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Photoelectron Angular Distributions of s Electrons in Open-Shell Atoms

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phenomena suggested to date[1,5] would predict pions at significantly higher energies. In particular, the condensation pions of Kitazoe and coworkers[5] should also emerge with low energies. The pion energy spectra may in fact provide information on the thermal state of the shock wave.

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Photoelectron Angular Distributions of *s* Electrons in Open-Shell Atoms

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It is shown that the photoelectron angular distribution of s electrons in an open-shell atom, having outer configuration ns^2np^q, is not described by the asymmetry parameter $\beta=2$, as predicted by more approximate theories, but has dramatic variations with energy even within LS coupling. Calculations for the 3s subshell in Cl are presented as an example.

One of the most striking predictions of the photoelectron angular distribution theory of Cooper and Zare[1] is that photoionization of an electron having orbital angular momentum $l=0$ leads to a pure $\cos^2\theta$ photoelectron angular distribution, irrespective of the photon's energy. Here θ is measured from the axis of linear polarization of the incident light and the differential cross section is

$$d\sigma/d\Omega=(\sigma/4\pi)[1+\beta P_2(\cos\theta)].$$

A pure $\cos^2\theta$ distribution corresponds to an asymmetry parameter $\beta=2$. Classically, this result is intuitively obvious: Since the initial state is spherically symmetric, the photoelectron angular distribution is centered about the electric vector of the incident light. Quantum mechanically, with neglect of retardation and relativistic effects, this result holds exactly for photoionization of atomic hydrogen.[2] For more complicated atoms, this result follows in the Cooper-Zare theory[1] from the approximation that in the final state

there is only a *single* outgoing p wave. In general β is a function of photon energy and varies between -1 and $+2$ as a result of interferences between *multiple* final-state channels.

The experimental evidence on the angular distribution of photoelectrons having initial angular momentum $l=0$ is sparse. This is due in part to the relatively small photoionization cross sections of $l=0$ electrons in many atoms and in part to the assumption that the result is already known to be $\beta=2$. Thus, for example, the value $\beta=2$ for the photoelectrons resulting from photoionization of He has been used by experimentalists for calibration purposes, although no published experimental measurement is known to us. Houlgate *et al.*[3] measured the photoelectron angular distribution resulting from photoionization of the $3s$ subshell in Ar, but still found $\beta=2$. Only for *much* heavier atoms have deviations from the value $\beta=2$ been found experimentally. Niehaus and Ruf[4] studied the $6s$ subshell in Hg and Dehmer[5] and Dill studied the $5s$ subshell in Xe; in both cases β was measured to be considerably different from 2. Dehmer and Dill[5,6] interpreted these measurements as an effect of relativistic interactions which mix the allowed $^1P_1$ channel with the spin-orbit–populated $^3P_1$ channel.

In this Letter we present theoretical formulas which indicate explicitly that for photoionization of $l=0$ electrons *in open-shell atoms*, β may be *expected* to differ from 2. This result is obtained in LS coupling; spin-orbit and other relativistic interactions are not introduced. In addition, calculations are presented for the $3s$ subshell in atomic Cl, a relatively light atom, which demonstrate oscillations in the parameter β as a function of photon energy due to very large effects of anisotropic electron-ion interactions.[7,8]

Consider the photoionization of the ns subshell of an atom A having an open np subshell, i.e., the process

$$A\,ns^2np^q(L_0S_0)+\hbar\omega\rightarrow A^+nsnp^q(L_0S_c)\epsilon p(LS_0). \qquad (1)$$

To calculate the parameter β for Reaction (1) we shall adopt the angular-momentum–transfer formulation of Dill and Fano.[6,9,10] While other formulations for β in principle give equivalent results, they do not demonstrate the consequences of final-state anisotropic interactions as clearly. If the angular momentum of the photoelectron is $\vec{l}$, then the allowed values of the angular momentum transfer j_t are given (in LS coupling) by the relations

$$\vec{j}_t=\vec{j}_\gamma-\vec{l} \text{ and } \vec{j}_t=\vec{L}_0-\vec{L}_c, \qquad (2)$$

where the angular momentum of the photon has the magnitude $j_\gamma=1$. For Reaction (1), $l=1$, and hence j_t is restricted by the first relation in (2) to the values 0, 1, and 2. As shown in Refs. 6 and 8, and as shown below, in the approximation that a single radial wave function is used to represent the continuum p electron in (1), as in the Cooper-Zare theory for β, then only the value $j_t=0$ contributes to (1) and β is consequently restricted to the value 2. Hence the contributions of angular momentum transfers $j_t\neq 0$, due to anisotropic final-state interactions, are what cause β to deviate from 2. In Cl, as shown below, these contributions approach 100% of the contribution from all angular momentum transfers.

Dill, Manson, and Starace[7,8] have given the LS-coupling form of the general theory of Dill and Fano[6,9,10] appropriate for photoionization of the outer subshell of an atom having the configuration (closed shells) nl^N. Using the same methods as in Ref. 8 we obtain the following LS-coupling expressions for the parameter β appropriate to (1):

$$\beta=\frac{2|S_1(0)|^2-3|S_1(1)|^2+|S_1(2)|^2}{|S_1(0)|^2+3|S_1(1)|^2+5|S_1(2)|^2}, \qquad (3)$$

where

$$S_1(j_t)=C\sum_L(-1)^L(2L+1)\exp(i\delta_{\epsilon p}{}^{L_0S_cL})R_{\epsilon p}{}^{L_0S_cL}\begin{Bmatrix}L_0 & L_0 & j_t\\ 1 & 1 & L\end{Bmatrix}. \qquad (4)$$

In Eq. (4) the subscript "1" on the expression $S_1(j_t)$ indicates the orbital angular momentum of the continuum p electron; the constant C represents factors common to a particular ion-core term level which cancel in the ratio (3); $R_{\epsilon p}{}^{L_0S_cL}$ is the radial dipole matrix element (in the length form),

$$R_{\epsilon p}{}^{L_0S_cL}\equiv\int_0^\infty dr\,P([L_0S_c]\epsilon p, LS_0|r)rP(ns, L_0S_0|r), \qquad (5)$$

with the radial wave functions for both the initial-state ns orbital and the final-state ϵp orbital as-

sumed to depend on the term levels involved; and, finally $\delta_{\epsilon p}{}^{L_0 S_c L}$ is the phase shift of the final-state ϵp orbital with respect to a pure Coulomb p wave. We observe that in the limit that the dipole matrix elements and phase shifts become independent of the quantum numbers L_0, S_c, and L (in which case we say that the electron-core interaction is isotropic), then the sum over L in Eq. (4) may be performed analytically[11] to yield

$$S_1(j_t) = (-1)^{L_0+1} C \exp(i\delta_{\epsilon p}) R_{\epsilon p} [3(2L_0+1)]^{1/2} \delta(j_t, 0). \tag{6}$$

Hence, with substitution of Eq. (6) into Eq. (3) it is seen that in this approximation β equals 2 identically since only $j_t = 0$ contributes.

Lastly, as a measure of the contribution of angular momentum transfers $j_t \neq 0$ we define the cross section ratio

$$\frac{\sigma - \sigma(j_t=0)}{\sigma} = \frac{3|S_1(1)|^2 + 5|S_1(2)|^2}{|S_1(1)|^2 + 3|S_1(1)|^2 + 5|S_1(2)|^2}, \tag{7}$$

where σ is the total photoionization cross section for Reaction (1) and $\sigma(j_t=0)$ is the partial cross section corresponding to the angular momentum transfer $j_t = 0$.

For the case of atomic chlorine, Reaction (1) becomes

$$\mathrm{Cl}\, 3s^2 3p^5(^2P) + \hbar\omega \rightarrow \mathrm{Cl}^+\, 3s\, 3p^5(^{1,3}P)\epsilon p(^2D, {}^2P, {}^2S). \tag{8}$$

We present calculations of the asymmetry parameters $\beta(^1P)$ and $\beta(^3P)$ corresponding to the two possible ionic term levels. For each of these ionic terms, j_t may assume the three values 0, 1, and 2. The matrix elements corresponding to these three j_t values are

$$S_1(0) = \tfrac{1}{3} C\{\exp(i\delta_{\epsilon p}{}^{1S_c 0}) R_{\epsilon p}{}^{1S_c 0} + 3\exp(i\delta_{\epsilon p}{}^{1S_c 1}) R_{\epsilon p}{}^{1S_c 1} + 5\exp(i\delta_{\epsilon p}{}^{1S_c 2}) R_{\epsilon p}{}^{1S_c 2}\}, \tag{9a}$$

$$S_1(1) = \tfrac{1}{6} C\{2\exp(i\delta_{\epsilon p}{}^{1S_c 0}) R_{\epsilon p}{}^{1S_c 0} + 3\exp(i\delta_{\epsilon p}{}^{1S_c 1}) R_{\epsilon p}{}^{1S_c 1} - 5\exp(i\delta_{\epsilon p}{}^{1S_c 2}) R_{\epsilon p}{}^{1S_c 2}\}, \tag{9b}$$

$$S_1(2) = \tfrac{1}{6} C\{2\exp(i\delta_{\epsilon p}{}^{1S_c 0}) R_{\epsilon p}{}^{1S_c 0} - 3\exp(i\delta^{1S_c 1}) R_{\epsilon p}{}^{1S_c 1} + \exp(i\delta_{\epsilon p}{}^{1S_c 2}) R_{\epsilon p}{}^{1S_c 2}\}. \tag{9c}$$

Note that if the radial dipole matrix elements and phase shifts in Eq. (9) become independent of the term levels then $S_1(0) \rightarrow 3C$, $S_1(1) \rightarrow 0$, and $S_1(2) \rightarrow 0$; and hence, from Eq. (3), $\beta \rightarrow 2$. Deviation of β from 2 is thus both a consequence and a measure of *anisotropic* electron-ion interactions. The calculations were performed using restricted Hartree-Fock continuum orbitals calculated in the field of a relaxed ion core. The bound-state Hartree-Fock orbitals were obtained from standard sources[12,13]; the continuum orbitals were calculated using the numerical procedures of Kennedy and Manson.[14]

Figures 1(a) and 1(b) show our calculated values for $\beta(^1P)$ and $\beta(^3P)$, respectively, versus photoelectron kinetic energy. The deviation of β in each case from the value 2 is very large near threshold and tends toward 2 very slowly with increasing energy. Hence the value $\beta = 2$ cannot be assumed for normalization purposes in experimental measurements without introducing a possible additional source of error. In Fig. 2 we have plotted the ratio in Eq. (7) versus photoelectron kinetic energy for the ^3P ion term. Just above threshold this ratio goes to unity, indicating that the $j_t = 0$ component—the only one considered in the simple, approximate calculations

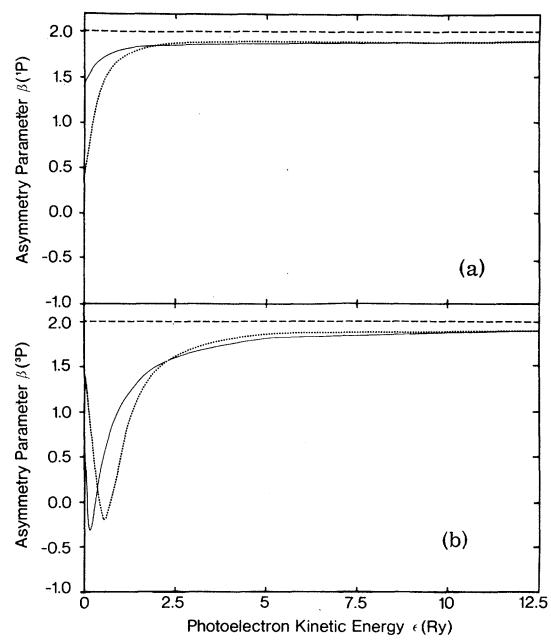

FIG. 1. Asymmetry parameters $\beta(^1P)$ and $\beta(^3P)$ vs photoelectron kinetic energy. The solid (dashed) curves employ the length (velocity) form for the radial dipole matrix elements. The dotted curves indicate the value $\beta = 2$ that would apply in the approximation of isotropic electron-ion interactions, as in a calculation based upon the theory of Ref. 1.

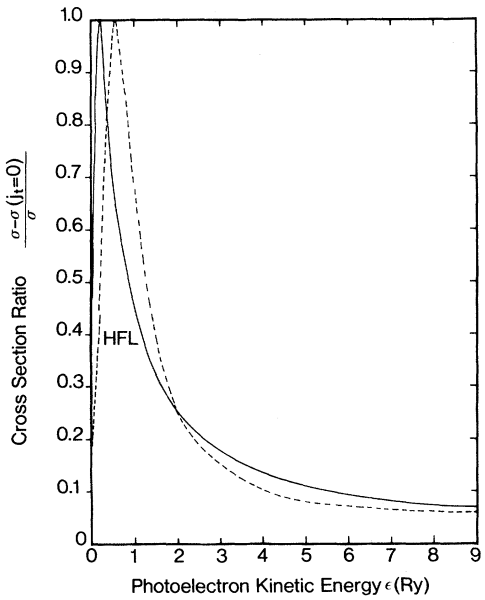

FIG. 2. The cross section ratio $[\sigma - \sigma(j_t = 0)]/\sigma$ plotted vs photoelectron kinetic energy for ionization to the ^3P term of the ion. The solid (dashed) curves employ the length (velocity) form of the radial dipole matrix element.

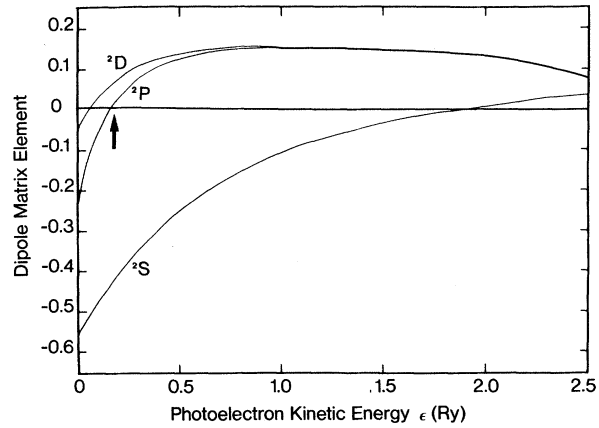

FIG. 3. The radial dipole matrix elements $R_{\epsilon p}^{L_0 S_c L}$ in the length form plotted vs photoelectron kinetic energy for $L_0 = 1$, $S_c = 1$ (i.e., the ^3P level of the ion), and $L = 2$, 1, and 0 (i.e., the $^2D, ^2P$, and ^2S final-state terms). The arrow indicates the kinetic energy at which $S_1(j_t = 0) = 0$, in which case only the angular momentum transfers $j_t \neq 0$ contribute to the cross section and the asymmetry parameter.

for β—*gives no contribution at all.* Lastly, Fig. 3 presents the energy dependence of the three radial dipole matrix elements that contribute to the ionization cross section for producing the ^3P ion term. It is clear from this figure that the minimum in $\beta(^3P)$ in Fig. 1(b) occurs near the Cooper zeros of the ^2D and ^2P radial dipole matrix elements. The arrow in Fig. 3 indicates the energy at which $S_1(j_t = 0) = 0$. From Eq. (9a) we see that at this energy the positive ^2D and ^2P radial matrix elements just cancel the negative ^2S radial matrix element.

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