the energy range from threshold to 1 a.u. (27.21 eV) above are presented in Fig. 1(c). For comparison, we have plotted on the same energy scale in Figs. 1(a) and 1(b) the nonrelativistic Hartree-Fock predictions for the continuum oscillator strength $df/dE$ for channels (2) and (3), respectively.[16] Note that the discrete members of these channels have been plotted also, following the prescription of Fano and Cooper.[17,18] As compared with the calculations reported in (I) we find

 

TABLE I. Block form of the final-state interaction matrix V. Each box indicates the interactions between each pair of final-state channels.

| | $5p^6\epsilon p(^2P_{1/2,3/2})$ | $5p^56s(^1P)\epsilon d(^2P_{1/2,3/2})$ | $5p^56s(^3P)\epsilon d(^2P_{1/2,3/2})$ |
|---|---|---|---|
| $5p^6\epsilon p(^2P_{1/2,3/2})$ | Spin orbit | Coulomb | Coulomb |
| $5p^56s(^1P)\epsilon d(^2P_{1/2,3/2})$ | Coulomb | 0 | Coulomb |
| $5p^56s(^3P)\epsilon d(^2P_{1/2,3/2})$ | Coulomb | Coulomb | 0 |

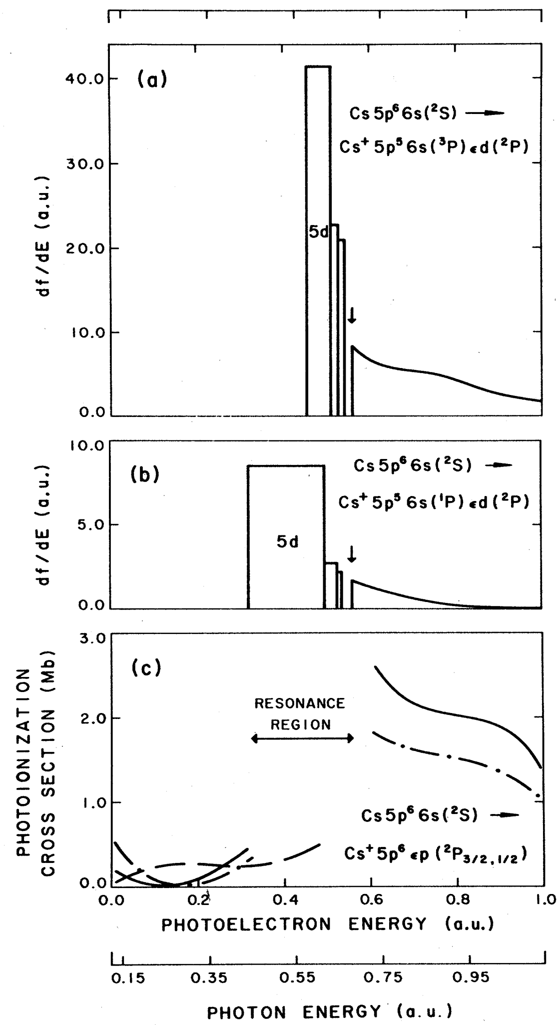


FIG. 1. (a) Oscillator-strength distribution in the Hartree-Fock approximation for the channel Cs $5p^66s(^2S) + \hbar\omega \rightarrow Cs^+ 5p^56s(^3P)\epsilon d(^2P)$. (b) Oscillator-strength distribution in the Hartree-Fock approximation for the channel Cs $5p^66s(^2S) + \hbar\omega \rightarrow Cs^+ 5p^56s(^1P)\epsilon d(^2P)$. (c) Present results for the $6s$-subshell photoionization cross section in cesium in dipole-length (solid line) and dipole-velocity (dash-dot line) approximations. The dashed line gives the RPA results of Amusia and Cherepkov (Ref. 12).

that at threshold both dipole-length and dipole-velocity cross sections are increased in value, particularly the dipole-velocity cross section. Also, the location of the minimum in the cross section is shifted to higher energies (by 2 and 4 eV, respectively, in the dipole-length and -velocity approximations). For photoelectron energies $\epsilon \gtrsim 0.3$ a.u. our calculated cross section rises rapidly under the influence of the $5p \rightarrow 5d$ resonance transitions, whose oscillator strengths are indicated in Figs. 1(a) and 1(b). We have not given any quantitative predictions in the resonance region since the resonances induce such rapid oscillations in our calculations that we do not have a sufficiently fine energy mesh to describe them. Furthermore, the resonance profiles are very sensitive to the energy locations and wave functions used for the discrete members of the channels (2) and (3); however, we have not attempted to optimize our description of these discrete states. Nevertheless, we expect that the shapes of the discrete oscillator strength distributions shown in Figs. 1(a) and 1(b) will be reflected in the $6s \rightarrow \epsilon p$ cross section in the resonance region. Above the threshold of the $d$ channels, i.e., for photoelectron energies $\epsilon \gtrsim 0.6$ a.u., the $6s$ cross section decreases smoothly from a value of approximately 2 Mb. Most of this oscillator strength in the $6s \rightarrow \epsilon p$ channel is also borrowed from the $5p \rightarrow \epsilon d$ channels. Indeed, our calculations reported in I, which ignored interchannel interactions, predict that the $6s \rightarrow \epsilon p$ cross section is less than 0.076 Mb and 0.030 Mb, respectively, in the dipole-velocity and dipole-length approximations at all energies above the near-threshold cross-section minimum.

We have also shown in Fig. 1(c) the nonrelativistic RPA calculation of Amusia and Cherepkov,[12] which includes the same interchannel interaction between the $6s$ and $5p$ subshells that we have included as well as the effect of ground-state correlations, which we have not included. Their cross section rises monotonically near threshold and does not exhibit the minimum observed experimentally. Their cross section levels off at energies where our calculation indicates a minimum, and then tends to rise, as ours does, but at energies about 0.17 a.u. higher in energy than ours. While no resonance behavior is shown in their cross section, it is possible that the location of the discrete resonances in their calculation may be quite different from that in our calculation. However, we do not know this. The major difference between our calculations and theirs appears to be our lack of ground-state correlations. It is well known that such correlations can substantially shift the location of minima in the cross section[19]; however, in general, qualitatively different cross sections are not expected. A more minor difference between our calculations and theirs is our neglect of the effect of the $5p \rightarrow \epsilon s$ transition on the $6s \rightarrow \epsilon p$ cross section. The cross section for the $5p \rightarrow \epsilon s$ transition is about an order of magnitude smaller than that for the $5p \rightarrow \epsilon d$ transitions which we have considered. While there are probably non-negligible effects of the $5p \rightarrow \epsilon s$ transition on the location of the minimum in the $6s \rightarrow \epsilon p$ cross section (since at the minimum the strong amplitudes nearly cancel), we do not expect any large effects on the cross-section values elsewhere. For this reason, and also since we have ignored ground-state correlation effects, which are probably larger, we have ignored the effect of $5p \rightarrow \epsilon s$ transitions.

We conclude that the Cs $6s$ photoionization cross section is dominated by $5p \rightarrow 5d$ resonance transition in the region just above the cross-section minimum near threshold to 1 a.u. above threshold. Experimental data for this region would be most welcome to theoreticians. Furthermore, we conclude that the exact location of the cross-section minimum near threshold is determined by a sensitive balance between final-state interchannel and spin-orbit interactions on the one hand and ground-state correlations on the other.

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[1] F. L. Mohler and C. J. Boeckner, J. Res. Natl. Bur. Stand. 3, 303 (1929).

[2] H. J. J. Braddick and R. W. Ditchburn, Proc. R. Soc. London A143, 472 (1934).

[3] G. V. Marr and D. M. Creek, Proc. R. Soc. London A304, 233 (1968).

[4] T. B. Cook, F. B. Dunning, G. W. Foltz, and R. F. Stebbings, Phys. Rev. A 15, 1526 (1977).

[5] M. J. Seaton, Proc. R. Soc. London A208, 418 (1951).

[6] E. J. McGuire, Phys. Rev. 161, 51 (1967).

[7] I. L. Beigman, L. A. Vainstein, and V. P. Shevelko, Opt. Spectros. (USSR) 28, 229 (1970).

[8] J. C. Weisheit, Phys. Rev. A 5, 1621 (1972).

[9] D. W. Norcross, Phys. Rev. A 7, 606 (1973).

[10] J. J. Chang and H. P. Kelly, Phys. Rev. A 5, 1713 (1972).

[11] G. McGinn, J. Chem. Phys. 58, 772 (1973).

[12] M. Ya Amusia and N. A. Cherepkov, Case Stud. At. Phys. 5, 47 (1975), Fig. 26d.

[13] K.-N. Huang and A. F. Starace, Phys. Rev. A 19, 2335 (1979) (referred to in the text as I). We point out here a misprint in this paper. Namely, in Eqs. (22b), (23b), and (24) as well as in the last line of Sec. IV B there should not be absolute-value signs on $M_{3/2E}$ and $M_{1/2E}$. Instead of $|M_{3/2E}|$ and $|M_{1/2E}|$ the quantities in Eq. (16) should appear, but *without* the exponential factor. Our numerical results are unaffected by these misprints.

[14] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Systems* (Springer, Berlin, 1957), Chaps. 38 and 39.

[15] H. P. Kelly, Phys. Rev. B 136, 896 (1964); 144, 39 (1966).

[16] For continuum energies, the photoionization cross section $\sigma$ is related to the continuum oscillator strength $df/dE$ as follows: $\sigma = 2\pi^2 c^{-1}\, df/dE = (4.033\,64\ \mathrm{Mb})df/dE$.

[17] U. Fano and J. W. Cooper, Rev. Mod. Phys. 40, 441 (1968), Sec. 2.4. See especially pp. 446 and 447 and Fig. 1.

[18] Note that, as explained in Ref. 17, the histograms representing the discrete oscillator strength should in principle extrapolate smoothly onto the continuum oscillator strength distribution above threshold. The reason our histogram plot in Fig. 1(a) does not extrapolate smoothly onto the continuum is due to our use of the Hartree-Fock $5p$ orbital eigenenergy rather than a more accurate approximation to the binding energy for channel 2. On the other hand, a more accurate approximation to the binding energy would have introduced the complication of nonorthogonality of the core orbitals corresponding to the three channels in Eqs. (1)–(3).

[19] See, e.g., C. D. Lin, Phys. Rev. A 9, 171 (1974).