PHOTOEXCITATION OF POLARIZED ATOMS BY POLARIZED RADIATION

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An expression for the probability of the excitation of polarized atoms by polarized radiation is obtained by using the graphical technique of angular momentum. In the case of pure states, the applied method is an alternative to that of density matrix formalism. The obtained expression enables one to describe the polarization state of an excited atom and can be used to derive formulas for the special cases under specific experimental conditions as well as for the photoexcitation as the first step process. The application of obtained expressions for the description of multistep processes is discussed. The photoexcitation of unpolarized atoms by polarized radiation and polarized atoms by unpolarized radiation is considered as the examples of practical application of the obtained general expression.

Keywords: excited atoms, polarization, photoexcitation

PACS: 31.50.Df, 29.25.Pj

1. Introduction

The laser [1] and tunable synchrotron radiation allows one not only to ionize an electron of a specific outer or inner shell of an atom but also to excite it to a specific orbital [2, 3]. The state of the produced atom is polarized if the radiation is polarized. Thus, the excitation of atoms by polarized radiation is one of the ways to produce atoms in polarized states for further measurements [4, 5]. In the case of inner-shell ionization, the ions can emit the fluorescence radiation [6, 7] that provides information about the polarization state of an atom before the ionization. The decay of atomic states via the emission of a photon is the only way when electrons are excited from outer shell. The emission of an electron is another way of decay of an excited atomic state in the case of the inner-shell ionization [8]. Thus, the photoexcitation can be used as a first step to create a resonant state with well-defined angular momentum and parity for further investigations of polarization and angular correlation phenomena [8, 9].

The main aim of the present work was the derivation of a general expression for the probability of the excitation of polarized atoms with polarized radiation and the application of this expression for specific cases to describe the polarization used in measurements. The application of the general formula for description of the photoexcitation as the first step process is also considered. For derivation of the expression for excitation probability, the graphical technique of angular momentum [10] is applied. So far, the investigations of the photoexcitation of polarized atoms was performed only by using the methods of density matrix [11, 12], therefore, our treatment is an alternative to the density matrix formalism. Some improvements of the graphical technique and methods of the ordinary atomic theory suggested in [13–15] have also found an application in the present work.

2. Derivation of the photoexcitation probability

To obtain the general expression for the excitation of polarized atoms by a polarized radiation the following process was studied:

$$A(\alpha_0 J_0 M_0) + h\nu(\widehat{\epsilon}_\lambda \mathbf{k}_0) \rightarrow A^*(\alpha_1 J_1 M_1). \quad (1)$$

Here an atom A in the state $\alpha_0 J_0 M_0$ is excited by the electromagnetic radiation into the state $\alpha_1 J_1 M_1$, where α defines the configuration and other quantum numbers, J is total angular momentum, and M is its projection. The electromagnetic radiation is described by the wave vector $\mathbf{k}_0$ and unit vector $\widehat{\epsilon}_\lambda$ of the polarization (λ is the helicity, $\lambda = \pm 1$). The system of atomic units is used in the present work ($\hbar = e = m = 1$,

$c = 137$, unless these constants are displayed explicitly). It is assumed that the fine structure splitting $\gg$ line width $\gg$ hyperfine structure splitting. Then the states of an atom can be characterized by the total angular momentum of all electronic shells. The modifications enabling the calculations of probability in a case when hyperfine structure is important will be described below. In the present work, it is assumed that the directions for the measurement of the projections M_0 and M_1 may be different.

The differential cross-section of the process (1) can be written as follows [12]:

$$\frac{d\sigma(\alpha_0 J_0 M_0 \widehat{\epsilon}_\lambda \mathbf{k}_0 \to \alpha_1 J_1 M_1)}{d\Omega}$$

$$= 2\pi^2 \left[\int \langle \alpha_1 J_1 M_1 | \mathbf{J}(\mathbf{r}) | \alpha_0 J_0 M_0 \rangle \mathbf{A}_{\lambda \mathbf{k}_0}(\mathbf{r}) \, d\mathbf{r} \right]$$

$$\times \left[\int \langle \alpha_1 J_1 M_1 | \mathbf{J}(\mathbf{r}) | \alpha_0 J_0 M_0 \rangle \mathbf{A}_{\lambda \mathbf{k}_0}(\mathbf{r}) \, d\mathbf{r} \right]^*. \tag{2}$$

Here $\mathbf{J}(\mathbf{r})$ stands for the operator of the current of electrons and $\mathbf{A}_{\lambda \mathbf{k}_0}(\mathbf{r})$ is the operator of the vector potential of electromagnetic field. Taking into account that $kr \gg 1$, assuming an arbitrary direction of the incidence $\mathbf{k}_0$ of the photon, and inserting the multipole expansion for the $\mathbf{A}_{\lambda \mathbf{k}_0}(\mathbf{r})$, the expression in the right square brackets of (2) acquires the form [12]

$$\int \langle \alpha_1 J_1 M_1 | \mathbf{J}(\mathbf{r}) | \alpha_0 J_0 M_0 \rangle \mathbf{A}_{\lambda k_0}(\mathbf{r}) \, d\mathbf{r}$$

$$= \sum_{p=0,1} \sum_{k=1}^{\infty} \sum_{q=-k}^{k} i^k (-i\lambda)^p \left[\frac{k+1}{k} \right]^{1/2} \frac{k_0^{k-1/2}}{(2k-1)!!}$$

$$\times D_{q\lambda}^k(\widehat{k}_0) \langle \alpha_1 J_1 M_1 | \mathcal{Q}_{kq}^p | \alpha_0 J_0 M_0 \rangle$$

$$= \sum_{k=1}^{\infty} \sum_{q=-k}^{k} \langle \alpha_1 J_1 M_1 | Q_q^{(k)} | \alpha_0 J_0 M_0 \rangle D_{q\lambda}^k(\widehat{k}_0), \tag{3}$$

$$\langle \alpha_1 J_1 M_1 | Q_q^{(k)} | \alpha_0 J_0 M_0 \rangle$$

$$= k_0^{k-1/2} \sum_{p=0,1} \left[\frac{k+1}{k} \right]^{1/2} \frac{i^k (-i\lambda)^p}{(2k-1)!!}$$

$$\times \langle \alpha_1 J_1 M_1 | \mathcal{Q}_{kq}^p | \alpha_0 J_0 M_0 \rangle. \tag{4}$$

Here $D_{q\lambda}^k(\widehat{k}_0)$ is the Wigner rotation matrix [16], and $p = 0$ indicates the operator of electric multipole transition (Ek)

$$\langle \alpha_1 J_1 M_1 | \mathcal{Q}_{kq}^0 | \alpha_0 J_0 M_0 \rangle$$

$$= \left[\frac{4\pi}{2k+1} \right]^{1/2} \sum_j \langle \alpha_1 J_1 M_1 | r_j^k Y_{kq}(\widehat{r}) | \alpha_0 J_0 M_0 \rangle, \tag{5}$$

and $p = 1$ is for the magnetic multipole transition (Mk)

$$\langle \alpha_1 J_1 M_1 | \mathcal{Q}_{kq}^1 | \alpha_0 J_0 M_0 \rangle$$

$$= \left[\frac{4\pi}{2k+1} \right]^{1/2} \sum_j \langle \alpha_1 J_1 M_1 | \nabla r_j^k Y_{kq}(\widehat{r})$$

$$\times \left[\frac{l_j}{k+1} + s_j \right] | \alpha_0 J_0 M_0 \rangle. \tag{6}$$

Note that the parities of the magnetic and electric multipole fields are $(-1)^k$ and $(-1)^{k+1}$, respectively. Only the magnetic (Mk) and electric (Ek) part contributes to transitions between specific electronic states owing to parity selection rules. Since we are considering pure photon states, there is no need to introduce Stokes parameters explicitly. In electrical dipole approximation, the matrix element (4) is as follows:

$$\langle \alpha_1 J_1 M_1 | Q_q^1 | \alpha_0 J_0 M_0 \rangle$$

$$= \sqrt{2k_0} \langle \alpha_1 J_1 M_1 | \mathcal{Q}_{1q}^0 | \alpha_0 J_0 M_0 \rangle. \tag{7}$$

The helicity $\lambda = \pm 1$ describes the right-hand and left-hand circular radiation. In the case of any polarization ϵ, the polarization may be expressed via the circular polarizations. Then

$$\mathbf{A}_{\epsilon \mathbf{k}_0}(\mathbf{r}) = \alpha \mathbf{A}_{\lambda=+1 \mathbf{k}_0}(\mathbf{r}) + \beta \mathbf{A}_{\lambda=-1 \mathbf{k}_0}(\mathbf{r}) \tag{8}$$

has to be inserted into Eq. (3) to obtain the expression for the excitation cross-section (2).

Since there is no dependence on the direction of $\mathbf{k}_0$ in the electrical dipole approximation, the calculated angular distributions of the emitted radiation are symmetric with respect to $\mathbf{k}_0$ reversal. Retaining terms up to the second order, the calculation then includes electric dipole – electric quadrupole (E1–E2) and electric dipole – magnetic dipole (E1–M1) cross terms, i.e. interference between the E1 photoionization amplitudes and those for E2 and M1 interactions, but excludes E1–M2, M1–M1, and E2–M1

cross terms that are of the order $(k_0 r)^2$. The E1–E2 and E1–M1 cross terms have odd parity and produce calculated asymmetry in angular distributions of radiation with respect to the reversal of the direction of $\mathbf{k}_0$ [17].

Sometimes it is more convenient to analyse the polarization state of a particle with respect to the direction that differs from the one used for the calculation of the matrix element, where all particles of the process (1) should be described in the same coordinate system, and their projections of the angular momentum are on the same quantization axis. For switching from the wave function $|JM\rangle$ defined in the laboratory fixed direction to the wave function $|J\widetilde{M}\rangle$ of the atomic frame, the coordinate rotation transformation [16] is used:

$$|JM\rangle = \sum_{\widetilde{M}} D^J_{\widetilde{M},M}(\widehat{J})\,|J\widetilde{M}\rangle. \qquad (9)$$

Here $D^J_{\widetilde{M},M}(\widehat{J})$ denotes the Wigner rotation matrix [16], and the hat on J indicates the rotation by solid angle that transforms the atomic frame into the laboratory one used for the polarization measurements of the characteristics depending on J. In the laboratory system of coordinates the matrix element $\langle \alpha_1 J_1 M_1 | Q^{(k)}_q | \alpha_0 J_0 M_0 \rangle$ can be written, by taking into account Eq. (9), in the form

$$\langle \alpha_1 J_1 M_1 | Q^{(k)}_q | \alpha_0 J_0 M_0 \rangle$$

$$= \sum_{\widetilde{M}_0,\widetilde{q},\widetilde{M}_1} \langle \alpha_1 J_1 \widetilde{M}_1 | Q^{(k)}_{\widetilde{q}} | \alpha_0 J_0 \widetilde{M}_0 \rangle$$

$$\times D^{J_0}_{\widetilde{M}_0 M_0}(\widehat{J}_0)\,D^{k}_{\widetilde{q}q}(\widehat{k}_0)\,D^{*J_1}_{\widetilde{M}_1 M_1}(\widehat{J}_1). \quad (10)$$

The matrix element $\langle \alpha_1 J_1 \widetilde{M}_1 | Q^{(k)}_{\widetilde{q}} | \alpha_0 J_0 \widetilde{M}_0 \rangle$ is defined in the atomic frame. The angular momentum part of this matrix element is represented by diagram A_1 in Fig. 1 [10], where the open lines are defined in the laboratory fixed coordinate system, the circles with D inside stand for the Wigner rotation matrices (9), and the rectangle at the lines representing the total angular quanta J of the initial and final states stands for the configuration and other quantum numbers. The application of the Wigner–Eckart theorem [10] enables us to separate the part that is independent of the space rotation (diagram A_2 in Fig. 1) and a Clebsch–Gordan coefficient (diagram A_3 in Fig. 1) that, together with the Wigner rotation ma-

trices (see Fig. 1), describes the directions of an atom in the initial and final states and a photon. The second Clebsch–Gordan coefficient comes up from the diagram of the complex conjugate matrix element (2).

For further simplification of the part describing the space rotation dependence, the following relation can be used:

$$D^J_{\widetilde{M}M}(\widehat{J})\,D^{*J'}_{\widetilde{M}'M}(\widehat{J})$$

$$= \sum_{K,N} T^{*K}_N(J,J',M|\widehat{J}) \begin{bmatrix} J' & K & J \\ \widetilde{M}' & N & \widetilde{M} \end{bmatrix}. \quad (11)$$

The tensor in Eq. (11) is defined as [15]

$$T^{*K}_N(J,J',M|\widehat{J})$$

$$= (-1)^{J'-M} \left[\frac{4\pi}{2J+1}\right]^{1/2} \begin{bmatrix} J & J' & K \\ M & -M & 0 \end{bmatrix} Y^*_{KN}(\theta,\phi). \quad (12)$$

Three Clebsch–Gordan coefficients for each momentum J_0, J_1, and k (see Eq. (11)) are used to perform the summation over the projections $\widetilde{M}_0$, $\widetilde{M}'_0$, $\widetilde{M}_1$, $\widetilde{M}'_1$, $\widetilde{q}$, $\widetilde{q}'$ in Eq. (10) and its complex conjugate. All Clebsch–Gordan coefficients coming up from diagram A_1 and its complex conjugate counterpart as well as for each angular momentum from Eq. (11) are shown in diagram A_4. This summation was performed graphically, and the result is represented by diagram A_5. The closing of open lines from A_5 produces the diagram A_6 invariant under space rotation and the diagram A_7 describing rotation properties of the excitation cross-section (2). The bow on the open lines of diagram A_7 represents the operator (11). The final expression for the excitation probability (2) can be written by using diagrams A_2, A_6, and A_7 in the following form:

$$\frac{d\sigma(\alpha_0 J_0 M_0 \widehat{\epsilon}\mathbf{k}_0 \to \alpha_1 J_1 M_1)}{d\Omega}$$

$$= C \sum_{K_0,K_r,K_1,k,k'} \frac{1}{2K_1+1} B^r(K_0,K_r,K_1,k,k')$$

$$\times \sum_{N_0,N_r,N_1,q} \begin{bmatrix} K_0 & K_r & K_1 \\ N_0 & N_r & N_1 \end{bmatrix} T^{*K_0}_{N_0}(J_0,J_0,M_0|\widehat{J}_0)$$

$$\times T^{*K_r}_{N_r}(k,k',q|\widehat{k}_0)\,T^{K_1}_{N_1}(J_1,J_1,M_1|\widehat{J}_1), \quad (13)$$
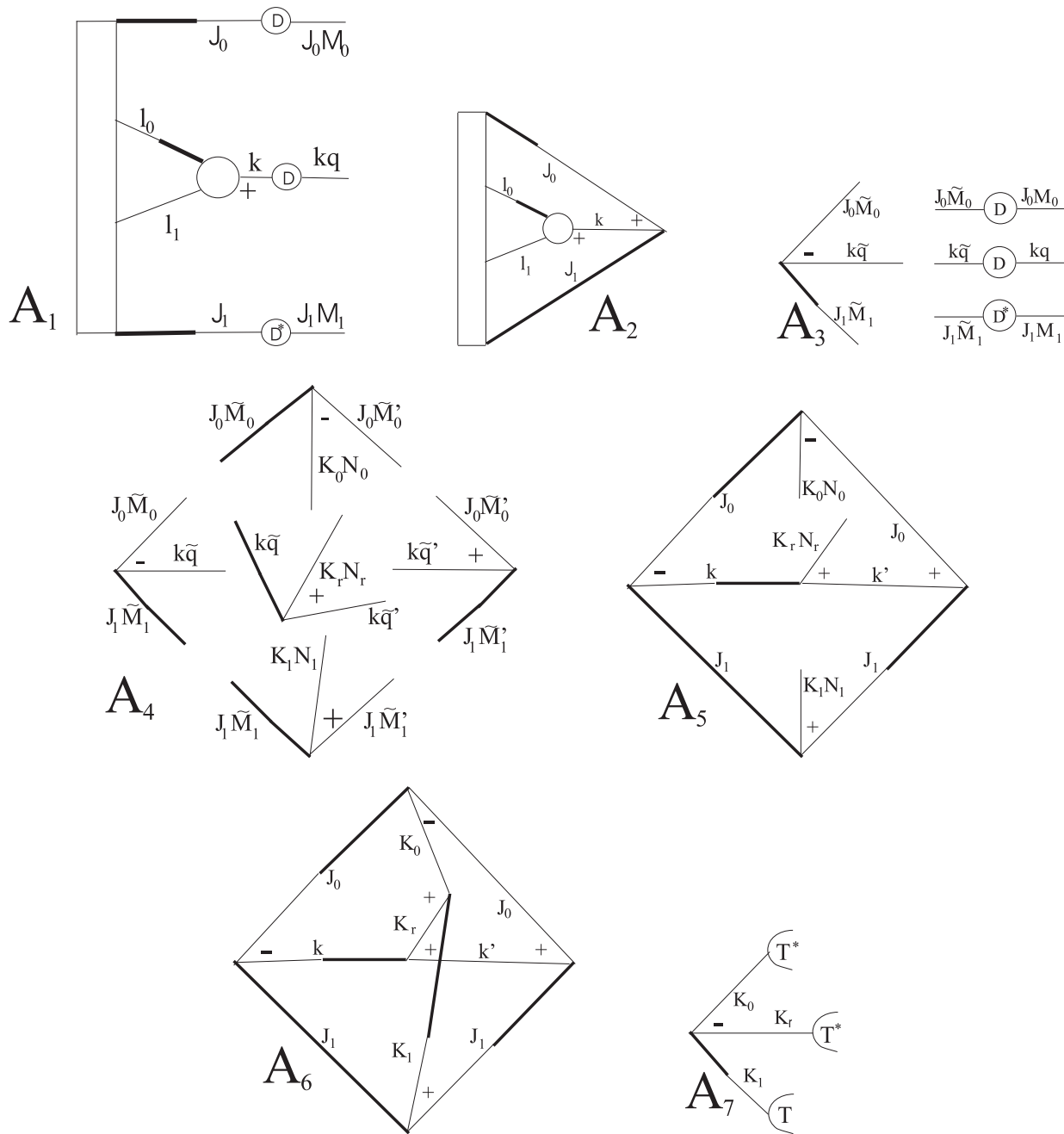
Fig. 1. The diagrams of angular momentum theory used to derive the expression for the probability of excitation of polarized atoms by polarized radiation.

where

$$B^r(K_0, K_r, K_1, k.k')$$

$$= (\alpha_1 J_1 || Q^{(k)} || \alpha_0 J_0)(\alpha_1 J_1 || Q^{(k')} || \alpha_0 J_0)^*$$

$$\times \left\{ \begin{matrix} J_0 & K_0 & J_0 \\ k & K_r & k' \\ J_1 & K_1 & J_1 \end{matrix} \right\} [(2J_0 + 1)(2J_1 + 1)$$

$$\times (2k + 1)(2K_1 + 1)]^{1/2}. \tag{14}$$

In Eq. (14) the relation

$$(\alpha_1 J_1 || Q^{(k)} || \alpha_0 J_0) = [2J_1 + 1]^{1/2} \langle \alpha_1 J_1 || Q^{(k)} || \alpha_0 J_0 \rangle \tag{15}$$

is taken into account. In Eq. (13), $C = 2\pi^2$.

In the case when hyperfine structure is important, the reduced matrix element $(\alpha_1 J_1 || Q^{(k)} || \alpha_0 J_0)$ in Eq. (14) should be changed by $(\alpha_1 J_1(I)F_1 || Q^{(k)} || \alpha_0 J_0(I)F_0)$. A simple relation between these two matrix elements holds:

$$(\alpha_1 J_1(I)F_1\|Q^{(k)}\|\alpha_0 J_0(I)F_0)$$

$$= (-1)^{F_0-J_1+I+k}[(2F_0+1)(2J_1+1)]^{1/2}$$

$$\times \begin{Bmatrix} F_0 & k & F_1 \\ J_1 & I & J_0 \end{Bmatrix} (\alpha_1 J_1\|Q^{(k)}\|\alpha_0 J_0). \qquad (16)$$

I is the spin of the nucleus. The values of J_0, J_1 in Eqs. (14) and (15) should be changed by F_0, F_1. The probability $W(\alpha_0 J_0 M_0\widehat{e}\mathbf{k}_0 \to \alpha_1 J_1 M_1)$ equals to the cross-section (2) divided by the density of the flow of the radiation. Then the same expressions of the cross-section can be used for the probability, only the definition of the constant C should be changed.

3. Photoexcitation as the first step of a multistep process

The excitation of an atom by laser or other electromagnetic radiation is often used to prepare the atom in a polarized state. Then the magnetic state of an atom is not observed, and summation over M_1 has to be performed coherently. In this case, the excitation (1) is the first step of the multistep process, while the second step is

$$A^*(\alpha_1 J_1 M_1) + b(\alpha) \to A(\alpha_2 J_2 M_2) + b'(\alpha'). \quad (17)$$

Here $b(\alpha)$ stands for the impacting particle or electromagnetic radiation in the state α, and $b'(\alpha')$ indicates one or more ionized and emitted particles. In two-step approximation, the probability of the processes (1) and (17) can be written as coherent sum, since the projection M_1 cannot be observed [18]:

$$W^t(\alpha_0 J_0 M_0\widehat{e}\mathbf{k}_0 \to \alpha_1 J_1\alpha \to \alpha_2 J_2 M_2\alpha')$$

$$= C'\Bigg|\sum_{M_1}\langle\alpha_2 J_2 M_2\alpha'|H_2|\alpha_1 J_1 M_1\alpha\rangle$$

$$\times \langle\alpha_1 J_1 M_1|H_1|\alpha_0 J_0 M_0\rangle\Bigg|^2$$

$$= C'\sum_{M_1,M_1'}\langle\alpha_2 J_2 M_2\alpha'|H_2|\alpha_1 J_1 M_1\alpha\rangle$$

$$\times \langle\alpha_2 J_2 M_2\alpha'|H_2|\alpha_1 J_1 M_1'\alpha\rangle^*$$

$$\times \langle\alpha_1 J_1 M_1|H_1|\alpha_0 J_0 M_0\rangle$$

$$\times \langle\alpha_1 J_1 M_1'|H_1|\alpha_0 J_0 M_0\rangle^*. \qquad (18)$$

H_1 and H_2 are the operators of the interaction in the first and second processes, respectively. Then the product (11) should be changed by

$$D^{J_1}_{\widetilde{M_1}M_1}(\widehat{J}_1)\,D^{*J_1'}_{\widetilde{M_1'}M_1'}(\widehat{J}_1)$$

$$= \sum_{K_1,N_1,N_1'}\begin{bmatrix} J_1' & K_1 & J_1 \\ \widetilde{M_1'} & N_1 & \widetilde{M_1} \end{bmatrix}$$

$$\times T^{*K_1}_{N_1 N_1'}(J_1,J_1',M_1,M_1'|\widehat{J}_1), \qquad (19)$$

where the tensorial operator (12) acquires a more general form

$$T^{K_1}_{N_1 N_1'}(J_1,J_1',M_1,M_1'|\widehat{J}_1)$$

$$= (-1)^{J_1'-M_1'}\left[\frac{2K_1+1}{2J_1+1}\right]^{1/2}\begin{bmatrix} J_1 & J_1' & K_1 \\ M_1 & M_1' & N_1' \end{bmatrix}$$

$$\times D^{*K_1}_{N_1 N_1'}(\widehat{J}_1). \qquad (20)$$

Now the summation over M_1 and M_1' in Eq. (18) may be carried out. From the examination of expressions (29) in [4] and (13) of the present work, for both terms in (18) it follows that only the tensors $T^{K_1}_{N_1 N_1'}(J_1,J_1',M_1,M_1'|\widehat{J}_1)$ depend on M_1 and M_1'. The sum over M_1, M_1' of the product of these tensors is equal to

$$\sum_{M_1,M_1'} T^{K_1}_{N_1 N}(J_1,J_1,M_1,M_1'|\widehat{J}_1)$$

$$\times T^{*K_1'}_{N_1' N'}(J_1,J_1,M_1,M_1'|\widehat{J}_1)$$

$$= \frac{\sqrt{(2K_1+1)(2K_1'+1)}}{2J_1+1}D^{*K_1}_{N_1 N}(\widehat{J})\,D^{K_1'}_{N_1' N'}(\widehat{J})$$

$$\times \sum_{M_1,M_1'}(-1)^{2J_1-2M_1'}\begin{bmatrix} J_1 & J_1 & K_1 \\ M_1 & M_1' & N \end{bmatrix}\begin{bmatrix} J_1 & J_1 & K_1' \\ M_1 & M_1' & N' \end{bmatrix}$$

$$= \frac{2K_1+1}{2J_1+1}D^{*K_1}_{N_1 N}(\widehat{J})\,D^{K_1}_{N_1' N'}(\widehat{J})$$

$$\times \delta(K_1,K_1')\delta(N,N'). \qquad (21)$$

Then the quantization axis can be chosen along the z axis of the laboratory coordinate system, and

$$D^{*K_1}_{N_1 N}(0,0,0)D^{K_1}_{N_1' N}(0,0,0) = \delta(N_1,N)\delta(N_1',N).$$

The square root of the multiplier $(2K_1 + 1)/(2J_1 + 1)$ is convenient to attribute to both terms of the following expression:

$$W^t(\alpha_0 J_0 M_0 \widehat{\epsilon} \mathbf{k}_0 \to \alpha_1 J_1 \alpha \to \alpha_2 J_2 M_2 \alpha')$$

$$= \sum_{K_1, N_1} W_{K_1 N_1}(\alpha_0 J_0 M_0 \widehat{\epsilon} \mathbf{k}_0 \to \alpha_1 J_1 \alpha)$$

$$\times W_{K_1 N_1}^A (\alpha_1 J_1 \alpha \to \alpha_2 J_2 M_2 \alpha'). \qquad (22)$$

Here the sum over M_1, M_1' in Eq. (18) is changed by the sum over K_1, N_1, i. e. the probability of the two-step process is expanded as the sum of state multipoles [11]. Then the expressions for the photoexcitation probability and that of the second process slightly change. For the excitation probability, it is

$$\frac{\mathrm{d}W_{K_1 N_1}(\alpha_0 J_0 M_0 \widehat{\epsilon} \mathbf{k}_0 \to \alpha_1 J_1)}{\mathrm{d}\Omega}$$

$$= \frac{C}{[2J_1 + 1]^{1/2}} \sum_{K_0, K_r, k, k'} B^r(K_0, K_r, K_1, k, k')$$

$$\times \sum_{N_0, N_r, q} \begin{bmatrix} K_0 & K_r & K_1 \\ N_0 & N_r & N_1 \end{bmatrix}$$

$$\times T_{N_0}^{*K_0}(J_0, J_0, M_0 | \widehat{J}_0) T_{N_r}^{*K_r}(k, k', q | \widehat{k}_0), \quad (23)$$

$$W_{00}(\alpha_0 J_0 k \to \alpha_1 J_1)$$

$$= \frac{C}{(2J_0 + 1)(2k + 1)[2J_1 + 1]^{1/2}}$$

$$\times |\langle \alpha_1 J_1 \| Q^{(k)} \| \alpha_0 J_0 \rangle|^2. \qquad (24)$$

The expression for the second term in Eq. (22) depends on the second step process. In the case of the Auger decay, it is presented by Kupliauskienė and Tutlys (see Eq. (4) in [19]).

The proposed method is easy to generalize for multistep process when intermediate states are not observed. In the case of a three-step process where the fluorescence radiation of the doubly charged ion formed after the photoionization of an atom and the Auger decay of a singly charged ion is registered, the summation over intermediate states gives

$$\sum_{M_1, M_1', M_2, M_2'} W^1(J_0 M_0 \mathbf{k}_1 \to J_1 M_1 M_1' \mathbf{p}_1 m_1)$$

$$\times W^2(J_1 M_1 M_1' \mathbf{p}_1 m_1 \to J_2 M_2 M_2' \mathbf{p}_2 m_2)$$

$$\times W^3(J_2 M_2 M_2' \mathbf{p}_2 m_2 \to J_3 M_3 \mathbf{k}_2)$$

$$= \sum_{K_1, N_1, K_2, N_2} W_{K_1 N_1}^1(J_0 M_0 \mathbf{k}_1 \to J_1 \mathbf{p}_1 m_1)$$

$$\times \frac{2K_1 + 1}{2J_1 + 1} W_{K_1 N_1 K_2 N_2}^2(J_1 \mathbf{p}_1 m_1 \to J_2 \mathbf{p}_2 m_2)$$

$$\times \frac{2K_2 + 1}{2J_2 + 1} W_{K_2 N_2}^3(J_2 \mathbf{p}_2 m_2 \to J_3 M_3 \mathbf{k}_2). \quad (25)$$

The square root of each multiplier $(2K + 1)/(2J + 1)$ in Eq. (25) is also convenient to attribute to both terms standing side by side.

4. Practical applications

The expressions (13) and (23) represent the general case of the excitation of polarized atoms by polarized radiation. Two cases are chosen to demonstrate their practical application. The first case is the excitation of randomly oriented atoms by the circularly and linearly polarized dipole radiation. The second one is the excitation of polarized atoms by unpolarized radiation. Since usually the photoexcitation is used to prepare an atom in a polarized state, the practical application of the expression (13) is limited. Therefore, the above-mentioned expressions will be obtained for the excitation probability in the first step process characterized by Eq. (23).

4.1. Photoexcitation of unpolarized atoms

To obtain the expression for the probability describing the excitation of an unpolarized atom by polarized dipole radiation, one needs to average the probability (23) over the initial states of an atom. The use of the expression (A1) from [15]

$$\sum_{M_0} T_{N_0}^{K_0}(J_0, J_0, M_0 | \widehat{J}_0) = \delta(K, 0)\delta(N, 0) \qquad (26)$$

in Eq. (23) enables us to write

$$\frac{\mathrm{d}W_{KN}(\alpha_0 J_0 \widehat{\epsilon} \mathbf{k}_0 \to \alpha_1 J_1)}{\mathrm{d}\Omega}$$

$$= \frac{4\pi C}{(2J_0 + 1)\sqrt{3(2J_1 + 1)}}$$

$$\times \sum_{K, q} (-1)^{1-q} B^r(0, K, K, 1, 1)$$

$$\times \begin{bmatrix} 1 & 1 & K \\ q & -q & 0 \end{bmatrix} Y_{KN}(\theta, \phi). \qquad (27)$$

Here θ, ϕ are polar and azimuthal angles between the direction of incoming radiation for the circularly polarized and unpolarized light, or its electrical vector **E** in the case of linearly polarized light, and laboratory z axis. When these directions coincide with the laboratory z axis, by using

$$Y^*_{KN}(0,0) = \left[\frac{2K+1}{4\pi}\right]^{1/2}\delta(N,0), \qquad (28)$$

$$Y_{K0}(\theta,\phi) = \left[\frac{2K+1}{4\pi}\right]^{1/2}P_K(\cos\theta), \qquad (29)$$

the final expression may be written in the form

$$W_{K0}(\alpha_0 J_0 q \rightarrow \alpha_1 J_1 M_1)$$

$$= (-1)^{1-q}\frac{C}{2J_0+1}\left[\frac{2K+1}{3(2J_1+1)}\right]^{1/2}$$

$$\times B^r(0,K,K,1,1)\begin{bmatrix}1 & 1 & K\\ q & -q & 0\end{bmatrix}, \qquad (30)$$

$$B^r(0,K,K,1,1)$$

$$= (\alpha_1 J_1\|Q^{(1)}\|\alpha_0 J_0)^2(-1)^{1+K+J_1+J_0}$$

$$\times\left[\frac{(2J_1+1)3}{2K+1}\right]^{1/2}\begin{Bmatrix}1 & 1 & K\\ J_1 & J_1 & J_0\end{Bmatrix}, \qquad (31)$$

and $P_K(\cos\theta)$ is the Legendre polynomial [16].

In the case of circularly polarized electrical dipole radiation, $k = 1$, $q = \pm 1$, $K = 0, 1, 2$, the excited atom is oriented along the laboratory z axis if $J_1 \geq 1/2$ because of the condition $K \leq 2J_1$. The ranks $K = 1$ and $K = 2$ describe the orientation and alignment [11], respectively. For the linearly polarized light, $q = 0$ and $K = 0, 2$ because the Clebsch–Gordan coefficient in Eq. (30) is equal to zero when $q = 0$ and $2 + K$ is odd [10]. The unpolarized light is described by the sum of equal parts of the right- and left-hand circularly polarized radiation. Then the terms of the sum with $K = 1$ cancel, and only the terms with $K = 0, 2$ bring the contribution like in the case of linearly polarized light, but one must have in mind that $q = 1$. This means that the excited atom is created in the aligned state even if excited by unpolarized radiation. For the description of orientation and alignment, the parameters A_1 and A_2, respectively, were introduced [12]. In the case of ex-

citation by electrical dipole radiation, their expressions are

$$A_1 = \frac{W_{10}}{W_{00}} = 3\sqrt{2J_1+1}(-1)^{J_1+J_0+1-q}$$

$$\times\begin{bmatrix}1 & 1 & 1\\ q & -q & 0\end{bmatrix}\begin{Bmatrix}1 & 1 & 1\\ J_1 & J_1 & J_0\end{Bmatrix}, \qquad (32)$$

$$A_2 = \frac{W_{20}}{W_{00}} = 3\sqrt{2J_1+1}(-1)^{J_1+J_0-q}$$

$$\times\begin{bmatrix}1 & 1 & 2\\ q & -q & 0\end{bmatrix}\begin{Bmatrix}1 & 1 & 2\\ J_1 & J_1 & J_0\end{Bmatrix}. \qquad (33)$$

For the linearly polarized dipole radiation, $A_1 = 0$, and

$$A_2^L = \sqrt{6(2J_1+1)}(-1)^{J_1+J_0}\begin{Bmatrix}1 & 1 & 2\\ J_1 & J_1 & J_0\end{Bmatrix}. \qquad (34)$$

In the case of circularly polarized dipole radiation, A_2 coincides with that of unpolarized radiation:

$$A_2 = \left[\frac{3(2J_1+1)}{2}\right]^{1/2}(-1)^{J_1+J_0-1}\begin{Bmatrix}1 & 1 & 2\\ J_1 & J_1 & J_0\end{Bmatrix} \qquad (35)$$

and

$$A_1 = 3\left[\frac{2J_1+1}{2}\right]^{1/2}(-1)^{J_1+J_0}\begin{Bmatrix}1 & 1 & 1\\ J_1 & J_1 & J_0\end{Bmatrix}. \qquad (36)$$

The calculated parameters describing the orientation A_1 (36) and alignment A_2 (34)–(35) of atoms excited by the electrical dipole radiation are presented in Table 1. The orientation and alignment of the resonance state in the case of photoexcitation are well-determined and both are model-independent because they do not depend on the values of the reduced matrix elements (see Eqs. (32)–(36)). Therefore, they can be used for the investigation of any atom in any experiment. The values of these coefficients can differ from those obtained by using the density matrix formalism by $\sqrt{2J_1+1}$ (e. g., [9]) because of our definition (25) where $[(2K+1)/(2J+1)]^{1/2}$ is attributed to each term standing side by side.

The data from Table 1 show that all atoms excited to the states $J_1 \geq 1/2$ by electric dipole radiation are oriented ($J_1 \geq 1/2$) and aligned ($J_1 \geq 1$). This polarization was observed in the second step process products. The transferred spin polarization of the resonantly excited $3d_J^{-1}5p$ Auger electrons of krypton was registered [20] to be 80%.

Table 1. Parameters describing orientation A_1 and alignment A_2 in the case of right circularly polarized dipole radiation and alignment A_2^{L} for the linearly polarized radiation.

J_0	J_1	A_1	A_2	A_2^{L}	J_0	J_1	A_1	A_2	A_2^{L}
0	1	1.2247	0.7071	−1.4142	5/2	3/2	−0.6708	0.1	−0.2
1/2	1/2	1	0	0		5/2	0.2928	−0.4276	0.8552
	3/2	1.1180	0.5	−1		7/2	0.9820	0.3873	−0.6546
1	0	0	0	0	3	2	−0.7071	0.1195	−0.2390
	1	0.6124	−0.3536	0.7071		3	0.25	−0.4330	0.8660
	2	1.0607	0.4183	−0.8367		4	0.9682	0.3134	−0.6268
3/2	1/2	−0.5	0	0	7/2	5/2	−0.7319	0.1336	−0.2673
	3/2	0.4472	−0.4	0.8		7/2	0.2182	−0.4364	0.8729
	5/2	1.0247	0.3742	−0.7483		9/2	0.9574	0.3028	−0.6055
2	1	−0.6124	0.0707	−0.1414	4	3	−0.75	0.1443	−0.2887
	2	0.3536	−0.4183	0.8367		4	0.1936	−0.4387	0.8775
	3	1	0.3464	−0.6928		5	0.9487	0.2944	−0.5889

4.2. Photoexcitation of polarized atoms

In the second case of the excitation of polarized atoms by unpolarized radiation, one needs to take into account the earlier mentioned description of unpolarized light and the condition that $K = 0, 2$, as well as that the laboratory z axis is aligned along the direction of incoming radiation. Then $N_r = 0$ due to the relation (28), and the contribution of terms of only the same multipolarity Eq. (22) acquires a simpler form:

$$W_{K_1 N_1}(\alpha_0 J_0 M_0 q \to \alpha_1 J_1)$$

$$= C \left[\frac{4\pi}{(2J_0 + 1)(2k + 1)(2J_1 + 1)} \right]^{1/2}$$

$$\times (-1)^{J_0 - M_0 + 1 - q} \sum_{K_0, K_r} [2K_r + 1]^{1/2}$$

$$\times B^r(K_0, K_r, K_1, 1, 1) \begin{bmatrix} K_0 & K_r & K_1 \\ N_1 & 0 & N_1 \end{bmatrix}$$

$$\times \begin{bmatrix} J_0 & J_0 & K_0 \\ M_0 & -M_0 & 0 \end{bmatrix} \begin{bmatrix} k & k & K_r \\ q & -q & 0 \end{bmatrix} Y_{K_0 N_1}(\theta, \phi).$$

(37)

The angles θ and ϕ of the total angular momentum J_0 are measured from the direction of incoming radiation.

In the electrical dipole approximation, $K_r = 0, 2$, Eq. (37) can be written as

$$W_{K_1 N_1}(\alpha_0 J_0 M_0 \to \alpha_1 J_1)$$

$$= W_{00}(\alpha_0 J_0 \to \alpha_1 J_1)$$

$$\times [1 + A_2(J_0 M_0, K_1 N_1, \theta, \phi)],$$

(38)

$$A_2(J_0 M_0, K_1 N_1, \theta, \phi)$$

$$= \sum_{K_0} \left[\frac{4\pi}{2K_0 + 1} \right]^{1/2}$$

$$\times Y_{K_0 N_1}(\theta, \phi) \beta(K_0, 2, K_1) \begin{bmatrix} K_0 & 2 & K_1 \\ N_1 & 0 & N_1 \end{bmatrix},$$

(39)

$$\beta(K_0, 2, K_1)$$

$$= (-1)^{J_0 - M_0 + 1 - q} 3(2J_0 + 1)$$

$$\times [(2J_1 + 1)(2K_1 + 1)(2K_0 + 1)(2K_r + 1)]^{1/2}$$

$$\times \begin{bmatrix} 1 & 1 & K_r \\ q & -q & 0 \end{bmatrix} \begin{bmatrix} J_0 & J_0 & K_0 \\ M_0 & -M_0 & 0 \end{bmatrix} \begin{Bmatrix} J_0 & K_0 & J_0 \\ 1 & 2 & 1 \\ J_1 & K_1 & J_1 \end{Bmatrix}.$$

(40)

Parameter $A_2(J_0 M_0, K_1 N_1, \theta, \phi)$ describes the alignment of the excited atom that was created by the photoexcitation of a polarized atom. It depends on the orientation of the atom in its initial state, but is independent of the value of the reduced matrix element. The values of $A_2(J_0 M_0, K_1 N_1)$ can be calculated if the orientation of the atom is known. In the case of J_0 aligned along the z axis that coincides with the direction of exciting radiation, the expression for $A_2(J_0 M_0, K_1 N_1)$ is simple, because $M_0 = J_0$, $K_0 + K_1 =$ even, and $N_1 = 0$:

$$A_2(J_0 J_0, K_1 0, 0, 0)$$

$$= 3(2J_0 + 1)$$

$$\times \sum_{K_0 \leq 2J_0} [5(2J_1 + 1)(2K_1 + 1)(2K_0 + 1)/6]^{1/2}$$

$$\times \begin{bmatrix} K_0 & 2 & K_1 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} J_0 & J_0 & K_0 \\ J_0 & -J_0 & 0 \end{bmatrix} \begin{Bmatrix} J_0 & K_0 & J_0 \\ 1 & 2 & 1 \\ J_1 & K_1 & J_1 \end{Bmatrix}.$$

(41)

Table 2. Parameters describing the alignment $A_2(J_0, J_0 = M_0, K_10, 0, 0)$ in the case of the photoexcitation of atoms polarized along the direction of incoming unpolarized radiation ($J_0 \leq 3$, $J_1 \leq 3$).

J_0	J_1	K_1					
		1	2	3	4	5	6
0	1		1.58811				
1/2	1/2	−1.1547					
	3/2	0.2582		1.7748			
1	1		−1.5811				
	2		1.0690		1.4432		
3/2	1/2	0.8660					
	3/2	−1.2394		−1.4199			
	5/2	0.4564		1.6424		1.0235	
2	1		0.7906				
	2		−2.0045		−1.0690		
	3		1.1066		1.7665		0.6725
5/2	3/2	1.1619		0.5916			
	5/2	−1.5937		−2.2082		−0.7311	
3	2		1.3363		0.4009		
	3		−2.4206		−1.9988		−0.4707

In Eq. (41), $K_1 \leq 2J_1$.

The calculated values for $A_2(J_0 J_0, K_10, 0, 0)$ in the case of $J_0 \leq 3$ and $J_1 \leq 3$ are presented in Table 2. They are also model-independent and can be used for the investigation of any atom in any experiment.

5. Concluding remarks

To summarize, a general expression for the excitation probability of polarized atoms by polarized radiation was obtained. The integration over the orbital and spin variables was performed by using the graphical technique of angular momentum. The approach is based on the ordinary atomic theory and is an alternative to the density matrix formalism in the case of pure states. Since usually the photoexcitation is used to prepare an atom in a polarized state, the general expression was also obtained for the probability of excitation as the first step process. The generalization of the obtained expressions in the case of a multistep process when intermediate states are not observed is presented. Two cases are chosen to demonstrate the practical application. The first case is the excitation of randomly oriented atoms by the unpolarized and by the circularly and linearly polarized dipole radiation. The second one is the excitation of polarized atoms by unpolarized radiation. The parameters describing the orientation and alignment in the case of photoexcitation are well-determined and both are model-independent because they do not depend on the values of the reduced matrix elements. Therefore, the calculated values of these parameters in the present work can be used for the investigation of any atom in any experiment.

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POLIARIZUOTO ATOMO SUŽADINIMAS POLIARIZUOTA SPINDULIUOTE

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Santrauka

Derinamo dažnio lazerio ir sinchrotroninė spinduliuotės atveria galimybę ne tiktai jonizuoti atomus, atplėšiant elektronus iš pageidaujamo sluoksnio, bet ir juos sužadinti į pageidaujamą būseną. Šitaip paruošiami poliarizuoti atomai, kurie toliau naudojami poliarizacijos reiškiniams jonizuojant atomus elektronais ir fotonais tirti. Norint vieningai teoriškai aprašyti atomo sužadinimo ir po jo einančius jonizacijos vyksmus bei tirti, kaip antrąjį vyksmą apibūdinantys dydžiai priklauso nuo pirmojo, reikalingos kuo bendresnės tokius vyksmus aprašančios formulės. Panaudojant judėjimo kiekio momento grafinį vaizdavimą ir atomo teorijos metodus, surasta poliarizuoto atomo sužadinimo poliarizuota spinduliuote tikimybės išraiška. Grynųjų būsenų atveju panaudotasis metodas yra alternatyvus iki šiol taikytam tankio matricos formalizmui. Surastoji formulė taip pat aprašo sužadinto atomo poliarizaciją. Ji panaudota nepoliarizuoto atomo sužadinimo poliarizuota spinduliuote ir poliarizuoto atomo sužadinimo nepoliarizuota spinduliuote tikimybės išraiškoms rasti, laikant, kad sužadinimas tėra pirmoji tolimesnio vyksmo stadija. Taip pat parodyta, kokius pakeitimus reikia padaryti, kad formulės būtų pritaikytos daugiapakopiams vyksmams aprašyti.