SPIN-ORBIT PERTURBATION IN HEAVY ALKALI ATOMS*

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(Received 15 June 1970)

In a photoionization experiment involving polarized alkali atoms and circularly polarized photons we measured the strength of the spin-orbit perturbation in potassium, rubidium, and cesium. Our results are pertinent to the theory of photoionization and to the production of polarized electrons. Extrapolation of the results to photon energies below the ionization threshold provides access to the anomalous doublet line-strength ratio of discrete P states.

The spin-orbit interaction is the cause of various anomalous effects in heavy alkali atoms. In 1930, Fermi showed that spin-orbit perturbation of alkali wave functions was responsible for the anomalous doublet line-strength ratio of the discrete P states.[1] In 1951 Seaton used similar arguments to explain the nonzero minimum of the photoionization cross section of the heavy alkalis.[2] On the basis of the work of Fermi and Seaton, Fano last year predicted that polarized electrons would result from photoionization of unpolarized cesium atoms by circularly polarized photons.[3] Stimulated by Fano's prediction, we experimentally examined the spin-orbit perturbation, which indeed does produce this polarization effect.[4]

Under the assumption that the P-state continuum wave functions are perturbed by a spin-orbit interaction, the radial matrix element for an electric dipole transition from the ground state $(n_0{}^2S_{1/2})$ to the P-state continuum will depend upon the total angular momentum j' of the final P state. The perturbed radial matrix elements can then be written in the form[3]

$$R_1 = R_0 - \tfrac{2}{3}\Delta R,$$
$$R_3 = R_0 + \tfrac{1}{3}\Delta R, \qquad (1)$$

where R_1 and R_3 are the radial matrix elements corresponding to $j' = \frac{1}{2}$ and $\frac{3}{2}$, respectively; R_0 is the unperturbed radial matrix element, and $\Delta R = R_3 - R_1$ is the deviation resulting from the perturbation of the final continuum P state. The various spin-orbit effects in the heavy alkali atoms can be described in terms of the single perturbation function $x(E)$, defined by

$$x(E) \equiv 3R_0(E)/\Delta R(E) = (2R_3 + R_1)/(R_3 - R_1), \qquad (2)$$

where E is the energy of the photon incident on the atom.

Access to the perturbation function $x(E)$ in the continuum is provided by any one of three polarization experiments, each of which involves the photoionization process, $A + \gamma \rightarrow A^+ + e^-$. To a very good approximation the spin-orbit interaction does not perturb the nuclear spin.[3] Therefore, the particle polarizations which characterize the polarization effects in the photoionization process are the circular polarization of the incident photon, P_{ph}; the electronic polarization of the atom, P_{at}, parallel to the direction of the incident photon; and the polarization of the final-state electron, P_{el}, also parallel to the direction of the incident photon. If any two of these three particle polarizations are experimentally controlled, the perturbation function $x(E)$ can be determined.

Photoionization of unpolarized atoms with circularly polarized photons, followed by measurement of the photoelectron polarization, results in the determination of the polarization parameter, P, given by

$$P = [P_{\mathrm{el}}/P_{\mathrm{ph}}]_{P_{\mathrm{at}}=0} = (2x+1)/(x^2+2). \qquad (3)$$

Photoionization of polarized atoms with circularly polarized photons, followed by measurement of the ion counting-rate asymmetry $\delta = (I^+ - I^-)/(I^+ + I^-)$, where I^+ and I^- are the counting rates corresponding to left- and right-circular photon polarization, respectively, results in the determination of the polarization parameter Q, given by

$$Q = \delta/(P_{\mathrm{at}}P_{\mathrm{ph}}) = (2x-1)/(x^2+2). \qquad (4)$$

Finally, photoionization of polarized atoms with unpolarized photons, followed by measurements of the photoelectron polarization, results in the determination of the polarization parameter R, given by

$$R = [P_{\mathrm{el}}/P_{\mathrm{at}}]_{P_{\mathrm{ph}}=0} = x^2/(x^2+2). \qquad (5)$$

Equations (3)-(5) follow directly from Eqs. (1) and (2) and the appropriate Clebsch-Gordan coefficients.

For purposes of optimization of signal, elimination of molecules, and suppression of back-

Table I. Calculated and measured atomic polarization.

	K^{39}	K^{41}	Rb^{85}	Rb^{87}	Cs^{133}
Abundance (%)	93.1	6.9	72.15	27.85	100
P_{at} obtained from calculation:					
Single isotope	0.259	0.267	0.168	0.251	0.125
Isotope mixture	0.260		0.191		0.125
Interpretation[a]	Exact value		Approximation, lower limit		Lower limit
P_{at} obtained from data analysis	0.254 ± 0.014[b]		0.203 ± 0.007		0.142 ± 0.002

[a]Interpretation depends on the hyperfine-structure interaction of the alkali atom.
[b]Error refers to one standard deviation.

ground photoelectrons, we chose to measure $Q(E)$ in order to determine the perturbation function, $x(E)$. Our first measurements with cesium[4] confirmed the existence of the polarization effect predicted by Fano, but they were not yet accurate enough to determine $x(E)$ quantitatively. Since then, we have measured $x(E)$ more accurately not only for cesium, where the spin-orbit interaction is strongest, but also for rubidium and potassium.

A beam of alkali atoms was polarized by passage through a sexipole magnet. Beyond the magnet, the atoms were ionized by circularly polarized photons in a weak magnetic field of 2.5 G parallel to the direction of photon incidence. A high-pressure xenon arc lamp was used in conjunction with a grating monochromator as the source of uv photons. The combination of linear polarizer and quarter-wave plate produced a circular polarization which varied between 0.82 and 0.91 in the range of photon energies used. The alkali ions extracted from the ionization region were detected by a Channeltron electron multiplier. Use of a central beam stop at the entrance of the sexipole magnet eliminated molecules from the alkali beam.

The perturbation function $x(E)$ passes through zero at the photon energy E_0, proximate to the energy where the photoionization cross section has its minimum. In the vicinity of E_0, $x(E)$ can be written in the form of a power series

$$x(E) = c_1(E-E_0) + c_2(E-E_0)^2 + c_3(E-E_0)^3 + \cdots. \quad (6)$$

In analyzing our data we neglected all terms of order higher than $(E-E_0)^3$. We employed a multiparameter regression-analysis computer program[5] to fit the function $Q(E)$ to our measurements of δ/P_{ph}, thereby directly determining the five parameters P_{at}, E_0, c_1, c_2, and c_3 as well as their errors and error correlations. The values of P_{at} are given in Table I together with the corresponding results of a calculation based on ideal state selection in the sexipole magnet.[6]

In Fig. 1 are shown the data points

$$Q_i = \delta(E_i, \Delta\lambda)[1+\eta(\Delta\lambda)]/[P_{ph}(E)P_{at}], \quad (7)$$

where $\delta(E_i, \Delta\lambda)$ is the ion counting-rate asymmetry obtained with the finite wavelength bandwidth $\Delta\lambda$, $\eta(\Delta\lambda)$ is a very small correction term which approximates the effect of the variation of the cross section over the wavelength interval $\Delta\lambda$,[7] and $P_{ph}(E_i)$ is the photon polarization measured for the photon energy E_i. The vertical error bars for one standard deviation were calculated from signal and background counting rates and the counting times. The horizontal error bars represent the uncertainty in the monochromator dial setting.

The $Q(E)$ curves, which were obtained from a cubic fit for $x(E)$, are given in Fig. 1 together with the values of the parameters E_0 and $(dE/dx)_0 = 1/c_1$. The nonlinearity in $x(E)$, governed by the coefficients c_2 and c_3, can be seen in Fig. 2. The bands of the $x(E)$ curves are determined by the functions $x+\delta x$ and $x-\delta x$, where δx is one standard deviation. Only in the case of potassium is the nonlinear part of $x(E)$ larger than the error δx. In the case of cesium, δx is small enough to permit meaningful extrapolation far into the discrete spectrum.

Below threshold, the physically allowed values of the perturbation function, $x(E)$, occur at discrete energy intervals. However, the function itself is continuous and passes through threshold continuously, as in fact do the functions for the quantum defect and the line strength per unit en-

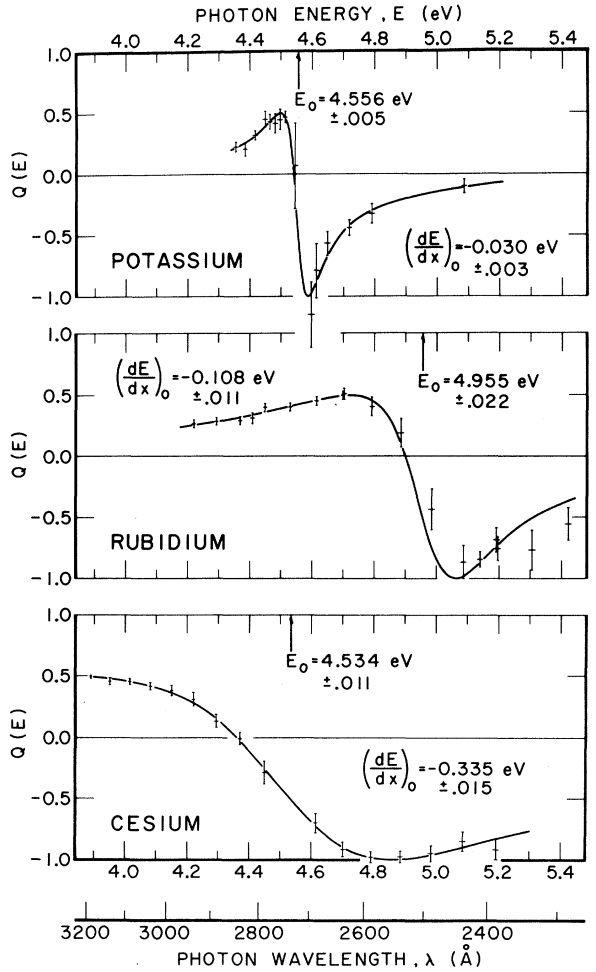

FIG. 1. Polarization parameter Q as function of photon energy E for potassium, rubidium and cesium.

ergy interval.[8] At the lower end of the discrete spectrum, values of x can be obtained from measurements of the doublet line-strength ratio ρ given by

$$\rho \equiv S(n P_{3/2})/S(n P_{1/2}) = 2(R_3^2/R_1^2) = 2(x+1)^2/(x-2)^2, \quad (8)$$

where $S(n P_{1/2})$ and $S(n P_{3/2})$ are the line strengths of the doublet. At energies closer to threshold, however, the doublets cannot be resolved spectroscopically. Thus the extrapolation of $x(E)$ from the continuum into the discrete spectrum of cesium represents the first experimental determination of the spin-orbit perturbation of P states of high quantum number n.

From Fig. 2 it can be seen that for cesium the value of the energy corresponding to $x=+2$ lies below threshold. Since $R_1=0$ when $x=+2$, the function $\rho(E)$ has a pole in the discrete spectrum. The extrapolated curve in Fig. 2 indicates that

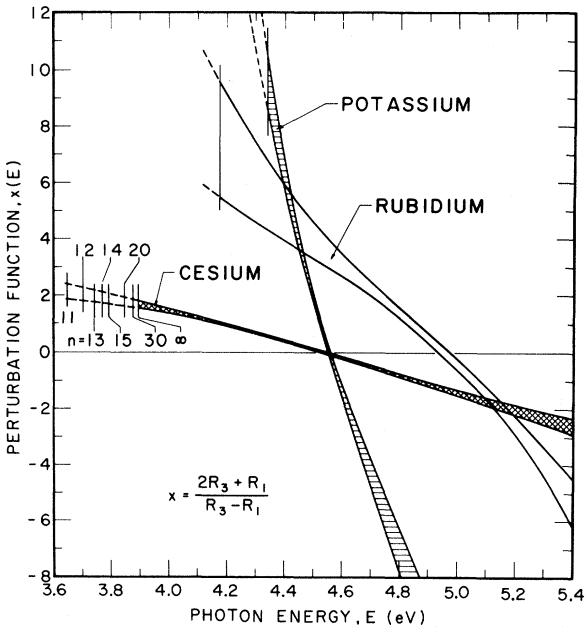

FIG. 2. The perturbation function $x(E)$ for the three alkali atoms. The width of the error bands corresponds to one standard deviation in x. Extrapolation below threshold is shown by dashed lines; n is the principal quantum number of the discrete P state.

the pole definitely lies below $n=20$ and probably occurs in the vicinity of $n=12$. This result contradicts the spectroscopic data of Kratz,[9] who observed a monotonic increase of $\rho(E_{n,P})$ from $n=6$, the lowest principal quantum number of cesium, to $n=21$, the upper limit determined by his spectral resolution. Kratz rejected the validity of a set of spectroscopic measurements which Sambursky[10] had performed twenty years earlier, in 1928. Sambursky's data, in fact, indicate the presence of a pole in $\rho(E)$ between $E_{10,P}$ and $E_{11,P}$, in good agreement with Fig. 2! For potassium and rubidium, the $x(E)$ curves of Fig. 2 show that the pole in $\rho(E)$ occurs above threshold; therefore, $\rho(E_{n,P})$ increases monotonically with n in the discrete spectrum. The threshold value of the doublet line-strength ratio, $\rho(E_{n\to\infty,P})$, is 3.85 ± 0.45 for potassium, 5.5 ± 1.5 for rubidium, and 250 ± 190 for cesium.

Since experiments with polarized electrons are being contemplated in many laboratories, the techniques for producing polarized electrons are of great current interest.[11] From the perturbation function, $x(E)$, the polarization parameters $R(E)$ and $P(E)$ which determine the polarization of the outgoing photoelectron can be calculated according to Eqs. (5) and (3), respectively. In Fig. 3 are shown $R(E)$ and $P(E)$ for cesium, the

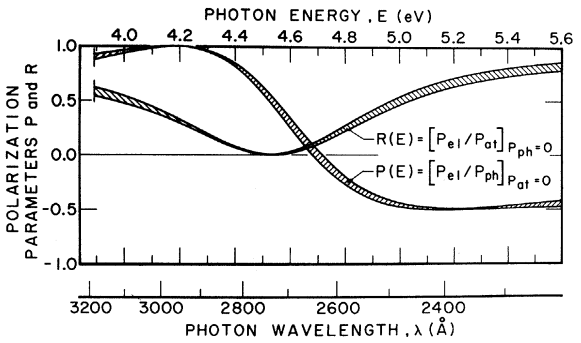

FIG. 3. Polarization parameters $R(E)$ and $P(E)$ for cesium. The width of the error bands corresponds to one standard deviation in the ordinate.

alkali with the most pronounced spin-orbit perturbation.

The electron polarization obtainable with sources based on ionization of polarized alkali atoms by unpolarized photons is given by $P_{el}=R(E)P_{at}$. As shown in Fig. 3, the curve $R(E)$ for cesium is substantially smaller than unity over the whole photon-energy range and, in fact, drops to zero at the photon energy $E_0=4.53$ eV. Therefore, the use of cesium in this type of polarized-electron source will not lead to highly polarized electrons.[12]

The electron polarization obtainable with sources based on ionization of unpolarized alkali atoms by circularly polarized photons is given by $P_{el}=P(E)P_{ph}$. Following Fano's suggestion,[3] Kessler and Lorenz recently measured the polarization of electrons produced with unpolarized cesium atoms and polarized but unfiltered light from a mercury-arc lamp. They obtained an average electron polarization of 0.65 ± 0.15, consistent with their estimate of 0.8.[13] Our result for the polarization parameter $P(E)$ is shown in Fig. 3. This curve provides a quantitative basis for the design of a Fano-type polarized electron source. Since the threshold value, $P(E_{th})=0.91\pm0.03$, is close to unity, photoionization near threshold appears to be a promising method for the production of highly polarized electrons occupying minimum phase space.

The authors gratefully acknowledge the encouragement and support given by Professor V. W. Hughes.

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*Work supported in part by the U. S. Office of Naval Research.

†NATO Postdoctoral Fellow.

‡U. S. Atomic Energy Commission Postdoctoral Fellow.

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[5]A large part of the computer program was provided by Dr. D. L. Mader and Dr. W. H. Wing of Yale University.

[6]If the electronic and nuclear spins of the atoms are completely decoupled inside the sexipole magnet, P_{at} can be calculated exactly. The decoupling was almost complete in the experiment because the central beam stop blocked the low-field region of the sexipole magnet.

[7]All measurements were made with $\Delta\lambda=16$ Å, except for three rubidium measurements which were made with $\Delta\lambda=32$ Å. The correction term, $\eta=[(dx/d\lambda)\Delta\lambda]^2/[3(x^2+2)]$, always lay within $0\leq\eta<0.015$.

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[12]For sources using lighter alkali atoms, this depolarization effect is not important.

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