Oscillations in the Energy Dependence of the Angular Distribution of Photoelectrons

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(Received 19 November 1970)

It is found that the angular distribution of photoelectrons from the argon $4d$ subshell oscillates as a function of energy for the first 150 eV above threshold.

The angular distributions of atomic photoelectrons are currently being studied by refined experimental techniques[1] by a number of groups.[2-11] Apart from its importance as a tool for obtaining information on atomic properties, photoelectron spectroscopy also provides information on the phase shifts of the continuum states as well as the matrix elements relevent to the photoionization process. In this Letter we describe one aspect of the angular distribution of atomic photoelectrons, previously unreported, which looks to be worthwhile for future studies.

The differential photoionization cross section for unpolarized incoming light is given, in the dipole approximation (which is excellent for low-energy photons), by[12,13]

$$d\sigma(\epsilon)/d\Omega = [\sigma_T(\epsilon)/4\pi][1-\tfrac{1}{2}\beta(\epsilon)P_2(\cos\theta)], \tag{1}$$

where ϵ is the photoelectron energy, $\sigma_T(\epsilon)$ is the total photoionization cross section, and $\beta(\epsilon)$ is the asymmetry parameter; the angle between the photon and photoelectron directions is θ, and

$$P_2(\cos\theta) = \tfrac{1}{2}(3\cos^2\theta - 1).$$

Using single-particle wave functions and LS coupling, it is found that the asymmetry parameter is[12,13]

$$\beta(\epsilon) = \frac{l(l-1)R_{l-1}(\epsilon)^2 + (l+1)(l+2)R_{l+1}(\epsilon)^2 - 6l(l+1)R_{l-1}(\epsilon)R_{l+1}(\epsilon)\cos[\delta_{l+1}(\epsilon)-\delta_{l-1}(\epsilon)]}{(2l+1)[lR_{l-1}(\epsilon)^2 + (l+1)R_{l+1}(\epsilon)^2]}, \tag{2}$$

with the dipole matrix element

$$R_{l\pm1}(\epsilon) \equiv \int_0^\infty P_{nl}(r) r P_{\epsilon,l\pm1}(r)\,dr, \tag{3}$$

where $r^{-1}P_{nl}(r)$ and $r^{-1}P_{\epsilon,l\pm1}(r)$ are the radial parts of the wave functions of the initial and final states of the photoelectron. Here the $\delta_{l\pm1}(\epsilon)$ are the phase shifts of the $l\pm1$ continuum functions with respect to free waves. The variation of $\beta(\epsilon)$ (and, thus, the angular distribution) with ϵ is seen from (2) to depend upon the dipole matrix elements and the continuum phase shifts; when one or more of these quantities exhibits a rapid variation with energy it is likely that $\beta(\epsilon)$ will as well.

To illustrate, we have performed a calculation on the $4d$ shell of xenon using a central field approximation.[14] Recent experience has shown that this type of calculation predicts angular distributions very close to those of the much more sophisticated Hartree-Fock self-consistent-field approximation.[15] We have chosen this case for two reasons: First, Xe is a monatomic gas so it is amenable to experimental investigation; second, the dipole matrix element and phase shift of the $4d \rightarrow \epsilon f$ channel vary rather rapidly with energy, the matrix element even changing sign and going through a zero at ~75 eV above threshold. The result of our calculation is shown in Fig. 1 where the oscillatory behavior of β is clearly exhibited. This oscillation follows closely the oscillations of $\cos[\delta_{l+1}(\epsilon)-\delta_{l-1}(\epsilon)]$ whose argument changes by $\sim 5\pi/2$ from threshold to 4 Ry. For the first rydberg above threshold the variation is due primarily to the Coulomb-wave

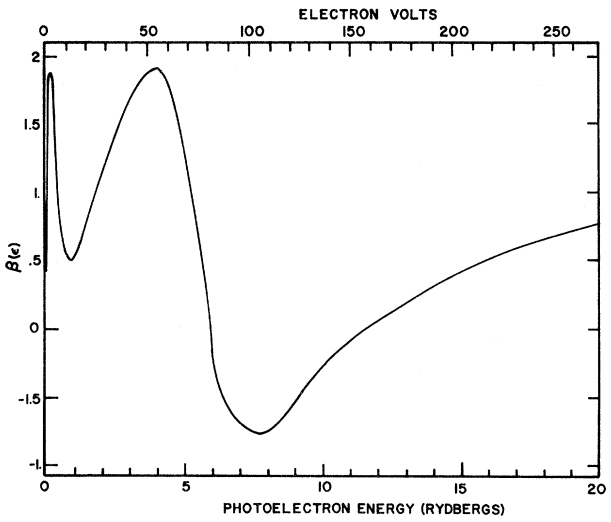

FIG. 1. Asymmetry parameter $\beta(\epsilon)$ as a function of photoelectron energy ϵ for the Xe $4d$ subshell.

phase shift with respect to free waves; from 1 to 4 Ry it is due mostly to the non-Coulomb shape resonance effect in the f-wave channel.[16] Above 4 Ry, the only major variation of β with energy occurs principally because of the Cooper minimum at $\epsilon = 5.6$ Ry and the associated change in sign of R_{l+1}. At still higher energies β is a smooth function of ϵ.

At this point it is worthwhile to point out that this calculation does not treat exchange exactly and omits correlation and spin-orbit effects which may be important.[17] Indications are that exchange effects will not significantly affect the angular distribution.[15] As for the other effects, while they may change the positions of the minima and maxima in β somewhat, it seems unlikely that they could greatly affect the overall oscillatory behavior.

In conclusion, we hope that experimental studies, as well as further theoretical work, will provide more information on this phenomenon.

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Time Evolution of Simple Quantum-Mechanical Systems. II. Two-State System in Intense Fields*

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The Schrödinger equation for a two-state quantum-mechanical system with sinusoidal perturbation is numerically integrated with respect to time. From these results a general formula for the induced transition probability (as a function of time, perturbation frequency, and perturbation strength) is extracted.

There has been a recent surge of interest in the properties of quantum systems in intense, coherent radiation fields.[1-3] We present here those preliminary results of a more general investigation which are relevant to this problem.

Consider a two-state system with states $|0\rangle$ and $|1\rangle$; we introduce an external time-dependent driving field in the Hamiltonian:

$$H = H_0 + V\sin(\omega t + \delta),$$

where

$$H_0|0\rangle = 0, \quad H_0|1\rangle = \hbar\omega_0|1\rangle,$$

$$\langle 0|V|0\rangle = \langle 1|V|1\rangle = 0,$$

and

$$\langle 0|V|1\rangle = V_{01} = \langle 1|V|0\rangle.$$

The time-evolution operator in the interaction representation satisfies the Schrödinger equation[4]:

$$i\hbar(\partial/\partial t)U_I(t) = V_I(t)U_I(t). \tag{1}$$

Since all the matrices in (1) are two-by-two, it is a trivial matter to integrate the equation numerically with initial condition $U_I(0) = 1$. In Fig. 1 we show the induced transition probability, $|\langle 1|U_I(t)|0\rangle|^2$, as a function of time for several values of ω/ω_0, with $\delta = 0$ and the perturbation