Relativistic Many-Body Approach to the Photoionization of Cesium

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A relativistic many-body technique which includes particle-hole excitations of the atomic core is developed to describe the photoexcitation of alkali atoms. The technique is used to study the photoionization of cesium near threshold, where the dipole amplitudes are sensitive to both relativistic and correlation effects. The predicted photoionization amplitudes are free from the gauge ambiguities of Dirac-Fock calculations and are in good agreement with semiempirical calculations and with measurements.

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The photoionization of cesium has been of considerable interest since it was pointed out by Fano[1] more than a decade ago that one expects a high degree of spin polarization for photoelectrons produced by circularly polarized light because of the large spin-orbit interaction. A number of measurements[2-5] have been made confirming Fano's prediction and giving a quantitative understanding of low-energy photoionization of cesium. Two different approaches have been used to understand such systems theoretically. On the one hand there are the *ab inito* studies ignoring the effects of core excitations, but including spin-orbit interactions, carried out at the Hartree-Fock (HF) or Dirac-Fock (DF) level of approximation.[6-8] These calculations give at best qualitative agreement with the measurements mentioned above, provided the calculations are carried out in the Coulomb gauge; however, such calculations are strongly gauge dependent,[8] and lead to qualitatively incorrect results in other gauges. A second approach has been to introduce a model potential[9-11] which includes core-polarization effects together with the spin-orbit interaction in a nonrelativistic framework. Model-potential calculations lead to photoionization parameters free from the gauge ambiguities of HF or DF calculations. Such calculations are not entirely satisfactory since they require the use of empirical cutoff parameters in the polarization potential.

The objective of the present work is to develop a relativistic many-body method to treat both strong spin-orbit effects and correlation effects in open-shell systems such as alkali atoms. For closed-shell systems the relativistic random-phase approximation[12-13] (RRPA) has been successfully applied to understand the interplay of relativistic and correlation effects. While nonrelativistic RPA calculations[14] have been carried out for the alkalis, these calculations give poor agreement with experiment near threshold since they take no account of spin-orbit effects. Moreover, generalizing the nonrelativistic RPA equations to the relativistic case in the most obvious way leads to unphysical couplings between final-state channels having different total angular momenta. The origin of such couplings is the averaging procedure used to obtain a suitable DF ground state.

To circumvent the difficulties in applying RPA to open-shell systems we employ techniques based on many-body perturbation theory.[15] We take our vacuum state to be the $(N-1)$-electron ionic core. The ionic core is described by self-consistent DF orbitals. If we ignore the effects of core excitation, the photoionization amplitude is described by the diagram in Fig. 1(a), where the solid lines represent DF orbitals in the self-consistent field of the core. The first-order core-polarization corrections[15] are due to the particle-hole excitations described in the diagrams of Figs. 1(b)–(e). The core-excitation diagrams shown in Fig. 1(f) are in turn subject to the correlation corrections illustrated in Figs. 1(g)–(l). By iterating the diagrams of Fig. 1 at the photon vertex we arrive at coupled equations, similar to those of the closed-shell RRPA,[13] which are suitable for numerical solutions. The specific equations will be presented elsewhere. Neglecting altogether the core excitations, the present calculation reduces to a relaxed-core DF calculation and leads to results in

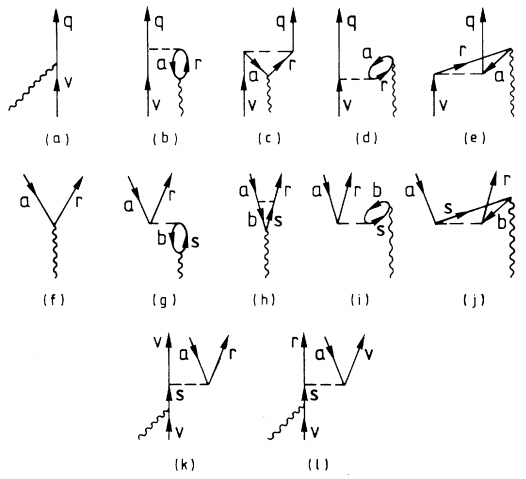

FIG. 1. Feynman diagrams describing cesium excitation including lowest-order correlation corrections. Solid lines: DF orbitals; v = valence orbital, a,b = core orbitals, q = valence excitation, r,s = core excitations. Dashed lines: Coulomb interaction. Wavy lines: Incident photon. (a) Amplitude for exciting valence electron in DF approximation. (b)–(e) Lowest-order correlation corrections to (a). (f) Core excitation amplitude; (g)–(j) core correlation corrections to (f); (k), (l) correlation corrections to (f) due to coupling with valence electron. Diagrams of this figure are iterated at the photon vertex in the present calculation.

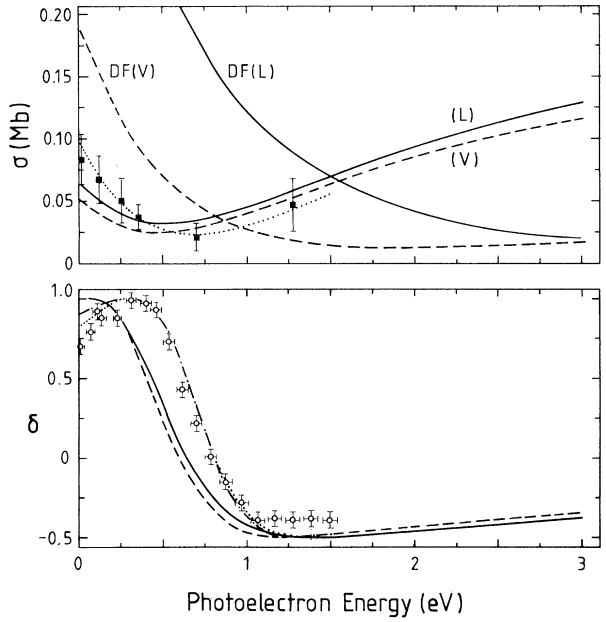

FIG. 2. Upper panel: Photoionization cross section for cesium. Experimental points: Measurements of Cook *et al.* (Ref. 17). DF(L), DF(V): Dirac-Fock calculations with length gauge (solid curve), and with velocity gauge (dashed curve). (L), (V): Present calculations including core excitations with use of length (solid curve) and velocity (dashed curve) gauges. Dotted curve: Model-potential calculation of Norcross (Ref. 11). Lower panel: Spin polarization δ of the total photoionization flux for cesium. Dash-dotted curve: values inferred from measurements of Baum, Lubell, and Raith (Ref. 5). Experimental points: measurements of Heinzmann, Kessler, and Lorenz (Ref. 4). Solid curve: present calculations in length gauge. Dashed curve: present calculations in velocity gauge. Dotted curve: model-potential calculation of Norcross (Ref. 11).

good agreement with previous DF calculations,[6,7] but only in qualitative agreement with experiment. Including only the effects of the dominant core excitation channels, one remarkably improves the agreement with both model-potential calculations[11-12] and with measurements; moreover, one reduces the gauge ambiguity inherent in the DF calculations to within several percent. Many-body calculations including the core-polarization corrections of Figs. 1(b)–(e) have been carried out previously for Na by Chang and Kelly.[16]

The differential cross section for photoionization at low energies may be written

$$\frac{d\sigma}{d\Omega}=\frac{\sigma}{4\pi}\left[1-\frac{\beta}{2}P_2(\cos\theta)\right], \tag{1}$$

where σ is the photoionization cross section and where β is a parameter describing the asymmetry in the photoelectron angular distribution. The angle θ is measured between the directions of the incident photon $\vec{k}$ and of the ionized electron $\vec{p}$. In the upper panel of Fig. 2 we compare the predicted energy dependence of the cross section σ given by the present calculation with results of DF calculations and with the atomic beam measurements of Cook *et al.*[17] The two DF curves labeled DF(L) and DF(V) are the results of calculations ignoring core excitations but carried out in gauges leading nonrelativistically to length (L) and to velocity (V) form transition amplitudes. The velocity-form results are in close agreement with those of Chang and Kelly[6] who also worked in the velocity (Coulomb) gauge. The corresponding length and velocity results including core excitation, labeled L and V, are found to be in substantially better agreement with each other and with the measured cross section than are the DF results. In this connection we mention that when solving our equations numerically we included only the dominant core-excitation channels: $5p_{1/2}\rightarrow d_{3/2}$, $5p_{3/2}\rightarrow d_{3/2}$, and $5p_{3/2}\rightarrow d_{5/2}$; together with the valence channels: $6s_{1/2}\rightarrow p_{1/2}$ and $6s_{1/2}\rightarrow p_{3/2}$.

Calculations including $5p \rightarrow s$ excitations carried out at several energies led to minor changes in the photoionization amplitudes with no improvement in the agreement between L and V results; further tests including excitations of the $4d$ subshell led to no substantial changes in the amplitudes, but did improve slightly the agreement between the two gauges. The spin polarization δ of the total photoionization flux determined by the present calculation is shown with solid (L) and dashed (V) curves in the lower panel of Fig. 2. Experimental values of δ inferred from the measurements of Baum, Lubell, and Raith[5] are shown with a dot-dashed line in the lower panel along with points measured by Heinzmann, Kessler, and Lorenz.[4] Again, the theoretical L and V calculations are in good agreement with each other while the agreement between theory and measurement is significantly improved over previous DF calculations.[6] The measurements shown in Fig. 2 all indicate that the theoretical amplitudes pass through zero at an energy of approximately 0.3 eV lower than the actual physical amplitudes.

The spin polarization[18-20] of photoelectrons produced by circularly polarized light is conveniently described in a coordinate system having Z axis along $\vec{p}$, Y axis along $\vec{k}\times\vec{p}$, and X axis along $[\vec{k}\times\vec{p}]\times\vec{p}$. In this coordinate system the components of the electron spin polarization vector are (in the notation of Huang[18])

$$P_X = \pm\xi \sin\theta/F(\theta), \tag{2a}$$

$$P_Y = \eta \sin\theta\cos\theta/F(\theta), \tag{2b}$$

$$P_Z = \pm\zeta \cos\theta/F(\theta), \tag{2c}$$

$$F(\theta) = 1 - \tfrac{1}{2}\beta P_2(\cos\theta), \tag{2d}$$

where the $\pm$ signs correspond to incident photons of helicity ± 1. The parameters ξ, η, and ζ are dynamical quantities which can be expressed in terms of the photoexcitation amplitudes. The spin polarization of the total photoelectron flux is given by

$$P_{\text{tot}} = \pm\delta = \pm\tfrac{1}{3}(\zeta - 2\xi). \tag{3}$$

In Fig. 3 we give the theoretical values of the dynamical parameters β, ξ, η, and ζ including the effects of core polarization. When we ignore the spin-orbit interaction, β takes on the energy-independent value of 2 while ξ, η, ζ, and δ all vanish; these parameters are therefore sensitive to relativistic effects. The fact that they are sensitive to correlation is illustrated by comparing the present result for β with the corresponding DF

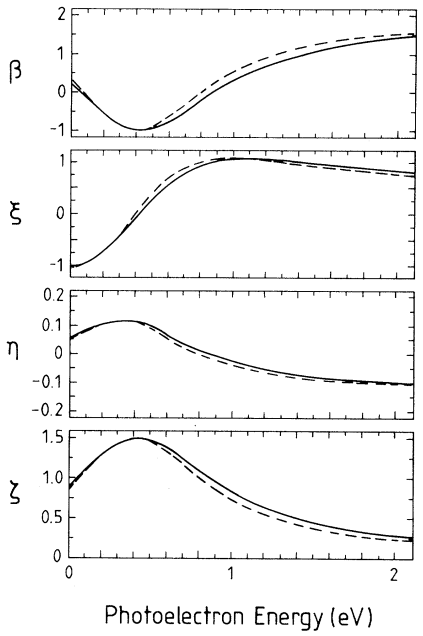

FIG. 3. Dynamical parameters for photoionization of cesium. β is the angular distribution asymmetry parameter, Eq. (1). ξ, η, and ζ are spin polarization parameters, Eq. (2). Solid lines and dashed lines, respectively, represent length and velocity gauge results of the present calculation.

calculation of Ong and Manson.[7] Whereas the present results predict that $\beta = -1$ at a photoelectron energy of approximately 0.4 eV, the DF calculation[8] gives $\beta = -1$ at an energy of approximately 1.5 eV.

The transverse spin polarization P_Y given in Eq. (2b) is of particular interest since P_Y is nonzero for unpolarized incident radiation. The size of P_Y is governed by the dynamical parameter η which is in turn proportional to the sine of the phase difference between the amplitudes for final states with angular momenta $\frac{1}{2}$ and $\frac{3}{2}$. The phase difference predicted in the present calculation is nearly energy independent and has the value 0.105 rad at threshold as compared with the value 0.10 rad obtained from a quantum-defect analysis of the cesium spectrum.

In summary we have developed a method for incorporating the dominant effects of correlations within a relativistic framework for simple open-shell systems. The gauge ambiguities of DF calculations are essentially eliminated with use of this technique and the agreement with the measured low-energy photoionization cross section is substantially improved over DF calculations. Despite the improvement, the present calcula-

tions are not in as close agreement with experiment as the best model-potential calculations, presumably because model-potential parameters include, empirically, effects of two-particle, two-hole final-state interactions and other many-body effects neglected here. Nevertheless, the present calculations do give for the first time a consistent relativistic many-body analysis of photoexcitation of alkali systems, and form the basis for more elaborate future investigations.

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