Spin polarization of photoions produced during ionization of unpolarized atoms

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Polarization of electrons and nuclei during photoionization of unoriented atoms by circularly polarized radiation may be accompanied by a preferential orientation of the electron-shell spin of the atomic residue. The resulting magnetization of the positive ions is similar in nature and magnitude to the magnetization of the electrons. The relation between the degrees of polarization of the electrons and ions is determined and formulas are derived for calculating the polarizations for atoms with ns^2 and np^6 outer shells.

INTRODUCTION

Particle polarization that accompanies the ionization of unoriented atoms by optical radiation has attracted much interest in recent years and has been discussed in detail in the literature. The polarization of photoelectrons has been thoroughly studied; the theoretical and experimental work in this area is reviewed in Refs. 1 and 2, where the applied and experimental implications of the results are discussed. In addition to the practical use of preferential spin orientation, such as generation of beams of polarized electrons, this phenomenon can also be exploited to study the fundamental properties of atoms. For example, together with the anisotropy in the angular distribution of the photoelectrons, the polarization can be used to ascertain how correlation and relativistic effects affect the internal structure of atoms.[3]

Preferential spin orientation due to spin-orbit interaction is most pronounced in atoms with a well-defined fine structure. For example, a sufficient condition for single-photon ionization to be accompanied by preferential orientation is that the initial or final states of the ionization transition have a sufficiently pronounced fine structure. However, polarized photoelectrons cannot be produced in significant amounts unless the frequency of the ionizing radiation is nearly resonant with the autoionizing states; since these generally lie deep in the single-electron continuous spectrum,[4] intense monochromatic vacuum ultraviolet sources are required.

Multiphoton optical resonant transitions[5] in which the intermediate states have a fine structure may be more effective. In this case the intense ionizing radiation shifts and splits the atomic energy levels, and this may either enhance or diminish the polarization. For instance, if the fine-structure splitting of the excited resonance state is small, the light-induced splitting of the ground state will be primarily responsible for the polarization; if the splitting of the excited state is large, its perturbation by the light field may appreciably decrease the polarization.[7,8]

In addition to polarized photoelectrons, the possibility of using hyperfine atomic effects to polarize nuclei has also been considered.[1,6,9] However, the weakness of the hyperfine interaction and the smallness of the associated nuclear magnetic moments make it difficult to observe the nuclear polarization experimentally, even when the nuclei are completely polarized.

Although much work has been done on polarization accompanying the ionization of atoms, the polarization of the ions that remain after ejection of the photoelectron and have paramagnetic electron shells has not been discussed. The probable reason is that the studies to date have been concerned with ionization of simple monovalent atoms, whose positive ions are diamagnetic. A paramagnetic ion, just as the photoelectron, may be polarized.

The mechanism of photoion polarization is the same as for photoelectrons. The circularly polarized radiation at the resonance frequency preferentially ionizes those atoms whose spins lie along a given direction. If the atomic spin $S = s_0 + s$ is the sum of the ion and valence (photo-) electron spins s_0 and s, the orientation of any two of the vectors S, s_0, s will be uniquely determined by the orientation of the third.

There is thus between the photoelectron and photoion polarizations a one-to-one correspondence which should have immediate practical applications. In particular, because the internal atomic structure can be equally well described by the photoelectron or photoion polarization, only the latter need be analyzed. Moreover, this correspondence also provides a way of indirectly measuring the polarization of the photoelectrons in terms of the ion polarization. This has several advantages over conventional methods, where serious difficulties are encountered (Refs. 1 and 2 may be consulted for discussions and references to original work).[1)] These two important applications, combined with the fact that ionized particles can be prepared in states with a specified spin orientation, provide the motivation for the following detailed investigation of the polarization of photoions associated with the ionization of unoriented atoms.

Specific calculations will be discussed for divalent atoms (e.g., helium, mercury, etc.) and inert-gas atoms with the ns^2 $^1S_0$ and np^6 $^1S_0$ outer-shell configurations in the ground state, respectively. We discuss formulas for calculating the ion polarization and present for the electron polarization general expressions which lead to the familiar relations for alkali-metal[5] and multivalent atoms.

We note that in addition to enhancing the fine structure, (which plays a major role in the polarization[5]) of the excited atomic state as compared with the alkali metals, the nonzero spin of the positive ion also causes the polarization to depend on the matrix elements of the ionization transition (there is no such dependence for the alkali metals, which

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have energy levels with a simple doublet structure). These features will be pointed out when we discuss the specific theoretical results.

We will employ throughout the one-electron approximation which is valid for atoms interacting with an external electromagnetic field at optical frequencies.[11] For simplicity we will neglect the shifting and broadening of the atomic lines by the ionizing field.

SPIN-DIRECTION DISTRIBUTION AND POLARIZATION OF PHOTOELECTRONS AND IONS

We will analyze the ionization of atoms by right-circularly polarized radiation of frequency ω and electric field vector

$$\mathbf{F}(t)=2F\,\mathrm{Re}\{\mathbf{e}_1\exp[i(kz-\omega t)]\},$$

where $\mathbf{e}_1$ is the cyclic unit vector (the z axis points along the wave vector $\mathbf{k}$). The probability

$$dW/d\Omega=2\pi|M_{fi}|^2 \tag{1}$$

that the radiation will ionize the atom depends on the matrix element for an N-photon transition, which is of the form

$$M_{fi}=\langle f|\hat{V}G\hat{V}\ldots\hat{V}G\hat{V}|i\rangle\,. \tag{2}$$

Here we employ atomic units $e=m=\hbar=1$; $|i\rangle$ and $|f\rangle$ are the initial and final states, and G is the Green function for the unperturbed atom. Since we are interested in the orientation of the photoparticle spins, we must express the final state $|f\rangle$ as a product of the wave function $\Psi_{E\mu}$ of a photoelectron of energy $E>0$ and the wave function $\Phi_{s_0\mu_0}$ of the remaining ion, with definite spin projections μ and μ_0 on the z axis. We can expand $|f\rangle$ in a complete set of atomic eigenfunctions $\varphi_{E,lJM}$ with fixed orbital and total angular momenta l and J:

$$|f\rangle=\Psi_{E\mu}\Phi_{s_0\mu_0}$$

$$=\sum_{SM_S}\sum_{lmJM}i^l e^{-i\delta_{lJ}}C^{SM_S}_{s_0\mu_0{}^1\!/_2\mu}C^{JM}_{lmSM_S}Y_{lm}^*(\mathbf{k}_f)\varphi_{E,lJM}(\mathbf{r}). \tag{3}$$

Here we have used the LS-coupling approximation and assumed for simplicity that only the spin of the ion is nonzero (the orbital angular momentum vanishes).

If we use the jl-coupling approximation to describe the states of the outer electron in the atom, the final-state wave function takes the form

$$|f\rangle=\Psi_{E\mu}\Phi_{j_0\mu_0}$$

$$=\sum_{KM_K}\sum_{lmJM}i^l e^{-i\delta_{lJ}}C^{KM_k}_{j_0\mu_0lm}C^{JM}_{KM_k{}^1\!/_2\mu}Y_{lm}^*(\mathbf{k}_f)\varphi_{E,lJM}(\mathbf{r}). \tag{4}$$

As usual, we have assumed that the photoelectron wave function is a superposition of a plane wave and a spherical incoming wave of momentum k_f; the $C^{c\gamma}_{a\alpha b\beta}$ are Clebsch-Gordan coefficients.

We now substitute the explicit expressions for the wave functions and the dipole interaction operator

$$\hat{V}=-F(\mathbf{e}\mathbf{d})e^{-i\omega t}$$

for the interaction of the atom with the field, where $\mathbf{d}$ is the atomic dipole moment operator. After integrating the composite matrix element (2) over the angular variables and over the emergence angles of the photoelectron, we get an expression for the probability that N-photon ionization will result in a specified polarization. Thus,

$$W_\mu={}^1\!/_2W+\mu w^e \tag{5}$$

gives the photoelectron emission probability when the projection $\mu=\pm 1/2$ of the spin vector s is specified. We have an analogous expression

$$W_{\mu_0}=\frac{W}{2s_0+1}$$

$$+\sum_{p=1}^{\min(2s_0,2N)}(-1)^{s_0+\mu_0+1}\left[\frac{3s_0}{(s_0+1)(2s_0+1)}\right]^{1/2}C^{p0}_{s_0-\mu_0s_0\mu_0}w_p{}^i \tag{5'}$$

for the probability of photoionization that leaves the ion with a definite projection μ_0 of its spin. Here

$$W=\sum_\mu W_\mu=\sum_{\mu_0}W_{\mu_0}$$

is the total probability for N-photon ionization. The quantities w^e and w_p^i thus characterize the ionization probability distribution for states with a specified photoelectron and photoion polarization.

If we define the degree of polarization as the average projection of the particle spin divided by the maximum possible value ($\mu_{\max}=s=1/2$, $\mu_{0\max}=s_0$), we obtain

$$P^e=w^e/W,\quad P^i=w_1{}^i/W. \tag{6}$$

The polarizations P^e and P^i thus characterize w^e and w^i, and the latter clearly determine the ratio P^e/P^i. In what follows we will use the LS- and jl-coupling schemes to examine the behavior of these quantities. We observe that $\bar{\mu}_0$ is independent of w_p^i at $p\neq 1$ (in theory, only the higher-order moments $\overline{\mu_0{}^p}$ of the random variable μ_0 can depend on w_p^i at $p>1$). We will therefore consider only the quantities $w_1{}^i$ in what follows.

THE LS-COUPLING APPROXIMATION

In this approximation we can express the quantities W, w^e, and $w_1{}^i$ determining the probabilities (5) and (5') as

$$W=2\pi F^{2N}\sum_{Jl}|A_{Jl}|^2, \tag{7}$$

$$w^e=2\pi F^{2N}(-1)^{s_0+3/2+J_t+l_t}(2S+1)$$
$$\times\left[\frac{6N(2N+1)}{N+1}\right]^{1/2}\begin{Bmatrix}1 & S & S\\ s_0 & 1/2 & 1/2\end{Bmatrix}$$
$$\times\sum_{lJJ'}[(2J+1)(2J'+1)]^{1/2}\begin{Bmatrix}1 & J & J'\\ l & S & S\end{Bmatrix}\begin{Bmatrix}1 & J & J'\\ J_i & N & N\end{Bmatrix}A_{Jl}A^*_{J'l}, \tag{8}$$

$$w_1{}^i=w^e\left[\frac{(s_0+1)(2s_0+1)}{6s_0}\right]^{1/2}\begin{Bmatrix}1 & S & S\\ 1/2 & s_0 & s_0\end{Bmatrix}\begin{Bmatrix}1 & S & S\\ s_0 & 1/2 & 1/2\end{Bmatrix}^{-1}. \tag{9}$$

Here

$$A_{Jl}\equiv[3(2J+1)]^{-1/2}\sum_{J_1\ldots J_{N-1}}R_{lJ,J_{N-1}\ldots J_1,l_iJ_i}$$
$$\times\prod_{k=1}^{N-1}(-1)^{J_k+J_t}(2k+1)^{1/2}\begin{Bmatrix}1 & k & k+1\\ J_i & J_{k+1} & J_k\end{Bmatrix},$$
$$R_{lJ,J_{N-1}\ldots J_1,l_iJ_i}=\langle lJ\|\mathbf{d}G_{J_{N-1}}\mathbf{d}\ldots\mathbf{d}G_{J_1}\mathbf{d}\|l_iJ_i\rangle, \tag{10}$$

where R is the reduced composite matrix element for the ionization transition; l_i and J_i are the orbital angular momentum of the valence electron and the total angular momentum of the ground-state atom; the curly brackets denote the Racah 6*j*-symbol.

According to (9), w^e and $w_1{}^i$ [and hence also P^e and P^i, cf. Eq. (6)] are linearly related in the *LS*-coupling scheme; the relation between P^e and P^i can be expressed in the form

$$P^i=P^e,\qquad S=s_0+1/2, \tag{11a}$$
$$P^i=-P^e(s_0+1)/s_0,\qquad S=s_0-1/2. \tag{11b}$$

These results have a simple interpretation if we recall that the dipole interaction between the atom and the ionizing radiation leaves both the total spin vector **S** and its electron and ion components **s**, $\mathbf{s}_0$ unchanged. We have already noted in the Introduction that when radiation with specified polarization and frequency preferentially ionizes atoms whose total spin **S** lies along a given direction, the component spins **s** and $\mathbf{s}_0$ of the dispersing particles should also have a preferred orientation. The relation (11) between P^i and P^e follows as a consequence. In particular, it is clear why $P^i=P^e$ for $S=s_0+1/2$ (parallel spins) while $P^i=-P^e$ for $S=s_0-1/2$ (opposite spins). Equations (8) and (9) imply that $w^e=w_1{}^i=0$ for singlet states (total atomic spin $S=0$), i.e., there is no polarization.

As an illustration we will derive expressions for P^i and P^e for ionization of triplet ($S=1$) states of atoms with two outer (valence) electrons. We will consider some specific examples of multiphoton ionization ($N\geqslant 2$) and omit the case of one-photon ionization, for which appreciable polarization can occur only near the minimum of the photoionization cross section (the Fano effect).

1) *Two-photon ionization of* n^3P_0*-states*. In this case, Eqs. (6)–(11) yield

$$P^i=P^e=\frac{(5\rho_{10}+\rho_{12})^2-36\rho_{32}{}^2}{(5\rho_{10}+\rho_{12})^2+54\rho_{32}{}^2},\qquad \rho_{ll_1}\equiv\langle El|rg_{l_1}r|n^3P_0\rangle; \tag{12}$$

where ρ is the radial composite matrix element for the two-photon ionization transition and q_{l_1} is the partial radial Green function for the subspace of states in which the valence electron has a specified orbital moment l_1. According to (12), $P^i=P^e\approx 1$ near the resonances for the $^3S_1$-states, for which $\omega\approx\omega(n'\,^3S_1)$ and $|\rho_{10}|\gg|\rho_{12}|$. If $\omega\approx\omega(n'\,^3D_1)$ and $|\rho_{12}|\gg|\rho_{10}|$, we have

$$P^i=P^e\approx-(36-x^2)/(54+x^2),$$

$$x\equiv\langle E^3P_2|r|n'\,^3D_1\rangle/\langle E^3F_2|r|n'\,^3D_1\rangle.$$

Here x is the ratio of the matrix elements for the transition from the $n'\,^3D_1$ resonance state to the $^3P_2$ and $^3F_2$ states in the continuous spectrum.

If we neglect the fine splitting of the $n'\,^3D_J$ states, the expressions for $P^i=P^e$ for ionization from the $n\,^3P_2$ and $n\,^3P_1$ states agree with (12) apart from a factor of $-1/2$ and $1/2$, respectively. This shows that even though Eq. (12) implies that $|P_J^{e(i)}\approx 1$, the polarization of the photoparticles may be insignificant well away from resonance if the populations of the $n\,^3P_J$ sublevels are equal (because $\Sigma_{J=0}^2(2J+1)P_J^{e(i)}=0$). Near a resonance, however, only one of the $n\,^3P_J$ states can be selectively ionized, so that $P^{e(i)}$ may differ appreciably from zero.

2) *Two-photon ionization of* $n\,^3S_1$*-states*. In this case we cannot neglect the fine structure of the intermediate $n'\,^3P_J$ states, because their splitting is primarily responsible for the polarization. The general expression for $P^{e(i)}$ is quite elaborate; we will give the results here only for the case of exact resonances:

$$\text{a)}\quad P^{e(i)}=(x^2-1)/(x^2+1),\qquad \omega=\omega(n'\,^3P_0),$$
$$\text{b)}\quad P^{e(i)}=(5x^2-2)/(5x^2+16),\qquad \omega=\omega(n'\,^3P_1),$$
$$\text{c)}\quad P^{e(i)}=(50x^2+2389)/(50x^2+3160),\qquad \omega=\omega(n'\,^3P_2),$$

where

$$x\equiv\langle E^3S_1|r|n'^3P_J\rangle/\langle E^3D|r|n'^3P_J\rangle$$

is the ratio of the radial matrix elements for the transitions from the $n'\,^3P_J$ resonance state to the $^3S_1$ and ^3D states of the continuous spectrum.

3) *Three-photon ionization of* $n\,^3S_1$*-states*. Here the expressions for the polarization become even more complicated because both one- and two-photon resonances can occur. Considering only the case of exact resonance, we have the expressions

$$\text{a)}\quad P^{e(i)}=\frac{(5x_0+x_2)^2-36}{(5x_0+x_2)^2+54},\qquad \omega=\omega(n'\,^3P_0),$$
$$\text{b)}\quad P^{e(i)}=\frac{7(5x_0+x_2)^2-27}{7(5x_0+x_2)^2+108},\qquad \omega=\omega(n'\,^3P_1),$$
$$\text{c)}\quad P^{e(i)}=\frac{7(5x_0+x_2)^2+10278}{7(5x_0+x_2)^2+13608},\qquad \omega=\omega(n'\,^3P_2),$$

for one-photon resonances involving the $n\,{}^3P_J$ levels; here

$$x_l \equiv \langle E^3P|rg_lr|n'\,{}^3P_J\rangle / \langle E^3F|rg_2r|n'\,{}^3P_J\rangle$$

is the ratio of the composite matrix elements for the second-order transition from the resonance to the continuum. Similarly, for two-photon resonances we find that

$$\text{a)}\ P^{e(i)} = \frac{x^2-36}{x^2+54}, \quad 2\omega = \omega(n'\,{}^3D_1),$$

$$\text{b)}\ P^{e(i)} = \frac{7x^2-28}{7x^2+218}, \quad 2\omega = \omega(n'\,{}^3D_2),$$

$$\text{c)}\ P^{e(i)} = \frac{882x^2+253042}{882x^2+276003}, \quad 2\omega = \omega(n'\,{}^3D_3),$$

where

$$x \equiv \langle E^3P|r|n'\,{}^3D_J\rangle / \langle E^3F|r|n'\,{}^3D_J\rangle$$

is the ratio of the matrix elements from the $n'{}^3D_J$ resonance state to the ${}^3P$ and ${}^3F$ continuum states.

The above formulas show that the polarization $P^{e(i)}$ is ≈ 1 and is almost independent of x near the one-photon and two-photon resonances involving the ${}^3P_2$ and ${}^3D_3$ states, respectively. By contrast, $P^{e(i)}$ is quite sensitive to x near the ${}^3P_{0,1}$ and ${}^3D_{1,2}$ resonances. This sensitivity can clearly be exploited to determine the simple and composite matrix elements for electromagnetic transitions in atoms.

Relations similar to the ones above can also be derived for ionization from other atomic states. In all cases the polarization is zero unless the initial or intermediate state has a fine splitting; in this case, values $P^{e(i)} \approx 1$ can be achieved at the resonance frequencies. Specific calculations indicate that the spin-orbit interaction for states in the continuous spectrum is always negligible for $N \geqslant 2$.

THE *jl*-COUPLING APPROXIMATION

This case is more complicated, because the valence electron spin is not directly related to the spin of the ion residue; instead, we have $\mathbf{K} = \mathbf{j}_0 + l$, $\mathbf{J} = \mathbf{K} + \mathbf{s}$, where l is the orbital angular momentum of the valence electron and the intermediate angular momentum ($\mathbf{K}$). Simple general relations of the type (9), (11) therefore cannot be derived.

We now substitute expansion (4) into (2), (1), carry out the integration over the angular variables and over the emergence angle of the photoelectron, average over the initial states, and sum over the final states to get an expression of the form (5). Here s_0 is understood to be the total angular momentum j_0 of the residue and μ_0 is its projection on the z axis. We find that

$$W = 2\pi F^{2N} \sum_{lKJ} |A_{lKJ}|^2, \tag{13}$$

$$w^e = 2\pi F^{2N}\left[\frac{6N(2N+1)}{N+1}\right]^{1/2} \times \sum_{lKJJ'} (-1)^{J_l+K+1/2+N}[(2J+1)(2J'+1)]^{1/2} \times \begin{Bmatrix} 1 & J & J' \\ K & 1/2 & 1/2 \end{Bmatrix} \begin{Bmatrix} 1 & J & J' \\ J_i & N & N \end{Bmatrix} A_{lKJ}A^*_{lKJ'}, \tag{14}$$

$$w_1{}^i = 2\pi F^{2N}\left[\frac{N(2N+1)(j_0+1)(2j_0+1)}{j_0(N+1)}\right]^{1/2} \times \sum_{lKJK'J'} (-1)^{j_0+J_l-1/2+l_l+J+J'+K+K'} \times [(2J+1)(2J'+1)(2K+1)(2K'+1)]^{1/2} \times \begin{Bmatrix} 1 & J & J' \\ 1/2 & K' & K \end{Bmatrix} \begin{Bmatrix} 1 & J & J' \\ J_i & N & N \end{Bmatrix} \times \begin{Bmatrix} 1 & K & K' \\ l & j_0 & j_0 \end{Bmatrix} A_{lKJ}A^*_{lK'J'}, \tag{15}$$

where A_{lKJ} is expressible as a combination of the reduced composite matrix elements (10) in the *jl*-coupling scheme, and the remaining notation is the same as in (8), (10). For *jl*-coupling we are unable to derive an analog of the simple relation (9) between $w_1{}^i$ and w^e, whereas Eq. (9), which holds for *LS*-coupling, implies that the electron and ion polarizations are proportional [cf. (11)].

As an example we give the expressions

$$P^i = \frac{9\rho_{3/2}{}^2 - 6\rho_{1/2}\rho_{3/2} - 3\rho_{1/2}{}^2}{5\rho_{1/2}{}^2 + 2\rho_{1/2}\rho_{3/2} + 11\rho_{3/2}{}^2}, \qquad P^e = \frac{5\rho_{1/2}{}^2 + 2\rho_{1/2}\rho_{3/2} - 7\rho_{3/2}{}^2}{5\rho_{1/2}{}^2 + 2\rho_{1/2}\rho_{3/2} + 11\rho_{3/2}{}^2}, \tag{16}$$

for two-photon ionization of metastable inert-gas atoms in the $n\,{}^3P_0$ states (the $ns'[1/2]_0$ states in the *jl*-coupling terminology).

Here

$$\rho_K \equiv \langle Ed'[{}^5/_2]_2|rg_{p'[K]_1}r|ns'[{}^1/_2]_0\rangle$$

is the radial composite matrix element for an ionization transition proceeding through states with intermediate angular momenta $K = l \pm j_0 = 1/2,\ 3/2$. Equation (16) clearly implies that $P^i = P^e = 0$ if there is no spin-orbit splitting of the intermediate states $n'p'[K]_1$, i.e., if $\delta \equiv E(n'p'[3/2]_1) - E(n'p'[1/2]_1) = 0$ (here we have neglected the fine structure of the $d'[K]_2$ final states in the continuous spectrum).

The ratio P^i/P^e in this case depends on the frequency ω of the ionizing radiation. For example, close to resonance $[\omega = \omega(n'p'[1/2]_1 - \varepsilon$, where $|\varepsilon| \ll \omega]$, we have

$$P^i = -3\delta\frac{4\varepsilon+\delta}{18\varepsilon^2+12\varepsilon\delta+5\delta^2}, \qquad P^e = \delta\frac{12\varepsilon+5\delta}{18\varepsilon^2+12\varepsilon\delta+5\delta^2},$$

for the polarizations as functions of the mismatch ε. Although P^i and P^e both have the same maximum and minimum values (equal to $+1$ and $-2/3$, respectively), these extrema are reached for different values of ε. Specifically, $P^i = 1$ for $\varepsilon = -(2/3)\delta$, whereas $P^e = 1$ for $\varepsilon = 0$; $P^i = -2/3$ for $\varepsilon = (1/6)\delta$, while $P^e = -2/3$ for $\varepsilon = -(5/6)\delta$.

In calculating the degree of polarization for ionization from the $np^6\,{}^1S_0$ ground state, we may neglect the splitting ΔK of the intermediate levels with respect to the quantum number K (this splitting is much smaller than the orbital moment splitting Δl of the valence electron). We then have the simple relation $P^i_{j_0=1/2} = P^e_{j_0=1/2}$ for ionization along the upper branch (corresponding to an ion residue with angular momentum $j_0 = 1/2$), and $P^i_{j_0=3/2} = -5/3 \cdot P^e_{j_0=3/2}$ for ionization along the lower branch ($J_0 = 3/2$). In particular, we have

$$P^e_{j_0=1/2} = \frac{(5\rho_{10}+\rho_{12})^2-36\rho_{32}^2}{(5\rho_{10}+\rho_{12})^2+54\rho_{32}^2} \tag{17}$$

for two-photon ionization, where $\rho_{ll_1} \equiv \langle El\,|rg_{l_1}r|np^6\ {}^1S_0\rangle$ is the radial composite matrix element for the second-order ionization transition. The polarization $P^e_{j_0=3/2}$ is given by a similar expression (multiplied by $-1/2$); in this case, however, the wave functions and energy correspond to the lower branch $j_0 = 3/2$. We see from (17) that

$$P^e_{j_0=1/2} \approx 1, \qquad \omega=\omega(n's'[{}^1/_2]_1),$$

$$P^e_{j_0=3/2} \approx -{}^1/_2, \qquad \omega=\omega(n's[{}^3/_2]_1),$$

$$P^e_{j_0=1/2} = (x^2-36)/(x^2+54), \qquad \omega=\omega(n'd'[{}^3/_2]_1),$$

where

$$x \equiv \langle Ep|r|n'd'[{}^3/_2]_1\rangle / \langle Ef|r|n'd'[{}^3/_2]_1\rangle$$

is the ratio of the radial matrix elements for transitions from the $n'd'[3/2]_1$ resonance state to continuum states in which the valence electron has the angular momenta $l = 1$ and $l = 3$.

We have

$$P^e_{j_0=1/2} = \frac{(35\rho_{210}+7\rho_{212}+3\rho_{232})^2-2025\rho_{432}^2}{(35\rho_{210}+7\rho_{212}+3\rho_{232})^2+2700\rho_{432}^2}.$$

for three-photon ionization from the ground state. Here $P^e_{j_0=1/2} \approx 1$ for the one- and two-photon resonances $\omega \approx \omega(n's'[1/2]_1)$ and $2\omega \approx \omega(n'p'[3/2]_2)$, respectively. Moreover,

$$P^e_{j_0=1/2} = [(7x_{21}+3x_{23})^2-2025]/[(7x_{21}+3x_{23})^2+2700],$$

$$\omega=\omega(n'd'[{}^3/_2]_1),$$

$$P^e_{j_0=1/2} = (x^2-225)/(x^2+300), \qquad 2\omega=\omega(n'f'[{}^5/_2]_2),$$

where

$$x_{ll_1} \equiv \frac{\langle El|rg_{l_1}r|n'd'[{}^3/_2]_1\rangle}{\langle Eg|rg_3r|n'd'[{}^3/_2]_1\rangle}, \qquad x \equiv \frac{\langle Ed|r|n'f'[{}^5/_2]_2\rangle}{\langle Eg|r|n'f'[{}^5/_2]_2\rangle}$$

are the ratios of the radial matrix elements for the two- and one-photon transitions, respectively, from the initial resonance to the final continuum states with $l = 2, 4$.

As N increases, the frequency dependence of $P^{i,e}$ becomes even more complicated. Near an m-photon resonance with the state $n'l'[K]_{j=m}$ we have $P^e \approx 1$ for $l' = m-1$ and $P^e \approx -N/(N+1)$ for $l' = m+1$. In the latter case P^e depends on the ratio of the matrix elements for $(N-m)$th order ionization transitions from the resonance states to unbound final states in which the orbital angular momentum of the electron $l = N \pm 1$ and the total atomic moment $J = N$. We can thus use P^e and P^i, which determine the magnetic moments of photoionized inert-gas atoms, to estimate the ratio of the transition matrix elements.

CONCLUSIONS

The above calculations show that ionization of atoms by circularly polarized radiation can polarize photoions as well as photoelectrons. The electron shell of the ion residue has a nonzero magnetic moment, and the polarizations P^e and P^i of the electrons and ions are uniquely related (this relation can often be expressed as a simple analytic formula). This correspondence can be traced to the fact that the spins of the ion residue and valence electron contribute equally to the total atomic moment and determine the magnitude of the spin-orbit splitting.

This polarization is quite general and is independent of how the angular momenta of the valence electron and atomic core are coupled. The calculations can thus be generalized to other types of coupling and to atoms with an arbitrary internal structure and electron excitation spectrum. Other topics of interest include the polarization of ions produced by ejection of an electron from an inner shell, and the polarization of multiply charged ions when several electrons are removed by photoionization.

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1) The spin polarization state (magnetization) of photoions can probably be detected quite easily by resonant magnetooptical techniques exploiting the Faraday or Hanle effects, resonant absorption, scattering and fluorescence of "magnetized" particles, etc., (see, e.g., Ref. 10).

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