Production of Polarized Ions by Photoionisation

W. Bussert
Fachbereich Physik der Universität, Kaiserslautern, Federal Republic of Germany

H. Klar
Fakultät für Physik der Universität Freiburg und Sonderforschungsbereich 91 der Universität Kaiserslautern, Federal Republic of Germany

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The photoproduction of polarized ions is investigated theoretically. The alignment of photoions is expressed in terms of a possible initial atomic polarization, the polarization of the radiation and of reduced transition matrix elements. In particular it is shown that an ion alignment can result from unpolarized target atoms exposed to unpolarized light.

1. Introduction

During the last decades a great number of photoionisation experiments has been performed. Such experiments include measurements of the angular distribution of photoelectrons and their spin polarization, [1, 2] and references therein. More recently also photoelectrons from polarized atoms have been considered experimentally [3] and theoretically [4, 5]. The goal of all these investigations was mainly to study the dynamics in many-electron systems.

The present paper investigates the polarization of photoions from polarized as well as from unpolarized atoms. In particular we show that polarized ions may be obtained from unpolarized atoms exposed to unpolarized light. Photoionisation therefore constitutes an important method to produce polarized ions which may for instance be used as collision partners in further experiments (ion atom collisions). On the other hand the ion polarization involves dynamical parameters different from the angular distribution parameter and the three spin polarization parameters. A measurement of the ion polarization would therefore yield additional information about the photoionisation process. However, a direct measurement of an ion polarization is very difficult except in the case where fluorescence radiation from the excited ion is detected [6–8].

The paper is organized as follows: The general theoretical framework is developed in Sect. 2. Section 3 treats the case of unpolarized initial atoms. For simplicity one-electron atoms are considered separately in Sect. 4, and effects from the nuclear spin, finally, are discussed in Sect. 5. Atomic units are used throughout.

2. Theoretical Frame

In this section we disregard the nuclear spin and consider the ionisation of an atom by radiation in the electric dipole approximation,

\[ \text{atom} (c_0, J_0) + h\nu \rightarrow \text{ion} (c, J) + \text{electron} (p, m_1). \]

The initial atomic state labelled by the total angular momentum \( J_0 \) and by other quantum numbers \( c_0 \) may be polarized, i.e. its magnetic substates may be unequally populated. This polarization we describe by a density operator \( \rho_0 \), normalized to unity. The unnormalized density operator of the final state is then given by

\[ \rho = (\delta D) \rho_0 (\delta D)^\dagger. \] (1)

where \( D \) is the dipole operator and \( \delta \) is the polarization vector.

In the following we are interested only in the observation of ions. The cross section for photoproduction of ions in magnetic substates \( M \) with respect to
some direction $\hat{Q}$ is given by

$$I_M(\hat{Q}, \hat{Q}_0) = 4 \pi^2 x a_0^2 \omega \sum_{m_s} \int d\phi \langle cJ M p m_s | \rho | c J M p m_s \rangle.$$  
\(2\)

The ion states $|cJ M \rangle$ are eigenstates of the operators $J^2$ and $(\hat{Q} \cdot J)$ with eigenvalues $J(J+1)$ and $M$. $p$ and $m_s$ are the momentum and the spin projection of the not observed photoelectron. $x$ is the fine-structure constant, $a_0$ is the first Bohr radius and $\omega$ is the frequency of the light. The density operator of the initial state will be diagonal with respect to a certain direction $\Omega_0$. As usual we treat both linearly as well as circularly polarized incident radiation on an equal basis by introducing suitable coordinate systems.

In the case of linearly polarized light we choose a quantization axis parallel to the electric light vector $E$. For circularly polarized radiation we introduce the linear momentum of the radiation as quantization axis. Unpolarized radiation, finally, is conveniently treated as an incoherent superposition of right and left circularly polarized radiation.

In order to analyse the general expression (2) we expand the cross section into a series of state multipoles [9–11]

$$I_M(\hat{Q}, \hat{Q}_0) = \sum_K (-)^{K-J-M} \langle J-M | J M | K 0 \rangle \rho_{K0}(\hat{Q}, \hat{Q}_0).$$  
\(3\)

Combining (2) and (3) we easily get

$$\rho_{K0}(\hat{Q}, \hat{Q}_0) = 4 \pi^2 x a_0^2 \omega \sum_M (-)^{K-J-M} \langle J-M | J M | K 0 \rangle \cdot \sum_{m_s} \int d\phi \langle cJ M p m_s | \rho | c J M p m_s \rangle.$$  
\(4\)

Often it is also convenient to consider the alignment factor

$$A_{K0}(\hat{Q}, \hat{Q}_0) = \frac{\rho_{K0}(\hat{Q}, \hat{Q}_0)}{\rho_{00}(\hat{Q}, \hat{Q}_0)}.$$  
\(5\)

We come now to the evaluation of the r.h.s. of (4). In the coordinate frame introduced above the density operator of the initial atom is generally not diagonal, but a diagonalisation may be performed by a rotation $R(\hat{Q}_0)$

$$\langle J_0 M_0 | \rho_A | J_0 M_0 \rangle = \sum_{N_0} \langle J_0 M_0 | R(\hat{Q}_0) | J_0 N_0 \rangle \langle J_0 N_0 | \rho_A | J_0 N_0 \rangle \cdot \langle J_0 N_0 | R(\hat{Q}_0)^\dagger | J_0 M_0 \rangle.$$  
\(6\)

Using standard relations for rotation matrices, and expanding the diagonal density matrix elements into a series of state multipoles analogous to (3), (6) may be rewritten in the form

$$\langle J_0 M_0 | \rho_A | J_0 M_0 \rangle = \sum_{K_0} \left(-\right)^{K_0-J_0-M_0} \left(\frac{4 \pi}{2 K_0+1}\right)^{1/2} \cdot \langle J_0-M_0 J_0 M_0 | K_0 M_0-M_0 \rangle \cdot \rho_{K0}(\hat{Q}_0) \cdot Y_{K0-M_0-M_0}(\hat{Q}_0).$$  
\(7\)

The dipole transition matrix elements we expand now into partial waves and apply the Wigner-Eckart theorem,

$$\langle cJ M p m_s D_q | c_0 J_0 M_0 \rangle = \sum_{i=m_j m_f} i^l Y_{lm}(\hat{Q}).$$  
\(8\)

Here $J_f$ is the total (conserved) angular momentum, $M_f$ its $z$-component, and $q$ describes the polarization state of the light.

We substitute the expressions given by (7) and (8) into (4) and perform the summation over all magnetic quantum numbers [12]. The result may be cast into the following form

$$\rho_{K0}(\hat{Q}, \hat{Q}_0) = 16 \pi^2 x a_0^2 \omega \sum_{K \in \mathbb{X} |J_0 J_f| J_f} \left(-\right)^{J_f+J_i+1+q} \langle 2J_f+1 \rangle \langle 1-q 1 q | X 0 \rangle \left\{ \begin{array}{c} J_f \ 1 \\ J_i \ 1 \end{array} \right\} \left\{ \begin{array}{c} K_0 \ K \ X \\ J_0 \ J_i \ 1 \end{array} \right\} \rho_{K0}(J_0) \cdot g_{X K_0}(\hat{Q}, \hat{Q}_0)$$  
\(9\)

where the bipolar spherical harmonics are defined by

$$g_{X K_0}(\hat{Q}, \hat{Q}_0) = \sum_{K0 \in \mathbb{X} \backslash Q} \langle K Q K_0 - Q | X 0 \rangle Y_{K0}(\hat{Q}) Y_{K0-Q}(\hat{Q}).$$  
\(10\)

Equation (9) represents the connection between the degree of initial atomic polarization ($\rho_{K0}$) and the final ion polarization. Inspection of the $9-j$ symbol in (9) shows that the ion multipoles $K$ are limited by

$$0 \leq K \leq 2J$$
$$0 \leq K \leq 2J_i$$

$$|K_0 - X| \leq K \leq K_0 + X \quad \text{with} \quad X = 0, 1, 2.$$

In particular we note that in general an ion alignment ($K > 0$) also occurs if the initial atom was unpolarized ($K_0 = 0$). We investigate this important case in the next section.