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Partial-photoelectron-wave analysis in diatomic molecule photoionization by fluorescence polarization experiments

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Parameters $\beta 1$ and $\beta 2$ describing the angular distribution of circularly polarized fluorescence photons emitted by randomly oriented diatomic molecules excited by circularly polarized radiation are derived. We prove theoretically that measuring these parameters allows a partial-wave analysis of the emitted photoelectrons in the case of closed-shell diatomic molecules. The determination of the relative partial cross sections for emission of the $\varepsilon\sigma$, $\varepsilon\pi$, and $\varepsilon\delta$ photoelectrons is of fundamental importance for a sensitive test of corresponding calculations.

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Partial-wave analysis (PWA) is the main tool for studying high-energy particle reactions, allowing us to (i) express measured phase-space distributions in terms of complicated dynamical functions for partial waves, (ii) access specific density matrix elements for the reaction, (iii) conclude on reaction channels for production of particles with specific spin and parity, and (iv) study thereby long-range nuclear interactions (see, e.g., Refs. [1–3] and references therein). Photoionization, on the other hand, is one of the most fundamental processes in nature and is a main tool for the investigation of the electronic structure of matter. Recently, PWA has been utilized for studying photoionization processes of atoms [4–6], allowing experimental determination of partial cross sections for the emission of electron waves of different angular momenta at the same kinetic energy. Thereby, considerably more information on the mechanisms of interaction of matter with light is obtainable as compared to experiments measuring photoelectron intensities and kinetic energies only. Since the relative contributions of the partial waves are determined by the dynamics of a specific photoionization process, PWA is fundamental for explaining how a many-body quantum system responds collectively on a probing photon and is the most sensitive test for theoretical methods and models for atomic photoionization and structure [7].

Cross sections for partial waves emitted on a photoionization event may be experimentally determined by measurements of spin-sensitive intensities for angularly resolved Auger electrons [8–10] or via the determination of the degrees of linear and circular polarization of fluorescence emitted by a subsequent relaxation of the residual ion [4–6]. For both types of experiments with atoms the underlying theory is existing (see Refs. [8–10] and [4–6], respectively). However, to our knowledge, there are neither experimental nor theoretical studies of this problem in molecules, and it is not known whether a PWA in molecules is possible at all. Here we report a theoretical proof that PWA for molecular photoionization is possible and suggest an experiment for the determination of relative partial-wave cross sections in diatomic molecule photoionization via the degrees of linear and circular polarization of fluorescence emitted by a subsequent electric dipole transition within the residual ion. This theory paves the way for a multitude of new experiments. The present general theory formalism can be straightforwardly extended to the description of more complicated nonlinear polyatomic molecules and to other areas of photoionization processes, like, e.g., multiphoton photoionization. Moreover, the experimental determination of partial molecular photoionization cross sections provides a conclusive test for available computational approaches for the electron continuum problem in molecules, which is a very complicated and actual theoretical problem alone.

One-photon ionization of a closed-shell atom ($J_0 = 0$) results, in general, in the emission of three partial photoelectron waves with $j = J_1 + 1, J_1, J_1 - 1$, where J_0, J_1, and j are the total electronic angular momenta of the ground atomic state, residual ion, and photoelectron, respectively. Since the sum of the three relative partial cross sections, $\gamma_j = \sigma_j/\sigma_{\text{tot}}$, is normalized ($\Sigma_j \gamma_j = 1$), there are two independent partial-wave cross sections γ_j which must be determined. A quadrupole transition, occurring, e.g., in multiphoton photoionization processes, may result in the emission of additional partial electron waves γ_j. In this case, the experimental determination of more than two independent observables will be necessary. Below we restrict our attention to the case, where the dipole approximation is valid. In fluorescence experiments with atoms, PWA is based on the determination of the two parameters $\beta 1$ and $\beta 2$ for the angular distribution of fluorescence photons emitted by a subsequent relaxation of the ion. In contrast to the angular distribution parameter for photoelectrons, β^e, where all three partial waves and the corresponding phase differences enter coherently, both fluorescence parameters $\beta 1$ and $\beta 2$ are independent of the phase difference between outgoing partial photoelectron waves, and, for closed-shell atoms, depend on the partial cross sections γ_j incoherently [11–13] (see also Ref. [14] and references therein). These fluorescence angular distribution parameters may be obtained from an analysis of the degrees of linear (P_{lin}) and circular (P_{circ}) polarization of fluorescence photons as given in Eqs. (2.17a) and (2.17b) of Ref. [15] (see also Refs. [4–6]). Thus, a PWA for photoionization of closed-shell atoms can be performed based on fluorescence experiments *only*.

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The angular distribution parameter $\beta 2$ for fluorescence photons emitted by randomly oriented diatomic molecules excited by linearly polarized radiation has been derived already in previous work [16]. Good agreement between the measured and computed fluorescence angular distribution parameter $\beta 2$ was found there. In order to analyze both parameters $\beta 1$ and $\beta 2$, we report here a general theory for angular distribution of circularly polarized fluorescence photons emitted by randomly oriented diatomic molecules excited by circularly polarized radiation. We consider the following stepwise process in randomly oriented diatomic molecules AB excited from their ground states $\Omega_0 v_0 = 0$ by circularly polarized photons $\gamma_\pm$ of energy $\hbar\omega$ (± stands for positive and negative helicity or right- and left-handed circular polarization):

$$\begin{aligned} &AB(\Omega_0 v_0 = 0) + \gamma_\pm(\hbar\omega) \\ &\quad\to AB^+(\Omega_1 v_1) + e^-(\varepsilon\ell m\mu) \\ &\quad\to AB^+(\Omega_2 v_2) + e^-(\varepsilon\ell m\mu) + \gamma'_\pm(hc/\lambda). \end{aligned} \tag{1}$$

The outgoing photoelectron e^- with energy ε can be expanded in the asymptotical region via partial waves [17] with fixed projections m and μ of the orbital angular momentum ℓ and spin s, respectively. The photoionization event results in the population of vibronic states of the molecular ion AB^+ with a given projection of the total electronic angular momentum Ω_1 along the molecular axis and a given vibrational quantum number v_1. These excited ionic states decay further via emission of a circularly polarized fluorescence photon $\gamma'_\pm$ with energy hc/λ (± stands again for positive and negative helicity). The quantization axis in the laboratory frame (z) coincides with the propagation direction of the exciting photons and in the molecular frame (z') with the molecular axis.

Proceeding along the same lines as suggested in Ref. [17] for the calculation of the photoelectron anisotropy parameter of diatomic molecules, we have derived equations describing the angular distribution of the fluorescence emitted in the second step of process (1). For this purpose, both circularly polarized incoming $\gamma_\pm$ and outgoing $\gamma'_\pm$ photons, as well as the photoelectron partial waves $\varepsilon\ell m\mu$ with observable quantum numbers in the laboratory frame, have been, at first, transformed into the molecular frame. As a result, both photoionization and radiative decay transition matrix elements are given in the molecular frame. Afterward, the fluorescence intensity has been integrated over all directions of emission of the photoelectron, since it is not observed in coincidence with the fluorescence photon. Finally, the fluorescence intensity has been averaged over all orientations of the molecular axis in the laboratory frame.

For randomly oriented diatomic molecules excited by circularly polarized light $\gamma_\pm$, the angular distribution of circularly polarized fluorescence radiation $\gamma'_\pm$ emitted via the $|\Omega_1 v_1\rangle \to |\Omega_2 v_2\rangle$ radiative decay is given by the following formula for the differential fluorescence intensity:

$$\left(\frac{dI^{\pm\gamma'}_{\pm\gamma}(\theta,\omega)}{d\Omega}\right)^{\Omega_2 v_2}_{\Omega_1 v_1} = \frac{1}{2}\frac{I^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)}{4\pi}\left[1 + (-1)^{n+n'}\beta 1^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)\cos\theta - \frac{1}{2}\beta 2^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)P_2(\cos\theta)\right], \tag{2}$$

where θ is the angle between the direction of propagation of the exciting radiation (z axis) and the direction of detection of the fluorescence radiation and $P_2(\cos\theta)$ is the second Legendre polynomial. Here, each of the quantities n and n' are equal to 0 for positive helicities or 1 for negative helicities of both the incoming $\gamma_\pm$ and outgoing $\gamma'_\pm$ photons, respectively. We note that Eq. (2) for diatomic molecules coincides with the corresponding equation for atoms [13,15]. The total fluorescence intensity, $I^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)$, is the product of the total photoionization cross section, $\sigma_{\Omega_1 v_1}(\omega)$, and the fluorescence yield, $\chi^{\Omega_2 v_2}_{\Omega_1 v_1}$. Their explicit computational forms can be found in Ref. [16].

The presently derived equations for the angular distribution parameters $\beta 1^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)$ and $\beta 2^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega)$ read

$$\begin{aligned} \beta 1^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega) = {}& \frac{1}{N}\sum_{\Omega_0\Omega_2}\sum_{qq'}\sum_{\Omega_1\Omega'_1}\sum_{\ell m\mu}\sum_{kk'}\frac{9}{2}(-1)^{k+q'} \\ &\times\begin{pmatrix}1 & 1 & 1\\ k & -k' & q-q'\end{pmatrix}\begin{pmatrix}1 & 1 & 1\\ q & -q' & k-k'\end{pmatrix} \\ &\times\langle\Omega_2 v_2|\mathbf{d}_{q'}|\Omega'_1 v_1\rangle^*\langle\Omega'_1 v_1\varepsilon\ell m\mu|\mathbf{d}_{k'}|\Omega_0 v_0\rangle^* \\ &\times\langle\Omega_2 v_2|\mathbf{d}_q|\Omega_1 v_1\rangle\langle\Omega_1 v_1\varepsilon\ell m\mu|\mathbf{d}_k|\Omega_0 v_0\rangle, \end{aligned} \tag{3a}$$

$$\begin{aligned} \beta 2^{\Omega_2 v_2}_{\Omega_1 v_1}(\omega) = {}& \frac{1}{N}\sum_{\Omega_0\Omega_2}\sum_{qq'}\sum_{\Omega_1\Omega'_1}\sum_{\ell m\mu}\sum_{kk'}3(-1)^{k+q'+1} \\ &\times\begin{pmatrix}1 & 1 & 2\\ k & -k' & q-q'\end{pmatrix}\begin{pmatrix}1 & 1 & 2\\ q & -q' & k-k'\end{pmatrix} \\ &\times\langle\Omega_2 v_2|\mathbf{d}_{q'}|\Omega'_1 v_1\rangle^*\langle\Omega'_1 v_1\varepsilon\ell m\mu|\mathbf{d}_{k'}|\Omega_0 v_0\rangle^* \\ &\times\langle\Omega_2 v_2|\mathbf{d}_q|\Omega_1 v_1\rangle\langle\Omega_1 v_1\varepsilon\ell m\mu|\mathbf{d}_k|\Omega_0 v_0\rangle, \end{aligned} \tag{3b}$$

with the normalization coefficient

$$N = \sum_{\Omega_0\Omega_2}\sum_{\Omega_1 kq}\sum_{\ell m\mu}|\langle\Omega_2 v_2|\mathbf{d}_q|\Omega_1 v_1\rangle\langle\Omega_1 v_1\varepsilon\ell m\mu|\mathbf{d}_k|\Omega_0 v_0\rangle|^2. \tag{3c}$$

Summations over indices $\Omega_0, \Omega_1, \Omega'_1$, and Ω_2 in Eqs. (3) must be performed over all degenerate electronic states $|\Omega_0\rangle$, $|\Omega_1\rangle$, and $|\Omega_2\rangle$. Equation (3b) coincides with the equation for the angular distribution parameter $\beta 2$ for fluorescence photons emitted by randomly oriented diatomic molecules excited by linearly polarized radiation [16]. However, in contrast to Ref. [16], parameter $\beta 2$ enters the differential fluorescence intensity (2) with a factor of $-\frac{1}{2}$, similar to the case of atomic fluorescence $\gamma'_{\pm}$ excited by $\gamma_{\pm}$ photons [13,15].

Both photoionization amplitudes entering Eqs. (3a) and (3b) contain photoelectrons with equal quantum numbers. This is a consequence of the integration of the fluorescence intensity over all directions of photoelectron emission. Therefore, both parameters $\beta 1$ and $\beta 2$ are independent of the phase difference between the partial electron waves. In the general case, Eqs. (3a) and (3b) cannot be further simplified to an incoherent sum of partial photoionization cross sections, since they contain the coherent summation over amplitudes corresponding to the population-depopulation of different degenerate electronic substates $|\Omega_1\rangle$ and $|\Omega'_1\rangle$. Moreover, similar to the case of open-shell atoms [14], the cross terms between these excitation-deexcitation amplitudes are also allowed for open-shell molecules [18], resulting in interference effects observable in angularly resolved fluorescence spectra. Because of this coherent summation of the partial waves in Eqs. (3a) and (3b), PWA for photoionization of open-shell molecules could become complicated.

For closed-shell molecules with $\Omega_0 = 0$, however, Eqs. (3a) and (3b) can be simplified. There, the cross terms for different degenerate electronic states with $\Omega_1 \neq \Omega'_1$ are forbidden due to the two following required conditions: (i) both photoionization amplitudes must describe a photoelectron with equal quantum numbers $\varepsilon\ell m\mu$; and (ii) for both $3j$ symbols it is necessary that $k' - k = q - q'$. This immediately implies $\Omega_1 = \Omega'_1$, $k = k'$, and $q = q'$. Thus, for closed-shell molecules the parameters $\beta 1$ and $\beta 2$ are, indeed, given by an incoherent sum of the partial photoelectron waves, similar to the case of closed-shell atoms [11–14]. In addition, the radiative decay probabilities $|\langle\Omega_2 v_2|\mathbf{d}_q|\Omega_1 v_1\rangle|^2$ are independent of the signs of the $\pm\Omega_1$ and $\pm\Omega_2$. As a result, these probabilities cancel in Eqs. (3a) and (3b) due to the normalization coefficient (3c). From the latter fact it follows that both parameters are independent of the vibrational quantum number v_2, and one should expect the same angular distributions for all fluorescence bands within an $\Omega_1(v_1 = \text{const}) \to \Omega_2(v_2)$ vibrational progression [16].

For simplicity, closed-shell diatomic molecules with $\Omega_0 = 0$, consisting of second-period atoms, are considered below. According to the conservation of the projection of angular momentum on the molecular z' axis ($\Omega_0 + k = \Omega_1 + m + \mu$), photoionization may result in emission of three partial photoelectron waves with $|m| = 0, 1$, and 2, corresponding to $\varepsilon\sigma$, $\varepsilon\pi$, and $\varepsilon\delta$ photoelectrons, respectively. Thus, it is possible to introduce a partial cross section for the emission of photoelectrons with fixed quantum numbers $|m|$ by summing the total photoionization cross section (Eq. (3) of Ref. [16]) over the two $\pm m$ projections

$$\sigma^{|m|}_{\Omega_1 v_1}(\omega) = \frac{4\pi^2\alpha a_0^2\omega}{3g_{\Omega_0}} \times \sum_{\Omega_0\Omega_1}\sum_{\ell\mu k}\sum_{\text{sgn}(m)} |\langle\Omega_1 v_1 \varepsilon\ell m\mu|\mathbf{d}_k|\Omega_0 v_0\rangle|^2, \tag{4a}$$

$$\sigma_{\Omega_1 v_1}(\omega) = \sum_{|m|=0}^{2} \sigma^{|m|}_{\Omega_1 v_1}(\omega), \tag{4b}$$

and to define relative partial cross sections as

$$\gamma^{|m|}_{\Omega_1 v_1}(\omega) = \sigma^{|m|}_{\Omega_1 v_1}(\omega)/\sigma_{\Omega_1 v_1}(\omega), \tag{5a}$$

$$1 = \sum_{|m|=0}^{2} \gamma^{|m|}_{\Omega_1 v_1}(\omega). \tag{5b}$$

In the present designations, Eqs. (3a) and (3b) can now be simplified to the following incoherent sum of relative partial cross sections (5)

$$\beta 1^{\Omega_2}_{\Omega_1 v_1}(\omega) = \sum_{|m|=0}^{2} \frac{9}{2}(-1)^{k+q} \begin{pmatrix} 1 & 1 & 1 \\ k & -k & 0 \end{pmatrix} \times \begin{pmatrix} 1 & 1 & 1 \\ q & -q & 0 \end{pmatrix} \gamma^{|m|}_{\Omega_1 v_1}(\omega), \tag{6a}$$

$$\beta 2^{\Omega_2}_{\Omega_1 v_1}(\omega) = \sum_{|m|=0}^{2} 3(-1)^{k+q'+1} \begin{pmatrix} 1 & 1 & 2 \\ k & -k & 0 \end{pmatrix} \times \begin{pmatrix} 1 & 1 & 2 \\ q & -q & 0 \end{pmatrix} \gamma^{|m|}_{\Omega_1 v_1}(\omega), \tag{6b}$$

with the k and q values running over $0, \pm 1$ and given by the conservation of the projection of angular momentum on the molecular z' axis: $k = \Omega_1 + m + \mu$ and $q = \Omega_1 - \Omega_2$. From Eqs. (6a) and (6b) it is obvious that for closed-shell diatomic molecules it must be possible to determine two independent partial waves $\gamma^{|m|}$ from the measured parameters $\beta 1$ and $\beta 2$.

As an example, let us consider photoionization of the CO molecule into the $\text{CO}^+(A\,^2\Pi)$ state with subsequent fluorescence decay into the $\text{CO}^+(X^2\Sigma^+)$ state

$$\begin{aligned}&\text{CO}(1\pi^4 5\sigma^2 X\,^1\Sigma^+, \Omega_0 = 0, v_0 = 0) + \hbar\omega \\ &\to \text{CO}^+\big(1\pi^3 5\sigma^2 A\,^2\Pi, \Omega_1 = \pm\tfrac{1}{2}(\pm\tfrac{3}{2}), v_1\big) + \varepsilon\sigma(\varepsilon\pi, \varepsilon\delta) \\ &\to \text{CO}^+\big(1\pi^4 5\sigma^1 X\,^2\Sigma^+, \Omega_2 = \pm\tfrac{1}{2}, v_2\big) + \varepsilon\sigma(\varepsilon\pi, \varepsilon\delta) + hc/\lambda.\end{aligned} \tag{7}$$

Spin-orbit interaction of the 1π electrons results in the nondegeneracy of the $\Omega_1 = \pm\frac{1}{2}$ and $\Omega_1 = \pm\frac{3}{2}$ fine structure components of the CO^+ $A\,^2\Pi(\Omega_1)$ state. This nondegeneracy is the reason for the appearance of two components of each vibrational band of the $\text{CO}^+(A\,^2\Pi \to X\,^2\Sigma^+)$ fluorescence spectrum corresponding to the $\Omega_1 = \pm\frac{1}{2} \to \Omega_2 = \pm\frac{1}{2}$ and $\Omega_1 = \pm\frac{3}{2} \to \Omega_2 = \pm\frac{1}{2}$ transitions (typical separation is about $\Delta\lambda = 3$ nm at $\lambda = 500$ nm [16,19]). Therefore, summations in Eqs. (3) must be performed over the $\Omega_1, \Omega'_1 = \pm\frac{1}{2}$ degenerate substates for the $|\Omega_1 = \frac{1}{2}\rangle$ state, and over $\Omega_1, \Omega'_1 = \pm\frac{3}{2}$ ones for the $|\Omega_1 = \frac{3}{2}\rangle$ state, separately.

The fluorescence angular distribution parameters (6a) and (6b) for the process (7) are then given by the following incoherent sum of the relative partial cross sections (5)

$$\beta 1^{X\Omega_2}_{A\Omega_1 v_1}(\omega) = -\frac{3}{4}\gamma^{\varepsilon\sigma}_{A\Omega_1 v_1}(\omega) + \frac{3}{4}\gamma^{\varepsilon\delta}_{A\Omega_1 v_1}(\omega), \tag{8a}$$

$$\beta 2^{X\Omega_2}_{A\Omega_1 v_1}(\omega) = -\frac{\gamma^{\varepsilon\sigma}_{A\Omega_1 v_1}(\omega)}{10} + \frac{\gamma^{\varepsilon\pi}_{A\Omega_1 v_1}(\omega)}{5} - \frac{\gamma^{\varepsilon\delta}_{A\Omega_1 v_1}(\omega)}{10}, \tag{8b}$$

where the kinematic coefficients are the same for the $|A\,^2\Pi, \Omega_1 = \frac{1}{2}\rangle$ and $|A\,^2\Pi, \Omega_1 = \frac{3}{2}\rangle$ initial fluorescence states. The system of linear algebraic Eqs. (5b), (8a), and (8b) is nondegenerate. Thus, the relative partial-wave cross sections

(5) for the processes (7) can be determined from the measured parameters for angular distribution of the $\mathrm{CO}^+(A\,^2\Pi \rightarrow X\,^2\Sigma^+)$ fluorescence via

$$\gamma^{\varepsilon\sigma}_{A\Omega_1 v_1}(\omega) = \frac{1}{3} - \frac{2}{3}\beta 1^{X\Omega_2}_{A\Omega_1 v_1}(\omega) - \frac{5}{3}\beta 2^{X\Omega_2}_{A\Omega_1 v_1}(\omega), \tag{9a}$$

$$\gamma^{\varepsilon\pi}_{A\Omega_1 v_1}(\omega) = \frac{1}{3} + \frac{10}{3}\beta 2^{X\Omega_2}_{A\Omega_1 v_1}(\omega), \tag{9b}$$

$$\gamma^{\varepsilon\delta}_{A\Omega_1 v_1}(\omega) = \frac{1}{3} + \frac{2}{3}\beta 1^{X\Omega_2}_{A\Omega_1 v_1}(\omega) - \frac{5}{3}\beta 2^{X\Omega_2}_{A\Omega_1 v_1}(\omega). \tag{9c}$$

The parameters $\beta 1^{X\frac{1}{2}}_{A\frac{1}{2}v_1} = \beta 1^{X\frac{1}{2}}_{A\frac{3}{2}v_1}$ and $\beta 2^{X\frac{1}{2}}_{A\frac{1}{2}v_1} = \beta 2^{X\frac{1}{2}}_{A\frac{3}{2}v_1}$ can be measured by determining the degrees of linear and circular polarization of fluorescence photons induced by linearly and circularly polarized exciting radiation, as has been done for the Kr atom in Refs. [4–6].

In conclusion, the present work shows that a PWA for photoionization of randomly oriented closed-shells diatomic molecules is possible and how a PWA can be performed by fluorescence polarization experiments. Using the present theoretical background such experiments may be performed in the closest future. The representation of the observed results in terms of partial waves makes their interpretation easier and applies to many branches of physics. The presently developed formalism can be extended to the analysis of multiphoton ionization of molecules by intense electromagnetic fields.

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