Photodouble ionization of water
J. M. Randazzo\textsuperscript{1}, G. Turri\textsuperscript{2}, P. Bolognesi\textsuperscript{3}, C. Penson\textsuperscript{2}, J Mathis\textsuperscript{2}, L. U. Ancarani\textsuperscript{4,5} and L Avaldi\textsuperscript{3}

\textsuperscript{1} Instituto Baileo, CNEA and CONICET, 8400 S. C. de Bariloche, Rio Negro, Argentina.
\textsuperscript{2} Embry-Riddle Aeronautical University, Physical Sciences Department, Daytona Beach, FL, USA.
\textsuperscript{3} CNR-Istituto di Struttura della Materia, Area della Ricerca di Roma 1,0015 Monterotondo Scalo, Italy.
\textsuperscript{4} Université de Lorraine - CERES, LPCIT, 57000 Metz, France.

\textbf{ABSTRACT}

In this work the photo-double ionization (PDI) of water has been investigated. Water has been chosen for its relevance in the study of radiation damage, being the main constituent of bio-systems. The measurements have been performed at the Gas Phase beamline of Elettra at about 20 eV above the double ionization threshold with unequal energy sharing condition. A Generalized Sturmian Function approach has also been applied to describe the two electron continuum, where the structure of the initial state is considered and electronic correlation of the final states exactly calculated. Averaging results over the molecular orientation allow us compare between theory and experimental values.

\textbf{INTRODUCTION}

Double and multiple electron emissions from an atom or a molecule by the absorption of a single photon represent minor processes in the radiation-matter interaction. However, they are of extreme interest, because they are completely determined by electron correlation. The detection in coincidence of the two correlated electrons allows a detailed investigation of the different active mechanisms. A comprehensive understanding of the photon doubleionization, PDI, has been achieved in two-electron systems like the He atom and \textit{H}\textsubscript{2} molecule [1]. In other diatomic and polyatomic molecules experimental studies of the mechanisms of PDI by the measurement of the electron-electron coincidence angular distribution are scarce [2,3]. Here we give the first theoretical approach to model recent measurements of water PDI [4].

\textbf{EXPERIMENTAL SET-UP}

Figure I shows the multicoincidence end station at the Gas Phase photoemission beamline of the Elettra storage to obtain the data presented here.

\textbf{THEORETICAL APPROACH}

Here we will solve a model for the two electron continuum in the PDI of water molecules after single photon absorption, where electron correlation is included in the final state. The initial electronic state of the water molecule is described as the separable products of Moccia orbitals tabulated in [5]. Three dimensional contour plots for the 1\textit{a}, 2\textit{a}, 3\textit{a}, 1\textit{b}, and 1\textit{b}\textsubscript{a} single-electron Moccia orbitals for water are shown below, where atomic \textit{O} and \textit{H} positions are shown in blue and red spheres respectively.

In this way we can refer the two electron dynamics relative to the oxygen position, and decompose the initial states in eigenfunctions of the total angular momentum:

$$\phi_{\ell_1}(r_1)\phi_{\ell_2}(r_2) = \sum_{L_1=|\ell_1|}^{\ell_1}\sum_{M_1=-L_1}^{L_1} D_{L_1M_1}^{L_2M_2}(\alpha, \beta, \gamma) \phi_{L_1M_1}(r_1) \phi_{L_2M_2}(r_2)$$

where:

$$\Phi_{L_1M_1}(r_1, r_2) = \sum_{L_1} H_{L_1M_1}^{L_2M_2}(r_1, r_2) \chi_{L_1M_1}(r_1) \chi_{L_2M_2}(r_2)$$

And where the Wigner D-matrix \textit{D}_{L,M} is introduced to describe the different molecular orientations through the Euler angles \((\alpha, \beta, \gamma)\), and \textit{\chi}_{L,M} are spherical harmonics. After ionization, each electron (\(i = 1, 2\)) interact with molecular orientation averaged potential \([6]-[8]\):

$$U_{\ell,M}(r_1) = \frac{1}{2\pi} \sum_{L,M} \int d\Omega_{\ell,M} \int d\Omega_{\ell,M} \chi_{L,M}(r_1, r_2),$$

where:

$$V_{\ell,M}(r_1, r_2) = \frac{2}{\sin 3\theta} \sum_{n=0}^N \frac{N_{LM}}{r_1 - r_2} + \frac{N_{LM}}{r_1 - r_2} \Delta E_{LM}$$

Scattering wave function which describes ionization reads:

$$\Psi_{\ell,M}(r_1, r_2) = \sum_{L,M} D_{L_1M_1}^{L_2M_2}(\alpha, \beta, \gamma) \phi_{L_1M_1}(r_1) \phi_{L_2M_2}(r_2)$$

where partial wave term follows the driven equation:

$$\sum_{L_1} \frac{1}{2} \nabla_{\ell,M}^2 + U_{\ell,M}(r_1) = \epsilon \sum_{L_1} \nabla_{\ell,M}^2 \phi_{L_1M_1}(r_1, r_2)$$

Even if we have averaged the potential of the residual ion over its orientation, the scattering wave function depends on the orientation through the initial state. Equation (1) is solved through the Generalized Sturmian Method as we have done for helium PDI [7], after which we evaluate the differential cross section:

$$d^2\sigma_1 = \frac{4\pi^2}{\omega c} \left| \left( \Phi_{r_1}, \kappa \right) \Phi_{r_2}, \kappa \right|^2 \omega_{\kappa}$$

(2)

and its average over the Euler angles:

$$d^2\sigma = \frac{4\pi^2}{\omega c} \sum_{L_0} \left( \frac{1}{2L_0} + 1 \right) \sum_{E_{LM}} \sum_{E_{\ell,M}} \left( \Phi_{r_1}, \kappa \right) \Phi_{r_2}, \kappa \right| \omega_{\ell, M}$$

(3)

$$d^2\sigma_{1,2} = \frac{4\pi^2}{\omega c} \left( \Phi_{r_1}, \kappa \right) \Phi_{r_2}, \kappa \right| \omega_{\ell, M}$$

(4)

\textbf{RESULTS}

The binding energy spectrum was first analyzed.

\textbf{SUMMARY}

We have presented TDCS for water double photoionization by single photon absorption, photoelectron coincidence experiments, and the Generalized Sturmian Functions approach. The theoretical method describes the experimental results by considering the Moccia description of the initial electronic states, while taking into account the evolution of the two electron pair in the averaged potential of the residual molecular ion exactly. In average, calculations correctly reproduce the number of peaks, angular distributions, and relative amplitudes in cases. Further improvements by considering the structure of the molecular ion for the ionized electron will be presented in an publication.

\textbf{REFERENCES}


Figure II: The binding energy spectrum of H$_2$O$^{+}\text{+}$ measured in unequal energy sharing conditions \((\theta_1 = \theta_2 = 10\text{ eV})\). The bars indicate the dication states, while the two arrows the binding energies where the two TDCS have been measured.

Measured triple differential cross sections (TDCS), compared with theoretical results for 65 eV and 63 eV energy Figures III and IV respectively.

Figure III: TDCS at 65 eV energy, at unequal energy shearing regime. The angle of one of the electrons is fixed at \(\theta_1 = 0\text{o}, 30\text{o}\) and 60\text{o}.

Figure IV: Same as Figure III but for 63 eV energy.