Photoionization of the 7*d* excited state of cesium

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The photoionization cross section for the excited 7*d* state of atomic cesium has been calculated in the Hartree-Fock approximation. The results show excellent agreement with a recent measurement.

I. INTRODUCTION

The photoionization of excited states of atoms offers the chance to study systems of large physical extent. While the interactions are the same as for ground-state atoms, the manifestations of these interactions differ considerably for excited states, leading to theoretical predictions of new phenomenology,[1-4] notably the existence and generality of many more zeros in the dipole matrix elements than are found for ground states. There theoretical predictions have not yet been tested. As a matter of fact, there is very little experimental work on excited-state photoionization, an even less where absolute cross sections of excited electrons have been measured over an appreciable energy range.[5] This latter point is of particular significance for assessing theory; measurements at a single energy, while useful, are simply not sufficient for testing calculations.

Of particular interest are the excited *d* states of Cs where a number of interesting theoretical predictions have been made, including multiple minima in the $d \rightarrow f$ cross sections and minima in the $d \rightarrow p$ channels as well. A recent measurement of the Cs 7*d* cross section has been made;[6] the first absolute measurement of an excited *d* electron over a significant range of energy.

In order to assess theory, in this paper the results of Hartree-Fock (HF) calculations of the Cs 7*d* cross section are presented and compared with the measured cross section, along with central-field Hartree-Slater (HS) results.[7] We focus particularly on two points: the utility of HF calculations as quantitative predictors of excited-state photoionization cross sections; and the qualitative utility of HS calculations in the same capacity. We note that in one previous case for a much simpler system, Na 3*p*, both HF and HS gave good quantitative agreement.[5]

In Sec. II a brief review of the theory employed in the HF calculation is given. Section III presents our results and compares them with experiment and the HS cross section. Section IV presents a summary, conclusions and some final remarks.

II. BRIEF REVIEW OF THEORY

Within the framework of the electric dipole approximation, which is excellent for low-energy photons,[8] the photoionization cross section for a single electron in a subshell takes a fairly simple form.[9] In particular, for a transition $(L_cS_cnl)LS \rightarrow (L_cS_c\epsilon l')L'S$, an initial nl electron coupled to a core L_cS_c to give LS going to a final continuum $\epsilon l'$ coupled to a core L_cS_c giving $L'S$, the cross section is given by[9]

$$\sigma_{nl}^{L_cS_cL}(\epsilon) = \frac{4\pi\alpha a_0^2}{3}(\epsilon + I) \times \sum_{l',L'} (2L'+1) l_> \begin{Bmatrix} l & L & L_c \\ L' & l' & 1 \end{Bmatrix}^2 (M_{nl,\epsilon l'}^{L_cS_cLL'})^2 , \quad (1)$$

where I is the ionization potential of the nl electron in the particular state, a_0 is the Bohr radius, α is the fine-structure constant, $l_>$ is the maximum of l and l', $\{\begin{smallmatrix} a & b & c \\ d & e & f \end{smallmatrix}\}$ is the Wigner 6-*j* symbol,[10] and

$$M_{nl,\epsilon l'}^{L_cS_cLL'} = \int P_{nl}^{L_cS_cL}(r) r P_{\epsilon l'}^{L_cS_cL'}(r) dr , \quad (2)$$

where the P's are (r times) the radial wave functions for initial and final states of the electron undergoing the transition. The initial discrete wave function was obtained from a standard HF code,[11] while the final continuum HF wave function, in the field of the fully relaxed ion, was obtained from our own code.[12]

Note that the length form of the dipole matrix element, Eq. (2), may be transformed[8,13] into another form, the so-called velocity form, and these two forms must be equal for exact wave functions.[8,13] Thus equality of length and velocity results is a useful (but by no means infallible) measure of the accuracy of the HF calculation.

III. RESULTS AND DISCUSSION

HF cross sections for Cs 7*d* photoionization have been calculated for the first 20 eV above threshold. A close-up of the region where the measurements were made is presented in Fig. 1, where the experimental points and our HF cross sections are shown. The outstanding feature of this comparison is the excellent agreement between theory and experiment; the experiment error bar of each of the points intersects the theoretical curve.

At this point, it is important to reiterate that the HF calculation uses theoretical binding energies and contains no adjustable parameters, and the experiment is absolute; nothing is normalized to anything else. This makes the agreement more meaningful.

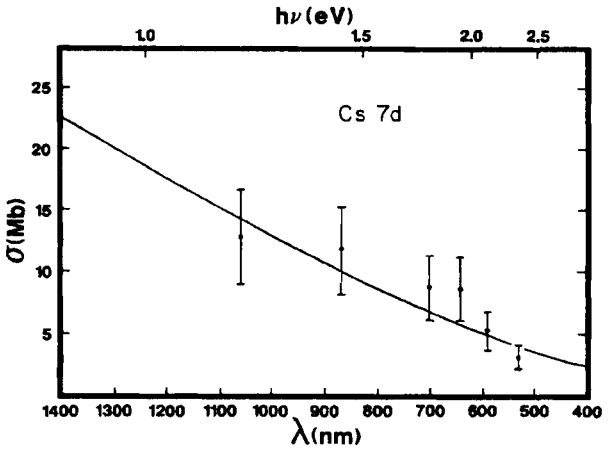

FIG. 1. Cs 7*d* photoionization cross section. The solid line is the Hartree-Fock result of this paper, and the points are the experimental results of Ref. 6. Note that only a single theoretical curve is given since the length and velocity results are virtually identical in this energy range.

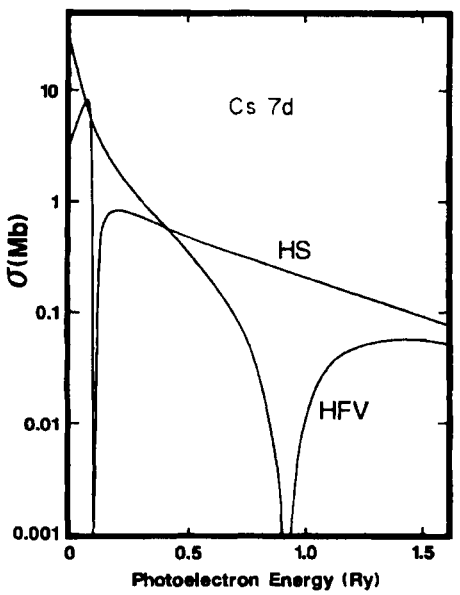

FIG. 2. Cs 7*d* photoionization cross section in Hartree-Fock velocity (HFV) approximation along with the Hartree-Slater results of Ref. 7.

In addition, only a single theoretical curve is given in Fig. 1, while, in fact, the HF calculations were performed in both length and velocity formulations. It turned out, however, that in the energy region depicted in Fig. 1, length and velocity were so close together, so as to be almost indistinguishable, differing by the order of the thickness of the curve. This agreement between length and velocity is a further indication of the accuracy of the theoretical cross section.

A comparison between HF and the central-field HS result over a broad energy range is shown in Fig. 2, where a vast quantitative difference between the two cross sections is seen. Despite the quantitative difference, several essential similarities exist. Both exhibit a $d \rightarrow f$ matrix element which is positive at threshold and changes to negative, thereby giving rise to the deep minima in the cross section as shown. Not shown in Fig. 2 is a second broad minimum, in both approximations at much higher energy. Thus both HF and HS show two zeros in the $d \rightarrow f$ channel. In addition, it is found that both have a zero in the $d \rightarrow p$, but it is not evident from Fig. 2 since the $d \rightarrow f$ channel so dominates the cross section.

The principal reason for the differences between HF and HS results involves the strength of the exchange interaction; the central-field approximation to exchange is significantly more attractive than the HF exchange, thus pulling the low-energy minimum closer to threshold. Thus, while the ϵf wave function has a shape resonance in both cases, it is much sharper and closer to threshold in HS than HF,[14] leading to the behavior shown.

Note further that only the velocity form of the HF cross section is shown in Fig. 2. As mentioned earlier, at energies much below the minimum, length and velocity are essentially equal. Near the minimum, they diverge slightly, with the length minimum being marginally below the velocity. At the larger energies, however, the length calculation is very unstable, compared to the velocity, particularly near a zero, so it is felt that the velocity form is more reliable here.[15]

It is also important to emphasize that the measured energy range,[6] which encompasses photoelectron energies from 0.037 to 0.122 Ry, where the HF results is monotone decreasing, is seen from Fig. 2 to include the HS minimum. Since HF is in good quantitative agreement with experiment in this region, it follows that HS is not, just as was found in a previous relative measurement on Cs 6*d*.[14]

IV. CONCLUDING REMARKS

The Hartree-Fock calculation has been shown to be in quite good agreement with the recent experimental measurement of the excited Cs 7*d* photoionization cross section. The Hartree-Slater result, while having the same qualitative features as the HF, was nevertheless quite far from the experimental measurements quantitatively, owing to the incorrect location of the minimum. This differs sharply from the case of Na 3*p*, where both HF and HS were in good agreement with each other and the experiment.[5]

It appears that except in special circumstances, like regions where intershell coupling might be uncharacteristically strong, the HF calculation should be a reasonably accurate predictor of excited-state photoionization cross sections. It would be very useful, however, to have some measurements near the minimum to benchmark the calculation; the minimum region is likely to be the most sensitive to the details of the calculation.

Finally we note that the very sophisticated random-phase approximation with exchange has been applied to photoionization of excited d states of Cs (although not the $7d$ state).[4] The results show good agreement with HF, even in the minimum region. This too points out the accuracy of HF calculations for excited-state photoionization cross sections.

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