Theoretical Study of Spin Polarization of Photoelectrons from Noble Gases

K.-N. Huang and W. R. Johnson
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

and

K. T. Cheng
Argonne National Laboratory, Argonne, Illinois 60439
(Received 9 October 1979)

Recent measurements by Heinzmann *et al.* of the spin polarization of photoelectrons ejected from xenon by unpolarized light are in serious disagreement with various theoretical calculations. This Letter reports an *ab initio* study of spin polarization with use of relativistic random-phase approximation. Excellent agreement is obtained between the present calculations and the experimental measurements by Heinzmann *et al.* for xenon, krypton, and argon.

The prediction[1] that polarized photoelectrons can be ejected from closed-shell atoms by unpolarized light has been verified by recent experiments.[2,3] In the case of xenon atoms a number of theoretical results for the degree of polarization are available. The results of the various theoretical calculations differ from one another; moreover, none of the existing theories agree with the recent measurements of spin polarizations made by Heinzmann, Schönhense, and Kessler.[3] Since the determination of polarization is required for a complete understanding of low-energy photoionization, it is imperative that reasonable agreement be obtained between theory and experiment.

In this Letter we present the results of *ab initio* calculations of the spin polarization of photoelectrons from xenon, krypton, and argon, based on the relativistic random-phase approximation (RRPA). Past experience shows that the nonrelativistic random-phase approximation[4] (RPA) is very successful in explaining cross sections for low-energy photoionization. These cross sections are sensitive to electron-electron correlation, and the RPA includes the correlations most important for low-energy photoionization. By treating relativity along with correlations the RRPA[5] is capable of explaining more refined details of atomic photoionization,[6] such as angular distributions for individual subshells and subshell branching ratios, which are sensitive to both relativistic and correlation effects. Since the RRPA includes the spin-orbit interaction in a nonperturbative manner, it is a particularly appropriate tool for the theoretical treatment of spin polarization. Indeed, we find that the RRPA does describe the measured spin-polarization parameters accurately over the entire range of measured energies. This fact is all the more remarkable since the empirical treatments of spin polarization obtained from multichannel quantum-defect analyses[7-9] of the xenon spectra lead to erroneous values for the spin-polarization parameters. A nonrelativistic RPA calculation of spin polarization by Cherepkov[10a] which involves some use of empirical data appears to have a mistake in sign. If the sign of Cherepkov's spin-polarization parameter is changed, then the RPA is found to agree approximately with the xenon measurements, although not as well as the RRPA.

Spin-polarization formulas for photoelectron have been given by Brehm,[11] Cherepkov,[10b] Lee,[1] and more generally by Huang.[12] We use the notation of Huang[12] and consider specifically the case of photoelectrons ejected by circularly polarized light incident on noble-gas atoms.

The polarization of incident light can be defined in a fixed (at the target atom) coordinate system X, Y, Z. The photoelectron polarization is defined in a rotated coordinate system x, y, z obtained from X, Y, Z by a rotation by Euler angles $(\varphi, \theta, 0)$ as shown in Fig. 1. The angular distribution and spin polarization is given by

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

$$P_x(\theta,\varphi) = \pm \frac{\xi \sin\theta}{1 - \frac{1}{2}\beta P_2(\cos\theta)}, \qquad (2)$$

$$P_y(\theta,\varphi) = \frac{\eta \sin\theta \cos\theta}{1 - \frac{1}{2}\beta P_2(\cos\theta)}, \qquad (3)$$

$$P_z(\theta,\varphi) = \pm \frac{\zeta \cos\theta}{1 - \frac{1}{2}\beta P_2(\cos\theta)}, \qquad (4)$$

where the ± signs are for light with positive or negative helicity. The five dynamical parameters

σ, β, ξ, η, and ζ completely describe low-energy photoionization. These dynamical parameters can be expressed in terms of reduced matrix elements of dipole transition operators.[5] For the present purpose where we consider the spin polarization of photoelectrons from noble-gas atoms by unpolarized light, only the polarization component P_y will be nonvanishing. The corresponding dynamical parameter η is given in terms of reduced matrix elements of the dipole transition operator by the following expression:

(i) For $P_{1/2}$ subshells,

$$\eta = -i\,\frac{3}{2\sqrt{2}}\,\frac{(D_{s_{1/2}}D_{d_{3/2}}{}^* - \text{c.c.})}{|D_{s_{1/2}}|^2 + |D_{d_{3/2}}|^2};$$

(ii) for $P_{3/2}$ subshells,

$$\eta = i\,\frac{[(3/4\sqrt{5})(D_{s_{1/2}}D_{d_{3/2}}{}^* - \text{c.c.}) - (3/2\sqrt{5})(D_{s_{1/2}}D_{d_{5/2}}{}^* - \text{c.c.}) + \tfrac{3}{4}(D_{d_{3/2}}D_{d_{5/2}}{}^* - \text{c.c.})]}{|D_{s_{1/2}}|^2 + |D_{d_{3/2}}|^2 + |D_{d_{5/2}}|^2},$$

where c.c. denotes the complex conjugate of the preceding term. A detailed analysis including all the three spin-polarization parameters ξ, η, and ζ will be presented in a separate paper.

In Fig. 2 we compare our theoretical values of η with the measurements of Heinzmann, Schönhense, and Kessler.[3] We also include in Fig. 2 the results from the multichannel quantum-defect analyses of Lee and Dill[7] and of Geiger,[8,9] as well as the nonrelativistic RPA results of Cherepkov.[10a] We have reversed the sign of Cherepkov's calculation in Fig. 2 since there appears to be a sign error in his published values.

The RRPA amplitudes used to obtain the theoretical curves in Fig. 2 include correlations between the $5p$, $5s$, and $4d$ atomic shells. The resulting RRPA equations involve thirteen interacting channels. Cross sections, angular distributions, and branching ratios obtained from these RRPA amplitudes have been reported elsewhere[6]; these quantities are also in excellent agreement with experiment.

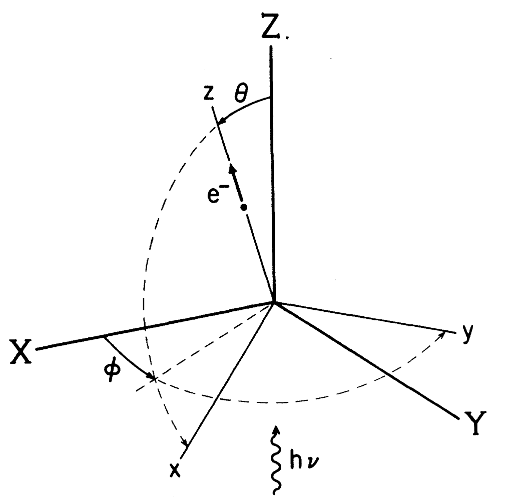

FIG. 1. Geometrical relationships used in spin-polarization formulas. Light is incident along the Z axis, and the photoelectron is ejected along the z axis. The photoelectron spin polarization is defined in the x,y,z coordinate system.

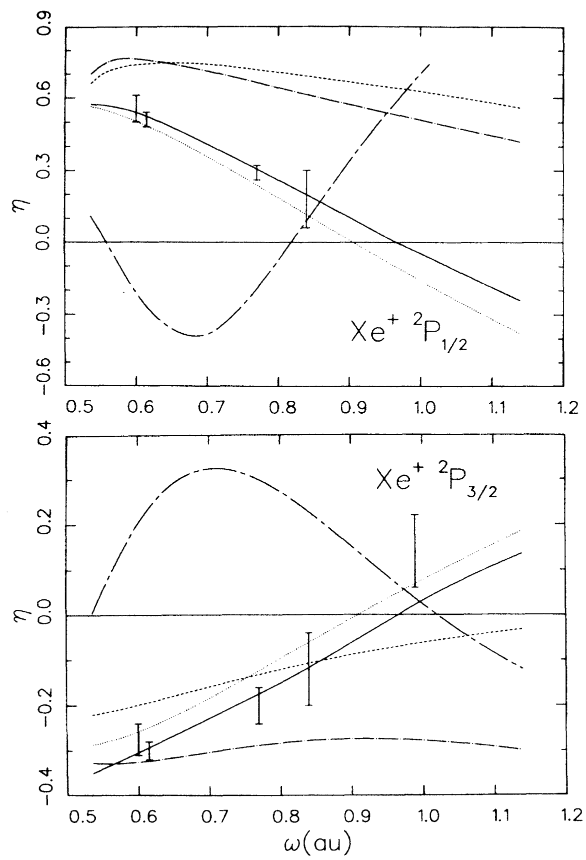

FIG. 2. Spin-polarization parameter η of xenon $5p$-shell photoionization. Experimental data with error bars are from Ref. 3. Curves are theoretical calculations: The solid lines are results using RRPA; The dashed lines are nonrelativistic RPA results from Ref. 10a with the sign reversed; and multichannel quantum-defect results obtained with different sets of parameters: dotted lines, Ref. 7; dash-dotted lines, Ref. 8; and long-short–dashed lines, Ref. 9.

In Fig. 3 the spin-polarization parameter η is given for krypton together with previously unpublished experimental values from Heinzmann, Schönhense, and Kessler.[13] The calculations of the photoionization amplitudes are similar to the xenon calculations; correlations between the three shells $4p$, $4s$, and $3d$ are included in the amplitudes. Again we find that the theoretical spin-polarization calculations are in good agreement with experiment. Finally, in Fig. 4, we compare our RRPA calculation for argon with experimental values of Heinzmann, Schönhense, and Kessler.[13] We also give the nonrelativistic values of Cherepkov[10a] in Fig. 4. Except for the sign mistake mentioned previously, Cherepkov's values are found to be in close agreement with the present values. This close agreement is expected since relativistic effects in argon as measured, for example, by the branching ratio $\sigma_{p3/2}:\sigma_{p1/2}$, are known to be small. In fact, it is apparent from Fig. 4 that the theoretical spin-polarization parameters $\eta_{p_{1/2}}$ and $\eta_{p_{3/2}}$ are in the ratio $-2:1$ which is just the ratio expected nonrelativistically. For krypton and xenon there are increasingly important deviations from the nonrelativistic ratios caused by the spin-orbit interaction.

It is seen that the polarization measurements place severe constraints on the empirical atomic parameters arising from multichannel quantum-defect analyses and that such measurements serve as a rigorous test of dynamical parameters predicted by photoionization theory. Additionally, spin-polarization measurements can help to separate those effects due to correlation from those due to relativity in heavy elements. We have already seen one example in xenon $5p$ photoionization, and we just mention another more striking case in the xenon $5s$ shell where completely correlated nonrelativistic calculations predict no

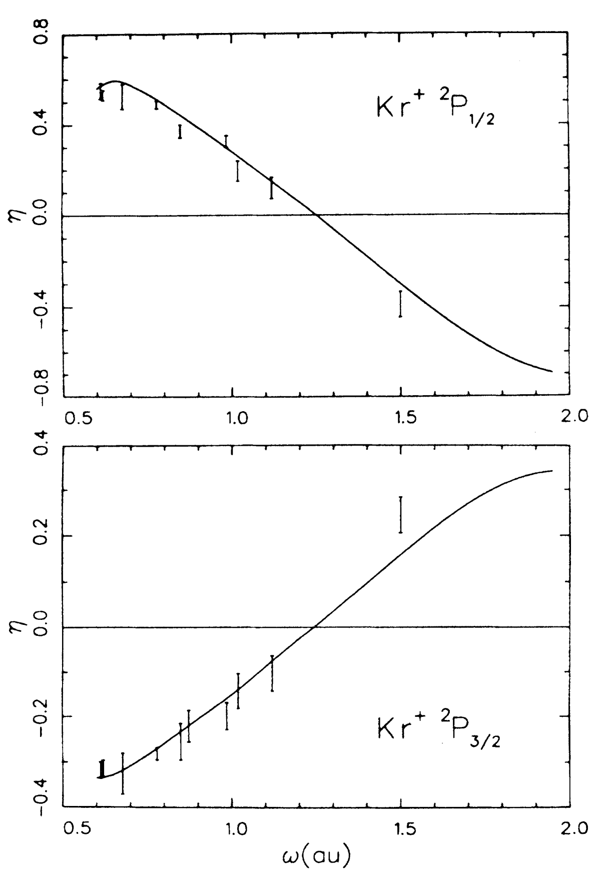

FIG. 3. Spin-polarization parameter η of krypton $4p$-shell photoionization. Data with error bars are unpublished experimental values from Heinzmann *et al.*

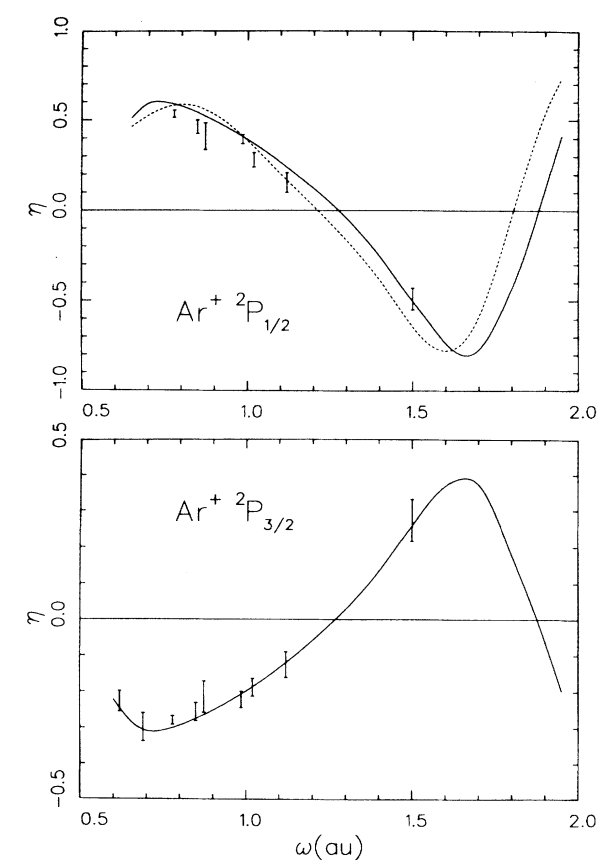

FIG. 4. Spin-polarization parameter η of argon $3p$-shell photoionization. Data with error bars are unpublished experimental values from Heinzmann *et al.* The solid curve is the present RRPA results, and the dashed curve is the nonrelativistic RPA results from Ref. 10a with the sign reversed.

spin polarization, but where correlated relativistic calculations predict very large spin polarization near the cross-section Cooper minimum.[14] To unfold such effects, further measurements of spin polarization are clearly desirable.

The authors express their gratitude to U. Heinzmann for providing experimental data for krypton and argon prior to publication. This research is partially supported by the National Science Foundation under contract No. PHY79-09229 and by the U. S. Department of Energy. One of us (W.R.J.) owes thanks to the Argonne National Laboratory for a Guest Faculty Participant appointment.

[1]C. M. Lee, Phys. Rev. A 10, 1598 (1974).
[2]U. Heinzmann, G. Schönhense, and A. Wolcke, in Abstracts of Contributed Papers and Proceedings of the International Workshop on Coherence and Correlation in Atomic Collisions, London, September, 1978, edited by H. Kleinpoppen and J. F. Williams (Plenum, New York, to be published).
[3]U. Heinzmann, G. Schönhense, and J. Kessler, Phys. Rev. Lett. 42, 1603 (1979).
[4]M. Ya Amusia and N. A. Cherepkov, Case Stud. At. Phys. 5, 47 (1975).
[5]W. R. Johnson and C. D. Lin, Phys. Rev. A 20, 964 (1979).
[6]W. R. Johnson and K. T. Cheng, Phys. Rev. A 20, 978 (1979).
[7]D. Dill, Phys. Rev. A 7, 1976 (1973).
[8]J. Geiger, Z. Phys. A 276, 219 (1976).
[9]J. Geiger, Z. Phys. A 282, 129 (1977).
[10a]N. A. Cherepkov, J. Phys. B 11, L435 (1978).
[10b]N. A. Cherepkov, Zh. Eksp. Teor. Fiz. 38, 933 (1974) [Sov. Phys. JETP 38, 463 (1974)].
[11]B. Brehm, Z. Phys. 242, 195 (1971).
[12]K.-N. Huang, to be published.
[13]U. Heinzmann, G. Schönhense, and J. Kessler, unpublished.
[14]K.-N. Huang and A. F. Starace, to be published; K. T. Cheng, K.-N. Huang, and W. R. Johnson, to be published.

Angular Distribution of Xe $5s \rightarrow \epsilon p$ Photoelectrons near the Cooper Minimum

M. G. White, S. H. Southworth, P. Kobrin, E. D. Poliakoff, R. A. Rosenberg, and D. A. Shirley
Materials and Molecular Research Division, Lawrence Berkeley Laboratory, and Department of Chemistry, University of California, Berkeley, California 94720
(Received 9 October 1979)

Angular distributions of Xe $5s \rightarrow \epsilon p$ photoelectrons were measured with use of pulsed synchrotron radiation and a time-of-flight photoelectron spectrometer. The asymmetry parameters β_{5s} differ markedly from the nonrelativistic value of 2, particularly near the Cooper minimum, in agreement with jj-coupling models. This confirms calculations based on the relativistic random-phase-approximation model of Johnson and Cheng, establishing the importance of the interchannel coupling in $5s \rightarrow \epsilon p$ photoionization.

Yang's theorem[1] gives the angular distribution of photoelectrons induced by linearly polarized radiation via an electric-dipole transition from randomly oriented targets as

$$\frac{d\sigma(\epsilon)}{d\Omega}=\frac{\sigma(\epsilon)}{4\pi}[1+\beta(\epsilon)P_2(\cos\theta)]. \quad (1)$$

Here ϵ is the electron kinetic energy, θ is the angle between the radiation polarization direction and the photoelectron propagation direction, and $\beta(\epsilon)$ is the "asymmetry parameter" that characterizes the photoelectron angular distribution. For the case of photoemission from an s orbital, Bethe[2] and Cooper and Zare[3] showed that in the LS-coupling limit, $\beta \equiv 2$ for all ϵ; i.e., a pure $\cos^2\theta$ distribution is expected. The He($1s \rightarrow \epsilon p$) angular distribution is, in fact, accurately described in this limit; however, a recent measurement by Dehmer and Dill[4] of the Xe($5s \rightarrow \epsilon p$) transition at $h\nu = 40.8$ eV yielded $\beta_{5s} = 1.4 \pm 0.1$. This deviation from the LS result has been attributed to anisotropic final-state interactions between the photoelectron and ion core. Specifically, spin-orbit interactions between the ϵp photoelectron and the Xe^+ core leads to two dipole-allowed continuum channels ($ns_{1/2} \rightarrow \epsilon p_{1/2}$ and $ns_{1/2} \rightarrow \epsilon p_{3/2}$) which can interfere, resulting in $\beta_{5s} < 2$.[4,5] Several theoretical calculations for β_{5s} have appeared in the literature,[5-9] each of which involves different levels of approximation in