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Photoelectron Polarization in Hg $6s^2$ Subshell Ionization with Unpolarized Light: New Aspect of the Fano Effect

G. Schönhense and U. Heinzmann
Fritz-Haber-Institut der Max-Planck-Gesellschaft, D-1000 Berlin 33, West Germany, and Physikalisches Institut der Universität Münster, D-4400 Münster, West Germany

and

J. Kessler
Physikalisches Institut der Universität Münster, D-4400 Münster, West Germany

and

N. A. Cherepkov
A. F. Ioffe Physical-Technical Institute, 194021 Leningrad, U. S. S. R.

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Experimental evidence is presented for the effect of photoionization of ns^2 atomic subshells with *unpolarized* radiation that leads to *highly polarized* photoelectrons in angular-resolved measurements. Like the Fano effect, the observed phenomenon is caused by spin-orbit interaction in the continuous spectrum. The spin polarization essentially depends on the phase-shift difference between the $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ continua.

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In a well-known paper Fano[1] predicted high spin polarization of photoelectrons which are ejected by circularly polarized light from alkali atoms near the Cooper minimum of the cross section. One year later the Fano effect was established experimentally.[2] The polarization of photoelectrons from *ns* subshells is direct evidence of spin-orbit interaction in the continuous spectrum, which leads to a difference between wave functions corresponding to the $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ outgoing partial waves. For the same reason the angular asymmetry parameter β in the Cooper minimum varies from $+2$ to -1 as a function of photon energy while in the *LS*-coupling scheme it should be equal to $+2$ irrespective of photon energy.[3]

The Fano effect also appears in ns^2 subshells,[4-6] since its cause, the spin-orbit interaction in the continuous spectrum, evidently exists there, and there is usually a Cooper minimum, also. But while in alkalis the minimum appears at energies between the first and the second ionization threshold, in ns^2 subshells it can occur at energies above the second threshold. Therefore interchannel interaction[7] has to be taken into account, resulting in the fact that the dipole matrix elements which describe the Fano effect quantitatively are no longer real. As a consequence, one obtains a high polarization of photoelectrons ejected at a definite angle by unpolarized light. This Letter reports on the first experimental verification of this new aspect of the Fano effect. The polarization of photoelectrons that have been ejected from mercury ($6s^2$) atoms by unpolarized vacuum ultraviolet radiation at the magic angle of 54°44′ has been measured with the apparatus of Schönhense *et al.*[8]

Theoretically the photoionization of ns^2 subshells is described by the moduli of two complex dipole matrix elements $|d_1|$ and $|d_3|$ of the $ns \to \epsilon p_{1/2}$ and $ns \to \epsilon p_{3/2}$ transitions, respectively, and the corresponding phase-shift difference $\Delta = \delta_3 - \delta_1$, all of these quantities being functions of photon energy $\hbar\omega$. In photoionization with unpolarized light the following parameters are observable: (i) The photoionization cross section of the subshell,

$$Q = \tfrac{2}{3}(4\pi^2\alpha\omega/3)(|d_1|^2 + 2|d_3|^2), \tag{1}$$

where $\alpha = \frac{1}{137}$ and atomic units $\hbar = m = e = 1$ are used; (ii) the angular asymmetry parameter β of the differential cross section,

$$\beta = \frac{2|d_3|^2 + 4\,\mathrm{Re}(d_3{}^* d_1)}{|d_1|^2 + 2|d_3|^2}; \tag{2}$$

and (iii) the degree of transverse polarization $p_\perp$ of photoelectrons ejected by unpolarized light at some angle θ,[5-7]

$$p_\perp = \frac{2\xi\sin\theta\cos\theta}{1 - \frac{1}{2}\beta(\frac{3}{2}\cos^2\theta - \frac{1}{2})}, \tag{3}$$

where

$$\xi = -\frac{3}{2}\,\frac{\mathrm{Im}(d_3{}^* d_1)}{|d_1|^2 + 2|d_3|^2}. \tag{4}$$

The spin parameter ξ is especially sensitive to the phases of the complex matrix elements. Introducing the ratio $\gamma = |d_1|/|d_3|$, we obtain from (2) and (4)

$$\beta = \frac{2 + 4\gamma\cos\Delta}{2 + \gamma^2}, \quad \xi = \frac{3\gamma\sin\Delta}{2(2 + \gamma^2)}. \tag{5}$$

For comparison, let us first discuss the situation in alkali atoms, at which the investigations were focused in the first years after the discovery of the Fano effect.[1] The behavior of the parameters γ and Δ near the Cooper minimum of the ns valence shells of these atoms is quite simple. The minimum appears at energies where only the valence shell can be ionized and no autoionization resonances appear. Hence photoionization is described in good approximation by two real matrix elements, the phase-shift difference between the two continua being very small[1] ($\sin\Delta \ll 1$). In the range of the Cooper minimum the dipole matrix elements change their signs.[9] Since the spin-orbit interactions for the $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ states have opposite signs, the matrix elements d_1 and d_3 are different, and d_1 always changes its sign at a lower energy than does d_3.[3] Thus γ is less than 1 for photon energies below the Cooper minimum and larger than 1 above it. The dashed curves in Fig. 1 illustrate this behavior of γ and Δ. The jumps of $\pm\pi$ in Δ correspond to the change of sign of d_1 and d_3. Since both signs of jump are equivalent, both are shown in the figure. As a consequence of the phase-shift difference being close to 0 (or to $\pm\pi$) the degree of polarization of photoelectrons ejected by unpolarized light from alkali atoms does not exceed several percent.

In cases where more than one channel is open, interchannel correlations cause the matrix elements to be complex. The full curves in Fig. 1 correspond to this case. As a guide we have used the results of Johnson and Cheng[7] for the

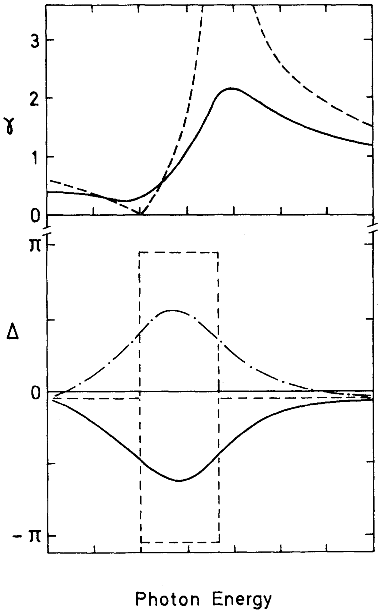

FIG. 1. An illustration of a typical behavior of the parameters γ and Δ as a function of the photon energy for real (dashed curves) and complex (full and dash-dotted curves) dipole matrix elements.

$5s^2$ subshell of Xe calculated in relativistic random-phase approximation (RRPA), according to which the moduli of the matrix elements are everywhere different from zero. There are no jumps in the phase-shift difference; instead Δ varies smoothly throughout the region of the Cooper minimum. In analogy to the two signs of jump there are two possibilities for Δ: It can be either negative or positive in the Cooper minimum (full and dash-dotted curves, respectively). As a matter of fact, Δ is negative in the $5s^2$ subshell of Xe (Ref. 7) and positive in the $6s^2$ subshell of Hg (see below). Only a measurement of the degree of polarization of photoelectrons ejected by unpolarized light enables one to find the sign of Δ from the experiment, since it is only the parameter ξ which is proportional to $\sin\Delta$, while the other parameters depend on $\cos\Delta$.[5,6]

The measured parameters ξ and β for Hg $6s^2$ are shown in Fig. 2 together with a β value from Niehaus and Ruf,[10] experimental cross-section data,[11] and several theoretical calculations. The three vertical dashed lines denote the ionization thresholds of the $6s$, $5d_{5/2}$, and $5d_{3/2}$ subshells, respectively. The region between the thresholds is strongly influenced by autoionization resonances.[12] The spin parameter ξ has been measured at three intense rare-gas resonance lines (Ne I 16.85 eV, He I 21.22 eV, and Ne II 26.9 eV). At 16.85 eV the measurement yielded the highest ξ value which has been experimentally found as yet. It corresponds to a maximum photoelectron polarization of more than 50% for emission angles around 30° with respect to the unpolarized photon beam.

The two measured values of the parameter β deviate significantly from the nonrelativistic value of 2. The β value of Niehaus and Ruf[10] at 21.22 eV shows good agreement with the present measurement. The subshell cross section Q has been measured by Shannon and Codling[11] using monochromatic synchrotron radiation. Their results indicate a cross-section (Cooper) minimum approximately 3 eV above the third threshold.

Comparison between the experimental and theoretical results shows the following:

(1) The RRPA,[13] which takes into account interchannel interactions with the $5d^{10}$ subshell, reflects in principle the correct behavior. But since it yields a Cooper minimum below the $5d$ thresholds, significant quantitative deviations from the measurements appear. The effect in the RRPA that the position of the minimum depends critically upon the correlations included is also known for other ns^2 systems.[7]

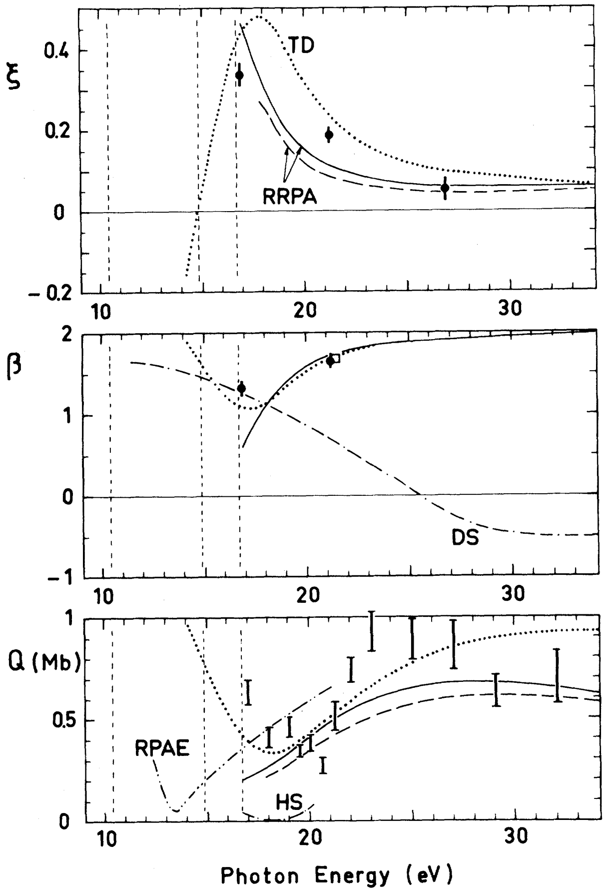

FIG. 2. Spin parameter ξ, asymmetry parameter β, and cross section Q for the $6s^2$ subshell of mercury. Experiment: points with error bars (one standard deviation), this work; square, Ref. 10; error bars for Q, Ref. 11. Theory: (solid curve) RRPA (experimental thresholds) (Ref. 13) for ξ, β, and Q; (dashed curve) RRPA (thresholds from theory) (Ref. 13) for ξ and Q; (dotted curve) Tamm-Dancoff (TD) calculation (Ref. 13) for ξ, β, and Q; (double-dash–dotted curve) Dirac-Slater (DS) calculation (Ref. 16) for β; (dash–double-dotted curve) RPAE (Ref. 14) for Q, with $\xi = 0$ and $\beta = 2$; (dash-dotted curve) Hartree-Slater (HS) calculation (Ref. 15) for Q with $\xi = 0$ and $\beta = 2$.

(2) The Tamm-Dancoff calculation[13] is based on the RRPA code of (1) but does not take into account ground-state correlations. It has been plotted only to illustrate the energy dependence of the parameters in the case that the Cooper minimum lies above the third threshold. The fact that all experimental results are rather close to the TD curves indicates indeed a position of the minimum above the $5d$ thresholds. This agreement may not lead to the conclusion

that the TD calculation (ground-state correlations omitted) yields in general better results than the full RRPA.

(3) Like the RRPA, the nonrelativistic random-phase approximation with exchange[14] (RPAE), which takes into account interchannel correlations with the $5d \rightarrow \epsilon f$ channel, shows the minimum of Q already in the autoionization region. The nonrelativistic values for β and ξ are 2 and 0, respectively. The discrepancies between this theory and experiment are therefore quite large.

(4) The single-particle calculations using the nonrelativistic Hartree-Slater[15] or the relativistic Dirac-Slater[16] central-potential models also do not agree with the experiments.

The results show that further investigations are desirable, both theoretical and experimental. In theory it is necessary to go beyond the RRPA and take into account two-particle–two-hole excitations which may give here an important contribution. In experiment it is necessary to measure ξ and β between the three present points using a tunable light source. The intense synchrotron radiation from storage rings should make such energy-, angle-, and spin-resolved measurements feasible.

The present paper verifies that photoelectrons from $l = 0$ subshells can be polarized not only when ejected by circularly polarized light, but also when ejected by unpolarized light. Since the same phenomena in subshells with $l > 0$ have been established previously,[8,17] it is possible to say now that photoelectrons ejected into a definite angle from any atomic subshell by light of any polarization (unpolarized included) are usually polarized. Investigation of transverse polarization of photoelectrons gives the most direct information on the phase-shift difference of interfering partial waves.

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