Angular Distribution of Auger Electrons Following Photoionization

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It is shown that the photoionization process of inner-shell electrons by unpolarized radiation leads to an alignment of the ionized atoms. From the nonisotropic angular distribution of the following Auger electrons the relative partial photoionization cross sections can be determined.

It has been predicted by theory[1] and confirmed by experiment[2] that Auger electrons following an inner-shell vacancy, caused by a directed unpolarized electron or proton beam, may have a nonisotropic angular distribution if an electron with quantum number $j > \frac{1}{2}$ has been removed and if the resulting atomic state has a quantum number $J'' > \frac{1}{2}$. This is due to the different ionization probabilities $Q(J''|M''|)$ of magnetic sublevels M'' of the atomic state J'' resulting in an alignment of the ionized atoms relative to the axis of the primary particle beam defining the quantization axis. The angular distribution depends also on the final atomic state of the Auger transition.

There arises the question whether Auger electrons following an inner-shell vacancy produced by photoionization may also have a noniostropic distribution. The answer will be determined essentially by the relative population $Q(J''M'')$ of magnetic sublevels M'' of the excited state with quantum numbers $S''L''J''M''$ after photoionization. Cooper and Zare[3] have pointed out that the population $Q(S''L''M_S''M_L'')$ will be independent of M_S'' and M_L'', so that $Q(J''M'')$ also will be independent of M''. Therefore, a subsequent radiation is expected to be isotropic and unpolarized, and subsequent Auger electrons will show an isotropic angular distribution. We cannot confirm this result and will show that the relative population $Q(J''M'')$ of atomic substates after photoionization by unpolarized radiation depends on $|M''|$. This yields a nonisotropic angular distribution of subsequent Auger electrons which will be discussed.

The appropriate coupling scheme describing an atomic state after the ionization of an inner-shell electron $n'l'j'$ is the $SLJM$ scheme because the fine-structure splitting is much larger than the width of the fs levels. This holds also for the final atomic state after the Auger transition. Therefore, the atomic initial state will be characterized by $SLJM$, the atomic state after photoionization by $S''L''J''M''$, and the atomic state after the Auger process by $S^fL^fJ^fM^f$. In order to simplify the theoretical treatment, randomly oriented initial atoms and unpolarized incoming radiation may be assumed. In this case it can be shown[4-6] that the summation over the magnetic quantum number M'' may be performed incoherently provided the axis of quantization (z axis) has been chosen to coincide with the direction of the incoming radiation. This results in a form of the angular correlation which can be described by a two-stage process: The relative population $Q(J''M'')$ of the M'' sublevels of the atom after photoionization can be multiplied with the probability $P(J''M'' \to J^fM^fm_s;\theta)\,d\Omega$ of emission of an Auger electron with spin projection m_s (relative to the z axis) in a solid angle $d\Omega$ in direction θ with respect to the incoming radiation. The angular distribution of Auger electrons is then given by

$$W(\theta) = (2J''+1)^{-1} \sum_{M''M^f} \sum_{m_s} Q(J''M'')P(J''M'' \to J^fM^fm_s;\theta). \tag{1}$$

The relative populations $Q(J''M'')$ of the atomic state after photoionization by unpolarized photons are equal to the photoionization cross sections $Q(J \to J''M'')$ which can be calculated using the expressions given by Cooper and Zare.[3] We need the transition matrix elements for photoionization (electric-dipole approximation) between $SLJM$ coupled wave functions for the two basic states of circular polarized photons and with quantization axis defined by the direction of the incoming radiation. These matrix elements follow from formula (III.21) given by Cooper and Zare with the modification of $\mu = +1$ instead of $\mu = 0$ in the tensor operator $C_\mu{}^1$ corresponding to the two circular states in the coordinate sys-

tem defined above. Using the notation of Cooper and Zare (with corrected phase factor), one gets

$$\langle \Psi_i | \sum_j r_j C_\mu^{\,1} | \Psi_f \rangle = \pm \sum_{lm} \sum_{M_L'' M_S''} \sum_{M_L M_S} \sum_{M'} a(l,m) l_>^{1/2} n^{1/2} (cfp)\, \sigma(n'l' \to \epsilon l) ([L][S][J''][J])^{1/2}$$
$$\times (-1)^{s-M_S+g+1+l'+l-m+M'+S-L-M-M''+M_L''+\mu} \begin{pmatrix} L'' & S'' & J'' \\ M_L'' & M_S'' & -M'' \end{pmatrix} \begin{pmatrix} L & S & J \\ M_L & M_S & -M \end{pmatrix}$$
$$\times \begin{pmatrix} S'' & s & S \\ M_S'' & m_s & -M_S \end{pmatrix} \begin{pmatrix} L & L'' & l' \\ -M_L & M_L'' & -M' \end{pmatrix} \begin{pmatrix} l & 1 & l' \\ m & \mu & M' \end{pmatrix}, \qquad (2)$$

where $\sigma(n'l' \to \epsilon l)$ is the radial dipole integral $\int_0^\infty r R_{n'l'} R_{\epsilon l}\, dr$ of the transition $n'l' \to \epsilon l$, n is the number of equivalent $n'l'$ electrons, (cfp) is the appropriate coefficient of fractional parentage, $l_> = \max(l, l')$, and $[K] = 2K+1$. The relative populations $Q(J''M'')$ are given by the incoherent addition of the two circular polarized states ($\mu = \pm 1$) with half the total intensity. For the special case of a $^1S_0$ initial atomic state, which is important for experimental investigations, this procedure yields

$$Q(J''M'') = Q(^1S_0 \to J''M'') = 2\pi \frac{[J'']}{[l']} \delta_{S'',1/2}\, \delta_{L'',l'} \sum_{lm} \sum_{M_L'' M_S''} n(cfp)^2 \sigma^2(n'p' \to \epsilon l) l_>$$
$$\times \begin{pmatrix} L'' & S'' & J'' \\ M_L'' & M_S'' & -M'' \end{pmatrix}^2 \frac{1}{2} \left[\begin{pmatrix} l & 1 & l' \\ m & 1 & M_L'' \end{pmatrix}^2 + \begin{pmatrix} l & 1 & l' \\ m & -1 & M_L'' \end{pmatrix}^2 \right]. \qquad (3)$$

From Eq. (3) it can be seen that $Q(J''M'')$ depends only on $|M''|$ and has different values for different $|M''|$ only in the case $l' \geq 1$. In the special case where an $n'p$ electron of a filled subshell is photoionized, one gets

$$Q(\tfrac{3}{2}|\tfrac{3}{2}|) = \tfrac{6}{27}\pi [3\sigma^2(n'p \to \epsilon s) + \tfrac{21}{5}\sigma^2(n'p \to \epsilon d)], \quad Q(\tfrac{3}{2}|\tfrac{1}{2}|) = \tfrac{6}{27}\pi [\sigma^2(n'p \to \epsilon s) + \tfrac{19}{5}\sigma^2(n'p \to \epsilon d)]. \qquad (4)$$

This result shows the pronounced alignment of the atomic state after photoionization. Using theoretical values of $\sigma(n'p \to \epsilon s)$ and $\sigma(n'p \to \epsilon d)$, calculated by McGuire,[7] the relative populations have been evaluated as a function of photon energy. Results for magnesium ($n'l' = 2p$) and calcium ($n'l' = 3p$) are shown in Fig. 1.

Experimentally, the alignment of atoms ionized in an inner shell $n'l'$ can be measured either through the angular distribution of the subsequent Auger electrons, or through the partial polarization of the competing characteristic x radiation. Because we believe that the measurement of the angular distribution of Auger electrons is easier to perform, we discuss this case only. The calculation of the angular correlation $W(\theta)$ of the Auger electrons [Eq. (1)] is quite identical with the procedure of calculating the angular distribution of Auger electrons following an inner-shell ionization caused by a directed unpolarized electron or proton beam.[8] If the final state has $J^f = 0$, then only one partial wave of the Auger electron is emitted, and the angular distribution of the Auger electron depends only on the relative populations $Q(J''|M''|)$ and takes the simple form

$$W(\theta) = 1 + A P_2(\cos\theta), \qquad (5)$$

where $P_2(\cos\theta)$ is a Legendre polynomial and

$$A = \frac{Q(\tfrac{3}{2}|\tfrac{1}{2}|) - Q(\tfrac{3}{2}|\tfrac{3}{2}|)}{Q(\tfrac{3}{2}|\tfrac{1}{2}|) + Q(\tfrac{3}{2}|\tfrac{3}{2}|)}, \qquad (6)$$

with the $Q(J''|M''|)$ being those of Eq. (4) assuming the atom is in a $^1S_0$ state before photoionization occurs. In Fig. 2 the asymmetry coefficient A of the angular distribution of Auger electrons following photoionization in magnesium ($n'l' = 2p$)

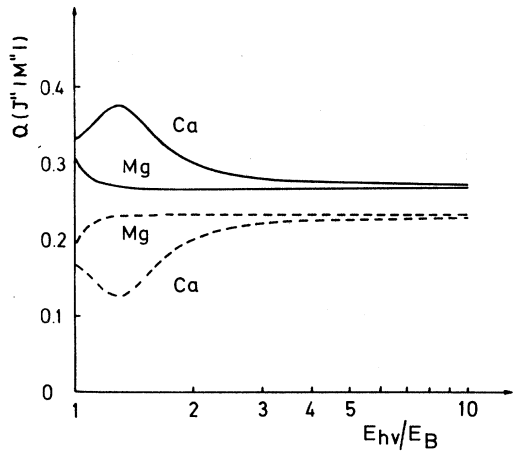

FIG. 1. Relative populations $Q(\frac{3}{2}|\frac{3}{2}|)$ (solid line) and $Q(\frac{3}{2}|\frac{1}{2}|)$ (dashed line) of the atomic state after photoionization in magnesium ($n'l' = 2p$) and calcium ($n'l' = 3p$) as a function of photon energy $E_{h\nu}$ in units of the binding energy E_B of the $n'l'$ electron. The results have been normalized to 1.0 for the total population.

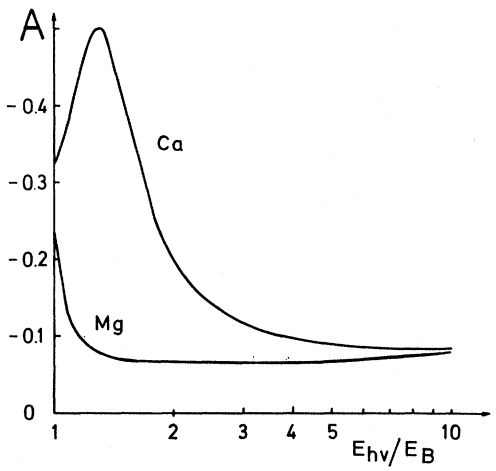

FIG. 2. Asymmetry coefficient A of the angular correlation function of Auger electrons following photoionization in magnesium ($n'l' = 2p$) and calcium ($n'l' = 3p$) as a function of photon energy $E_{h\nu}$ in units of the binding energy E_B of the $n'l'$ electron.

and calcium ($n'l' = 3p$) is plotted as function of the energy of the incoming unpolarized radiation. We have selected these two examples to demonstrate the striking differences in the angular distribution of Auger electrons of different elements.

Generally[5] it can be shown that in the dipole approximation of the photoionization process, the angular distribution of Auger electrons following photoionization by unpolarized radiation has the form

$$W(\theta) = 1 + AP_1(\cos\theta), \tag{7}$$

where the coefficient A is a function of two partial photoionization cross sections. So it should be possible to determine the ratio of these two partial cross sections by a measurement of the angular distribution of Auger electrons following photoionization. Experimental investigations are in progress.

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[1]W. Mehlhorn, Phys. Lett. 26A, 166 (1968).
[2]B. Cleff and W. Mehlhorn, Phys. Lett. 37A, 3 (1971).
[3]J. Cooper and R. N. Zare, in *Lectures in Theoretical Physics: Atomic Collision Processes*, edited by S. Geltman, K. T. Mahanthappa, and W. E. Brittin (Gordon and Breach, New York, 1969), Vol. XI C, p. 317.
[4]S. Devons and L. J. B. Goldfarb, in *Encyclopedia of Physics*, edited by S. Flügge (Springer, Berlin, 1957), Vol. XLII, p. 362.
[5]C. N. Yang, Phys. Rev. 74, 764 (1948).
[6]M. Frauenfelder and R. M. Steffen, in *Alpha, Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1966), p. 997.
[7]E. J. McGuire, Sandia Laboratories Research Report No. SC-RR-70-721, 1970 (unpublished).
[8]W. Mehlhorn and B. Cleff, to be published.

Apparent Pressure Dependence of the Reduced Mobility of Potassium Ions in Gases*

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The apparent pressure dependence of the reduced mobility of potassium ions in various gases, reported by Elford, is explained in terms of diffusion effects and reversible reactions involving clustered ions.

In a recent paper[1] Elford has reported measurements of the mobility of potassium ions in helium, neon, argon, hydrogen, and nitrogen. All the measurements evidence a variation of the reduced mobility with pressure for constant drift energy (i.e., E/N, where E is the drift field and N the neutral gas number density[2]). This implies either three-body elastic collisions or the production of a long-lived ion-neutral cluster. At the pressures used in Elford's drift tube (1–20 Torr typically), the gas density is so low that the former possibility may be ruled out (since the average molecular separation is about 1000 Å). On the other hand Elford claims that his observations are not consistent with clustering since his data would imply mobilities of the ion and the ion cluster which seem too similar (3% difference, assuming equilibrium between the ions and the clusters). One is then left with a contradiction between experiment and the kinetic-theory transport equation.[3] Since this raises questions of some consequence it seems appropriate to review