Angular Dependence of the Polarization of Photoelectrons Ejected by Plane-Polarized Radiation from Argon and Xenon Atoms

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The photoelectrons produced by ionization of argon and xenon atoms with linearly polarized vacuum-ultraviolet radiation of the helium resonance wavelength 58.43 nm have been analyzed simultaneously as to their energy, angle of emission, and spin. Degrees of spin polarization up to 0.5 have been found. The results verify the theoretically predicted angular dependence of the photoelectron polarization. The asymmetry parameter β describing the angular distribution of the cross section is determined via electron intensity and polarization measurements.

Recent theoretical investigations[1-4] have shown that photoelectrons from unpolarized target atoms can be spin polarized not only if the ionizing radiation is circularly polarized (Fano effect[5]) but even if unpolarized or linearly polarized radiation is used. The predictions for unpolarized light have been experimentally verified for atomic lead[6] and rare gases[7,8] by analyzing photoelectrons ejected at two fixed angles with respect to the photon beam. This Letter reports the first experiment performed to measure the angular distribution of photoelectron polarization for plane-polarized vacuum-ultraviolet (vuv) light over the full angular range.

The interest in these phenomena results from the intention to find new experimental methods which complement measurements of the cross section and its angular distribution in order to obtain detailed and possibly full information about the photoionization process. Symmetry arguments require that, contrary to Fano-effect measurements,[5,9] the average electron polarization would vanish if all the photoelectrons produced were extracted by an electric field independent of their direction of emission. It is the purpose of this Letter to show that the predicted polarization formula is valid for the systems investigated. The results for Ar and Xe at 58.43 nm are compared with both theoretical predictions of Cherepkov[2] and Huang, Johnson, and Cheng[4] and experimental values measured at fixed angles with unpolarized radiation of the same wavelength by Heinzmann, Schönhense, and Kessler.[8]

According to theory[1-4] the angular dependence of the photoelectron polarization in the case of totally plane-polarized light was expected to be (in dipole approximation)[10]

$$\vec{P}(\theta) = \frac{4\xi \sin\theta \cos\theta}{1+\beta(\frac{3}{2}\cos^2\theta - \frac{1}{2})}\hat{s}, \qquad (1)$$

where $\hat{s} = \hat{k}\times\hat{e}/\sin\theta$. The unit vectors $\hat{k}$ and $\hat{e}$ have the direction of electron momentum and electric vector of the incident radiation, respectively, and θ is the angle between $\hat{k}$ and $\hat{e}$ defined by the vector product. As in any polarization formula the denominator of Eq. (1) is proportional to the differential cross section with the well-known asymmetry parameter[11] β. Like β, the parameter ξ depends on photon energy and the initial and final states of the electronic transitions.[10] In the general case of partially plane-polarized radiation one can consider the incoming light wave to be an incoherent superposition of two totally polarized waves with orthogonal electric vectors defining the x and y axes of a coordinate system. The resulting angular distribution of electron polarization is thus obtained as the vector sum of two terms having the form of Eq. (1)

$$\vec{P}(\theta_x,\theta_y) = \frac{I_x}{I_x+I_y}\vec{P}(\theta_x) + \frac{I_y}{I_x+I_y}\vec{P}(\theta_y), \qquad (2)$$

where I_x and I_y are the light-intensity components in the direction of the x and y axis, respectively.

The technical problems of a revolving electron detector (Mott analyzer at 120 kV) can be avoided by using a rotatable light polarizer to vary θ. When the latter arrangement is used, the detector (defining the direction of observation $\hat{k}$) can be mounted at a fixed angle in such a way that the unit vector $\hat{s}$ remains constant during the measurement. If the photoelectrons are observed in the x-y plane defined by the electric vectors, $\hat{k}\times\hat{e}$ is always perpendicular to the plane and the electrons detected are transversely polarized in z direction. By use of the relation $\theta = \theta_x = \theta_y + 90°$ as well as the definition for the degree of plane polarization of the incoming radiation $P_r = (I_x - I_y)/(I_x + I_y)$, the expression for the spin polarization

can be derived from the above equations:

$$P_z(\theta) = P_\tau \frac{4\xi \sin\theta \cos\theta}{1 + (1 - P_\tau)\frac{1}{4}\beta + P_\tau \beta(\frac{3}{2}\cos^2\theta - \frac{1}{2})}, \quad (3)$$

where the denominator is again proportional to the differential cross section for partially plane-polarized radiation.[12]

The experimental arrangement,[13] which allows observation of the complete angular distribution $P_z(\theta)$, is shown in Fig. 1. The vuv radiation is produced in a rare-gas resonance lamp, an improved version of the type described by Heinzmann and Schönhense,[14] with a maximum light intensity of about 10^{13} photons/s at the helium resonance line, 58.43 nm (21.22 eV). The radiation is plane polarized by reflecting the incident beam on three gold coatings. Angles of incidence are 76°, 62°, 76°, as this combination provides a good compromise between high reflectivity and a sufficient degree of polarization.[12] A transmission of 8% (for linearly polarized light) was achieved. The gold mirrors are aligned such that the light beam is neither deviated not shifted under rotation of the three-mirror device. The degree of plane polarization is determined by the method presented by Rabinovitch, Canfield, and Madden,[15] based upon reflectance measurements on a plane mirror at 45° with respect to the light beam, a technique which is applicable for a gold surface at 58.43 nm.[16] Because of contamination of the gold films the degree of polarization decreased from the maximum value of 88% (± 4%) to 68% after 30 h of continuous lamp operation. The atomic beam is produced by a focusing nozzle consisting of two coaxial metal cones separated by an adjustable gap of some tenths of a millimeter serving as gas outlet. The light beam passes through a central bore (see Fig. 1). A cryopump reduces the target-gas background during the measurement to less than 10^{-3} Pa. Electric fields in the ionization region near to the target-gas focus are minimized by means of a graphitized copper cylinder coaxial to the light beam (not shown in Fig. 1). Earth's magnetic field is compensated by three pairs of Helmholtz coils to less than 1% to make sure that only photoelectrons which are ejected into a certain solid-angle interval can enter the spectrometer. The cylindrical mirror analyzer (energy resolution 0.7% full width at half maximum) serves to select electrons corresponding to one of the two final ionic states $^2P_{1/2}$ or $^2P_{3/2}$, which differ in energy by 0.18 and 1.30 eV for argon and xenon, respectively.

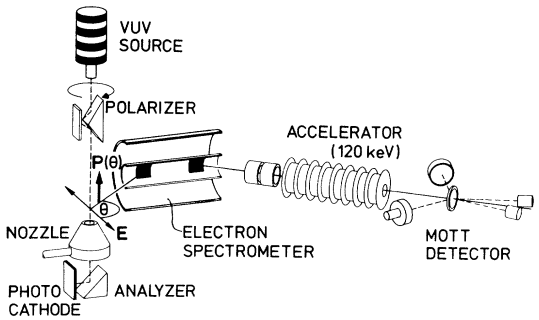

FIG. 1. Schematic view of the experimental setup. Spectrometer slits, and apertures in the light and electron path, as well as shielding elements, are omitted.

The photoelectrons with the energy selected are injected into an acceleration tube for 120 keV and hit the gold foil of a Mott detector. The spin polarization is determined from the left-right asymmetry[9] of the electron intensity scattered into the two detectors at 120°. Two additional detectors are mounted in the forward-scattering direction in order to control the local

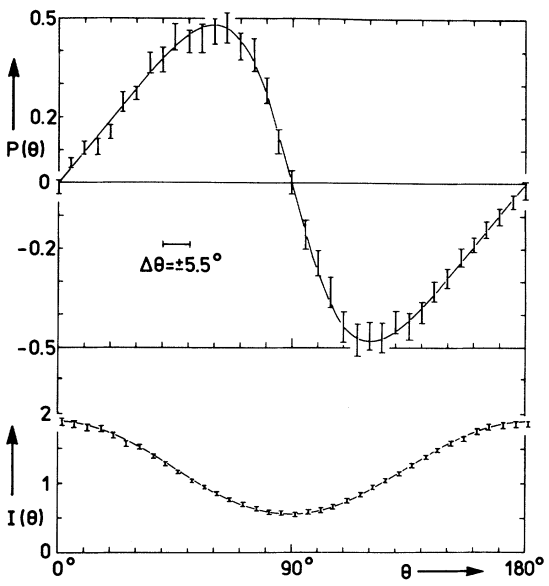

FIG. 2. Angular distribution of photoelectron polarization $P(\theta)$ (upper curve) and intensity $I(\theta)$ (lower curve, in arbitrary units) for the process $Ar + h\nu(21.22\ eV) \rightarrow Ar^+ {}^2P_{1/2} + e^-$. The error bars of the experimental values represent the single statistical error as well as the error of the light-polarization measurements and include, for the upper curve, a contribution for the uncertainty in the Sherman function of the Mott detector. All values shown are normalized to 100% light polarization. Full curves: least-squares fit yielding ξ and β given in Table I.

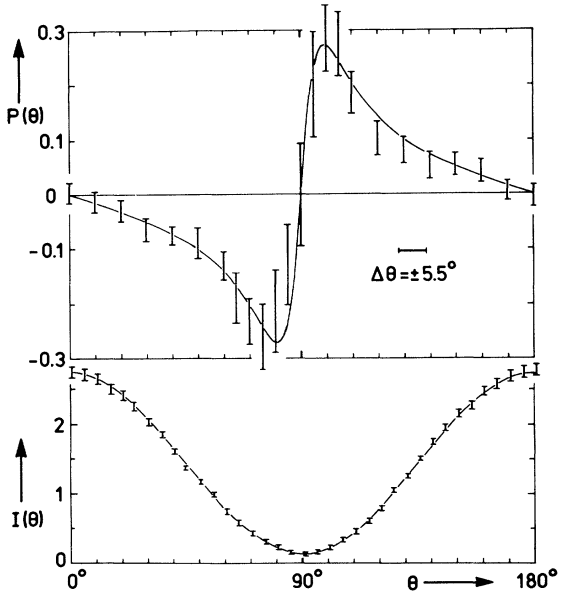

FIG. 3. The same as in Fig. 2 for the process Xe $+h\nu(21.22\ \text{eV}) \rightarrow \text{Xe}^+ {}^2P_{3/2} + e^-$.

stability of the electron beam. Counting rates varied between 2 and 30 per second. Instrumental asymmetries could be eliminated by taking advantage of the reversal of the electron polarization when rotating the light polarizer from θ to $360°-\theta$ [see Eq. (3)]. The overall intensity loss, which is due to the light polarizer, the angular separation, and the Mott-scattering procedure, exceeded seven orders of magnitude. To measure the angular distribution of the photoelectron intensity $I(\theta)$ [which is proportional to the denominator of Eq. (3)], the Mott detector was replaced by a Channeltron multiplier in order to avoid part of the intensity loss.

Figures 2 and 3 show the measured angular distributions $P(\theta)$ and $I(\theta)$ for the two examples $Ar^+ {}^2P_{1/2}$ and $Xe^+ {}^2P_{3/2}$, respectively. The full curves are results of a least-squares fit to the experimental values using Eq. (3) and taking into account the degrees of light polarization P_γ measured simultaneously (with an absolute error of $\pm 4\%$). The parameters ξ and β obtained from the fit procedure are listed in Table I, together with those of the two other final ionic states $Ar^+ {}^2P_{3/2}$ and $Xe^+ {}^2P_{1/2}$. For comparison, results of theoretical calculations as well as some experimental data are cited.[17] The ξ values of Cherepkov[2] have a sign other than predicted because of a sign error in his publication.[18]

A comparison of the measured polarization values with the solid curve [which represents Eq. (1)] shows, within the error limits, that the angular dependence of the photoelectron polarization has the shape predicted.[1-4] The parameters ξ and β giving the best fit agree well with results of other authors, except the ξ parameter of $Xe^+ {}^2P_{3/2}$, which is significantly lower than the value obtained with unpolarized light.[8] The polarization values were reproduced several times, varying the intensity of the atomic beam, always yielding the same results. The overall agreement of the parameters confirms the theoretical predictions.

In Fig. 3 the large β parameter has a strong influence on the shape of the polarization curve, resulting in a shift of the extrema towards $\theta = 90°$. In spite of the relatively small value $\xi = -0.06$, spin polarizations up to $P(\theta) = 0.3$ occur because the differential cross section (denominator of the polarization formula) is very small in the region around 90° (see lower curve). It can be concluded that, if counting rates are sufficiently high, measurement of $P(\theta)$ provides a very reliable method for the determination of both pa-

TABLE I. Angular-distribution parameters ξ and β at 58.43 nm: (a) derived from $P(\theta)$ (this work); (b) measurements with unpolarized light (Heinzmann, Schöhense, and Kessler, Ref. 8); (c) random-phase–approximation experiment calculation (Cherepkov, Ref. 2); (d) relativistic random-phase–approximation calculation (Huang, Johnson and Cheng, Ref. 4); (e) derived from $I(\theta)$ (this work); (f) measurements with linearly polarized light (Hancock and Samson, Ref. 12).

Ionic state	ξ values (a)	(b)	(c)	(d)	β values (a)	(e)	(f)	(c)
$Ar^+ {}^2P_{1/2}$	0.25 ± 0.01	0.264 ± 0.012	0.29	0.03	0.8 ± 0.1	0.89 ± 0.05	0.95 ± 0.02	1.01
$Ar^+ {}^2P_{3/2}$	-0.13 ± 0.01	-0.139 ± 0.007	...	−0.15	0.8 ± 0.1	0.91 ± 0.05		...
$Xe^+ {}^2P_{1/2}$	0.13 ± 0.01	0.141 ± 0.006	0.12	0.15	1.6 ± 0.1	1.58 ± 0.05	1.64 ± 0.04	1.73
$Xe^+ {}^2P_{3/2}$	-0.06 ± 0.01	-0.096 ± 0.007	...	−0.09	1.8 ± 0.1	1.73 ± 0.06	1.78 ± 0.04	...

rameters ξ and β, because fluctuations of light intensity or target density (for example, in metal-vapor beams) do not affect the polarization measurement (ratio of two counting rates simultaneously measured), and apparatus-related asymmetries can easily be eliminated.

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[1]N. A. Cherepkov, Zh. Eksp. Teor. Fiz. 65, 933 (1973) [Sov. Phys. JETP 38, 463 (1974)].
[2]N. A. Cherepkov, J. Phys. B 12, 1279 (1979).
[3]C. M. Lee, Phys. Rev. A 10, 1598 (1974).
[4]K. -N. Huang, W. R. Johnson, and K. T. Cheng, Phys. Rev. Lett. 43, 1658 (1979).
[5]U. Fano, Phys. Rev. 178, 131 (1969), and 184, 250 (1969).
[6]U. Heinzmann, G. Schönhense, and A. Wolcke, in *Proceedings of the International Workshop on Coherence and Correlation in Atomic Collisions, London, 1978, Abstracts of Contributed Papers*, edited by H. Kleinpoppen and J. F. Williams (Plenum, New York, 1979), p. 607.
[7]U. Heinzmann, G. Schönhense, and J. Kessler, Phys. Rev. Lett. 42, 1603 (1979).
[8]U. Heinzmann, G. Schönhense, and J. Kessler, to be published.
[9]J. Kessler, *Polarized Electrons* (Springer, Berlin, 1976).
[10]Unfortunately different definitions of ξ are in use. This Letter is consistent with the polarization formula in Ref. 7 which was originally given by Lee, Ref. 3.
[11]J. Cooper and R. N. Zare, J. Chem. Phys. 48, 942 (1968).
[12]W. H. Hancock and J. A. R. Samson, J. Electron Spectrosc. 9, 211 (1976).
[13]A similar apparatus with a laser light source is currently used by Kaminski, Kessler, and Kollath for investigation of two photon processes in atomic Cs.
[14]U. Heinzmann and G. Schönhense, in *Proceedings of the Eleventh International Conference on the Physics of Electronic and Atomic Collisions, Kyoto, 1979, Abstracts of Papers*, edited by K. Takayanagi and N. Oda (Society of Atomic Collision Research, Kyoto, 1979), p. 24, and to be published.
[15]K. Rabinovitch, L. R. Canfield, and R. P. Madden, Appl. Opt. 4, 1005 (1965).
[16]J. A. R. Samson, Rev. Sci. Instrum. 47, 859 (1976).
[17]For β parameters of other authors see, for example, J. L. Dehmer, W. A. Chupka, J. Berkowitz, and W. T. Jivery, Phys. Rev. A 12, 1966 (1975), and references therein.
[18]N. A. Cherepkov, private communication.

K-Edge Absorption Spectra of Ionic Potassium and Its $Z+1$ Analogy

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If the excited-state energy spectrum of a free atom is such that the energy needed for excitation to a localized orbital is higher than one to a delocalized orbital, the ligand field of the coordination atoms in a molecule may reverse their energy order and violate the $Z+1$ analogy. The comparison of a self-consistent calculation with the experimental K-edge absorption spectra of ionic potassium in molecular complexes shows that the energy order of $3d$ and $4s$ orbitals of potassium is reversed, contrary to the $Z+1$ analogy.

The availability of synchrotron radiation as an x-ray source in recent years has made it possible to measure x-ray absorption spectra with improved accuracy over a wide energy range. As a method for determining the structure of noncrystalline systems, the extended x-ray absorption fine structure (EXAFS) is incomplete. EXAFS, an excellent method for determining interatomic distances, is insensitive to other structural parameters, such as the coordination geometry. The latter information is contained in the absorption-edge spectrum and usage of such edge spectra has been demonstrated.[1-3] Unfortunately, the interpretation of absorption-edge spectra in general requires molecular-orbital calculations, which for large molecules are forbiddingly tedious. However, if the absorbing atom is highly ionic, the effect of the coordination atoms can be treated as a ligand field and the atomic approach is a reasonable approximation for a qualitative understanding of the low-lying absorption peaks.

Under such circumstances, the $Z+1$ analogy is often used as a starting point,[1,2,4] i.e., the $1s \rightarrow nl$ excited-state spectrum of atom Z (nuclear

