Selective population of ionic states produced in photoionization by linearly polarized light

L. Végh*
Physics Divison, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831
and Physics Department, University of Tennessee, Knoxville, Tennessee 37966-1501

R. L. Becker†
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6373

(Received 9 March 1992)

The elements of the statistical tensors describing the anisotropy of an ionic state are calculated for photoionization induced by linearly polarized light. Depending on the orbital angular momentum of the ionized subshell, there are some special photoelectron emission angles that correspond to selective population of a magnetic sublevel. We point out that, if only one magnetic substate is populated, then the elements of the statistical tensor depend only on a geometric factor that is independent of the orbital angular momentum of the photoelectron.

PACS number(s): 32.80.Fb, 32.80.Hd

INTRODUCTION

Coincidence experiments which study an angular correlation frequently provide more dynamical information than does a noncoincident measurement. In coincidence measurements, performed by using polarized beams, the information may be very detailed and states of specific properties may be prepared.

We study the photoionization process followed by an Auger decay where both electrons are detected. The angular correlation between the photoelectron and the Auger electron reflects the alignment of the decaying ionic state produced in angle-resolved photoemission [1].

The alignment is determined by the population of vacancies in the production of the decaying state. Generally the decaying state with a vacancy in a subshell with total angular momentum j is described by the spin density matrix $\langle jm|\rho|jm'\rangle$. The possibilities of the production of pure states in charged-particle-induced reactions was discussed by Fano and Macek [2]. The selective population of the vacancy with $m_l=0$ in the shell with orbital angular momentum l can be realized if the incident particle is measured at zero deflection angle [2] or if the incident energy is close to the threshold of an excitation process [2,3]. In other cases we cannot produce a pure state because of the complexity of the vacancy-production mechanisms. In this paper we study the selective population of magnetic sublevels in photoionization induced by linearly polarized light and we discuss the behavior of the elements of the statistical tensor describing the alignment of the ionic state.

THEORETICAL FRAMEWORK

It is convenient to choose the axis of quantization z along the direction of linear polarization. In this geometry the photoionization calculated in dipole approximation has the selection rule

$$m_l=m_l' , \tag{1}$$

which means that the projections of the orbital momenta l and l' of the electron before and after the photoionization are identical. When the photoelectron is detected along z, taking into account the value of a spherical harmonic at zero polar angle as

$$Y_{lm}(0,\phi)=\delta_{m0}\left[\frac{2l+1}{4\pi}\right]^{1/2} , \tag{2}$$

the vacancy must have $m_l=0$ and therefore $|m_j|=\frac{1}{2}$.

The selective population of a magnetic substate, which is based on relation (1), can be regarded as an important feature of photoionization. Considering relation (1) and the properties of the spherical harmonics, the specific angles of the photoelectron emission, which are characterized by the selective population, may be identified. From Eq. (2) and the similar relation for the emission at 180°

$$Y_{lm}(\pi,\phi)=\delta_{m0}(-1)^l\left[\frac{2l+1}{4\pi}\right]^{1/2} , \tag{3}$$

it is apparent that at photoelectron ejection to 0° and 180°, independently of the angular orbital momentum of the ionized subshell, the vacancy has $m_l=0$ and therefore $|m_j|=\frac{1}{2}$.

A further special photoelectron emission angle, at least in the photoionization of p and d subshells, is 90°. For this angle, in photoionization of a p subshell, the vacancy has $m_l=0$ and therefore $|m_j|=\frac{1}{2}$. In fact, if the photoelectron is emitted as an s wave, according to relation (1) $m_l=0$. If the photoelectron is a d-wave elelctron, since

$$Y_{21}(90°,\phi)=Y_{2\text{-}1}(90°,\phi)=0$$

and

$$Y_{20}(90°,\phi)=-\tfrac{1}{4}\sqrt{5/\pi} , \tag{4}$$

only the $m_l=0$ vacancy is populated.

In the photoionization of a d subshell at the emission angle of 90°, the vacancy has $|m_l|=1$. In fact, the intensity of p-wave electrons emitted at 90° with $m_l=0$ is zero, while for $|m_l|=1$ the intensity is maximal,

$$Y_{10}(90°,\phi)=0, \quad Y_{11}(90°,\phi)=-\sqrt{3/8\pi}e^{i\phi} . \tag{5}$$

If an f-wave photoelectron is emitted then according to the relations

$$\begin{aligned} &Y_{30}(90°,\phi)=0 , \\ &Y_{32}(90°,\phi)=Y_{3\text{-}2}(90°,\phi)=0 , \end{aligned} \tag{6a}$$

and

$$Y_{31}(90°,\phi)=\tfrac{1}{8}\sqrt{21/\pi}e^{i\phi} , \tag{6b}$$

only the population of $|m_l|=1$ state is allowed.

The anisotropy of an ionic state produced by impact ionization of the subshell with the total angular momentum j can be described by a set of statistical tensors $\rho_{kq}(j)$ expressed in terms of the matrix elements of the spin-density matrix $\langle jm|\rho|jm'\rangle$ by the relation

$$\rho_{kq}=\sum_{m,m'}(-1)^{j-m'}(jmj-m'|kq)\langle jm|\rho|jm'\rangle . \tag{7}$$

In the decay of the ionic state, the angular distribution of decay products are completely determined by a set of tensors ρ_{kq}. This angular distribution can be given [4] as

$$W(\theta,\phi)=\frac{W_0}{4\pi}\left[1+\sum_k \alpha_k \sum_q A_{kq}(j)\left[\frac{4\pi}{2k+1}\right]^{1/2} \times Y_{kq}(\theta,\phi)\right] . \tag{8}$$

Here α_k depends on the decay process and A_{kq} are the statistical tensors

$$A_{kq}=\frac{\rho_{kq}(j)}{\rho_{00}(j)} \tag{9}$$

normalized so that $A_{00}=1$.

We calculate the statistical tensors ρ_{kq} for the case of ionization produced by linearly polarized light with emission of a photoelectron of momentum $\mathbf{k}_e$. Now the spin-density matrix elements are built up as

$$\langle jm|\rho(\mathbf{k}_e)|jm'\rangle=\sum_{m_s}F_{jm}(\mathbf{k}_e,m_s)F^*_{jm'}(\mathbf{k}_e,m_s) , \tag{10}$$

where $F_{jm}(\mathbf{k}_e,m_s)$ is the amplitude of the photoionization in dipole approximation

$$\begin{aligned} F_{jm}(\mathbf{k}_e,m_s)&=\langle \mathbf{k}_e,m_s|z|jm\rangle \\ &=\sum_{j',l',m_l'}(l'm_l'\tfrac{1}{2}m_s|j'm')Y_{l'm_l'}(\hat{\mathbf{k}}_e) \\ &\quad\times\langle(l'\tfrac{1}{2})j'm'|z|(l\tfrac{1}{2})jm\rangle . \end{aligned} \tag{11}$$

Here m_s is the spin projection of the photoelectron. The dipole matrix element $\langle(l'\frac{1}{2})j'm'|z|(l\frac{1}{2})jm\rangle$ can be expressed by the reduced matrix element $M_{\epsilon l',nl}$

$$\langle(l'\tfrac{1}{2})j'm'|z|(l\tfrac{1}{2})jm\rangle =\frac{1}{\hat{l}'}(jm10|j'm')U(l\tfrac{1}{2}1j';jl')M_{\epsilon l',nl} , \tag{12}$$

see, for example Ref. [5]. We assume that $M_{\epsilon l';nl}$ does not depend on the total angular momentum of the photoelectron. Here $\hat{a}=\sqrt{2a+1}$ and $U(abcd;ef)$ is the Jahn coefficient

$$U(abcd;ef)=(-1)^{a+b+c+d}\hat{e}\hat{f}\begin{Bmatrix} a & b & e \\ d & c & f \end{Bmatrix} .$$

Using Eqs. (7) and (10)–(12) the ρ_{00} and ρ_{kq} statistical tensors have the form

$$\rho_{00}=\frac{\hat{j}}{\hat{l}^4}\sum_{l',l'',m}(10l'm|lm)(10l''m|lm)Y_{l'm}(\hat{\mathbf{k}}_e) \times Y^*_{l''m}(\hat{\mathbf{k}}_e)M_{\epsilon l';nl}M^*_{\epsilon l'';nl} \tag{13}$$

and

$$\begin{aligned} \rho_{kq}=&(-1)^l\frac{j}{\hat{l}^3}U(l\tfrac{1}{2}kj;jl) \\ &\times\sum_{l',l'',m,m'}(-1)^{m'}(lml-m'|kq)(10l'm|lm) \\ &\times(10l''m'|lm')Y_{l'm}(\hat{\mathbf{k}}_e)Y^*_{l''m'}(\hat{\mathbf{k}}_e) \\ &\times M_{\epsilon l';nl}M^*_{\epsilon l'',nl} . \end{aligned} \tag{14}$$

Now we determine the elements of the statistical tensors A_{kq} for the case when only one magnetic substate with m_0 is allowed to be populated due to the special angle of the photoelectron ejection. Then the sums over the magnetic quantum numbers in Eqs. (13) and (14) are restricted to the sums of two terms depending on m_0 and $-m_0$. In this case $(-1)^{m'}(lml-m'kq)$ can be factored out of the sum in Eq. (14). Using Eqs. (9), (13) and (14) and the symmetry properties of spherical harmonics and Clebsh-Gordan coefficients A_{k0} with even k is given by

$$A_{k0}=(-1)^{l+m_0}\hat{l}(lm_0l-m_0|k0)U(l\tfrac{1}{2}kj;jl) \tag{15}$$

and A_{kq} with $q=2m_0$, $m_0\neq 0$ appear as

$$A_{kq}=\frac{(-1)^l}{2}e^{iq\phi_{\hat{k}_e}}\hat{l}(lm_0lm_0|kq)U(l\tfrac{1}{2}kj;jl) . \tag{16}$$

Summing over the magnetic quantum numbers in the expression (14), we obtain

$$\rho_{kq}(j,\mathbf{k}_e)=(-1)^{k+1}\frac{(2j+1)\begin{Bmatrix} l & \frac{1}{2} & j\\ j & k & l\end{Bmatrix}}{\sqrt{4\pi}}\sum_{l',l''}(-1)^{l'}\hat{l}'\hat{l}''M_{\epsilon l',nl}M^{*}_{\epsilon l'';nl}$$

$$\times\sum_{K}(l'0l''0|K0)\frac{1}{\hat{K}}Y_{Kq}(\hat{\mathbf{k}}_e)\sum_{x}(-1)^{x}(2x+1)\begin{Bmatrix} l & l & k\\ 1 & x & l'\end{Bmatrix}\begin{Bmatrix} l' & l'' & K\\ 1 & x & l\end{Bmatrix}$$

$$\times(10xq|Kq)(10xq|kq)\ . \qquad (17)$$

This formula, given for photons linearly polarized along the z axis, can be regard as a special case of the general formula presented by Berezhko and Kabachnik [6] in terms of the photon density matrix.

RESULTS

Equations (15) and (16) show that the nonzero elements of the A_{kq} tensor at special photoelectron ejection angles contain only geometric factors and they are independent of the orbital angular momentum of the photoelectron. The nonzero elements of the A_{kq} statistical tensors for $k>0$ for the p and d shells at photoelectron emission angles of 0° and 180° are

$$p_{3/2}:\ A_{20}=-1,\quad d_{3/2}:\ A_{20}=-1\ ,$$

$$d_{5/2}:\ A_{20}=-\frac{2\sqrt{2}}{\sqrt{7}}\ ,\quad A_{40}=(\tfrac{6}{7})^{1/2}\ ,$$

and at 90°

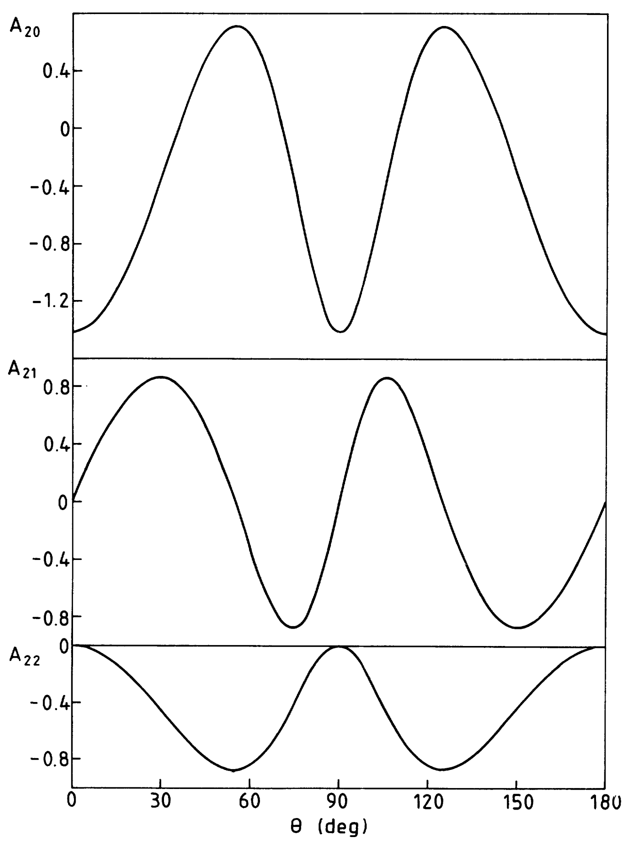

FIG. 1. The elements of tensor A_{kq} for photoionization of a p subshell induced by light linearly polarized along the z axis. The photoelectron is ejected in the xz plane with angle θ. The curves have been calculated by using Eqs. (9) and (17). To obtain the values valid for a subshell with $j=\frac{3}{2}$ the functions should be multiplied by $1/\sqrt{2}$.

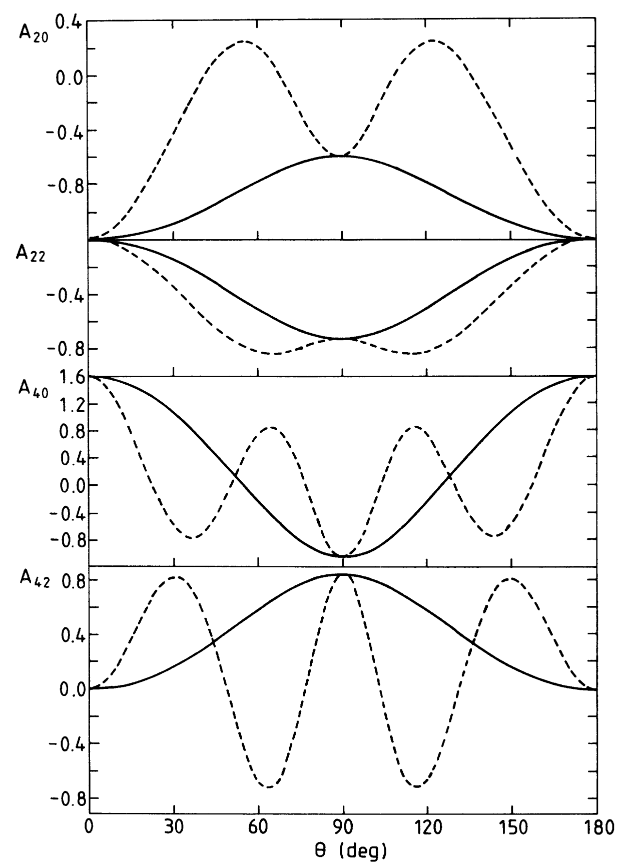

FIG. 2. The same as Fig. 1, but for photoionization of a d subshell. The elements of tensors A_{21}, A_{41} are not presented here. The multiplicative factors are the following: for the $j=\frac{3}{2}$ subshell, $(\frac{7}{10})^{12}$, and for the $j=\frac{5}{2}$ subshell for A_{2q} and A_{4q}, $2/\sqrt{5}$ and $1/\sqrt{3}$, respectively. The solid lines correspond to p-wave photoelectron emission, the dashed lines denote the curves for f-wave photoemission.

$$p_{3/2}:\quad A_{20}=-1\ ,$$

$$d_{3/2}:\quad A_{20}=-\tfrac{1}{2},\quad A_{22}=A^{*}_{2\text{-}2}=-\frac{\sqrt{3}}{2\sqrt{2}}e^{i2\phi_{\hat{\mathbf{k}}_e}}\ ,$$

$$d_{5/2}:\quad A_{20}=-(\tfrac{2}{7})^{1/2},\quad A_{22}=A^{*}_{2\text{-}2}=-(\tfrac{3}{7})^{1/2}e^{i2\phi_{\hat{\mathbf{k}}_e}}\ ,$$

$$A_{40}=-\frac{2\sqrt{2}}{\sqrt{3}\sqrt{7}},\quad A_{42}=A^{*}_{4\text{-}2}=\frac{\sqrt{5}}{\sqrt{3}\sqrt{7}}e^{i2\phi_{\hat{\mathbf{k}}_e}}\ .$$

A deviation of the measured and theoretical values at the special photoelectron ejection angles would imply a violation of the two-step concept of dipole photoionization followed by Auger decay.

Using Eqs. (9) and (17) we have calculated the elements of the A_{kq} tensor for p- and d-shell ionization as a function of the polar angle of the photoelectron ejection at zero azimuthal angle. For the sake of simplicity we assume that the photoionization is dominated by one of the two allowed angular orbital momenta of the photoelectron. Since $\rho_{kq}(j,\mathbf{k}_e)$ now depend on the total angular momentum j only through a $6j$ symbol, which is a simple multiplicative factor in Eq. (17), we present the A_{kq} functions in the figures without the

$$\frac{\begin{Bmatrix} l & \frac{1}{2} & j \\ j & k & l \end{Bmatrix}}{\begin{Bmatrix} l & \frac{1}{2} & j \\ j & 0 & l \end{Bmatrix}}=U(l\tfrac{1}{2}kj;jl)$$

factor. As Figs. 1 and 2 show, at the selective angles discussed above, A_{kq} does not depend on the orbital angular momentum of the photoelectron and its values are the same as given by expressions (15) and (16).

ACKNOWLEDGMENTS

This work was supported by the Chemical Sciences Division of the Office of Basic Energy Sciences, U.S. Department of Energy under Contract No. DE-AC05-84OR21400 with the Martin Marietta Energy Systems, Inc., and by the Hungarian OTKA Foundation under Contract No. 3011. We thank M. O. Krause and C. D. Caldwell for the inspiring discussions. L.V. thanks N. M. Kabachnik for his valuable remarks on the manuscript.

*Present address: Institute of Nuclear Research of the Hungarian Academy of Sciences, P.O. Box 51, H-4001 Debrecen, Hungary.

†Retired.

[1] B. Kämmerling and V. Schmidt, Phys. Rev. Lett. **67**, 1848 (1991); and M. O. Krause and C. D. Caldwell (private communication); C. D. Caldwell, M. O. Krause, S. B. Whitfield, M. G. Flemming, and A. Fahlman, in Abstracts of the 10th International Conference on Vacuum Ultraviolet Radiation Physics, July 1992, Paris.

[2] U. Fano and J. H. Macek, Rev. Mod. Phys. **45**, 553 (1973).

[3] W. Sandner and W. Schmitt, J. Phys. B **11**, 1833 (1978).

[4] E. G. Berezhko, N. M. Kabachnik, and V. V. Sizov, J. Phys. B **11**, 1819 (1978).

[5] D. A. Varshaliovich, A. N. Moskalev, and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).

[6] E. G. Berezhko and N. M. Kabachnik, J. Phys. B **12**, 2993 (1979).