Complete photoionization experiment and autoionizing states in Ne II.

Paolo A. Carpeggiani[1,2,], Elena Gryzlova[3], Maurizio. Reduzzi[1,2], Antoine Dubrouil[1], Davide Faccialà[1,2], Matteo Negro[1,2], Kyoshi Ueda[4], Svetlana I. Strakhova[3], Fabio Frassetto[5], Frank Stienkemeier[6], Yevheniy Ovcharenko[7], Micheal Meyer[8], Oksana Plekan[9], Paola Finetti[9], Kevin Prince[9], Carlo Callegari[9], Alexi N. Grum-Grzhimailo[3] and Giuseppe Sansone[1,2,10]*

(1) Dipartimento di Fisica, Politecnico Piazza Leonardo da Vinci 32, 20133 Milano Italy. (2) IFN-CNR Politecnico, Piazza Leonardo da Vinci, 32, 20133 Milan, Italy. (3) Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Moscow, 119991 Russia. (4) Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan. (5) IFN-CNR via Trasea 7, 35131 Padova, Italy. (6) Physikalisches Institut, Universität Freiburg, 79106 Freiburg, Germany. (7) Institut für Optik und Atomare Physik, TU Berlin, Berlin, Germany. (8) European XFEL, Albert-Einstein-Ring 19, 22761 Hamburg, Germany. (9) Elettra-Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy. (10) Physikalisches Institut der Albert-Ludwigs-Universität Stefan-Meier-Str. 19 79104 Freiburg, Germany

**Current: Institut für Photonik, TU Wien, Gußhausstraße 27-29, 1040 Wien.*

The interaction of intense extreme ultraviolet (XUV) pulses with an atom can lead to multi-photon absorption and multiple ionization of the target. By using intense, linearly and circularly polarized XUV pulses of the Free Electron Laser (FEL) FERMI [1], we realize the first experimental demonstration of a quantum mechanically complete experiment (CE) in an ionic system. The quest for a CE, as the strongest test of theory and as a source of minimum information from which any observable can be predicted, was first formulated at the end of the 1960s, [2], for electron-atom scattering, and then extended to atomic and molecular photoionization (PI). The complete information cannot be obtained by measuring only the photo-electron angular distribution (PAD) and the cross sections, but additional variables are required. Thus, CEs in atomic PI were realized by measuring the angle-resolved PAD, combined with the observation of other variables or, in case of atoms with open shells, by controlling the initial polarization of the target. All the CEs on PI so far have been performed with neutral targets. As atomic PI leads to polarized residual ions, CE in positive ions can be realized within the two photon double sequential ionization (TPDI) process, now available thanks to the intense XUV pulses of FELs.

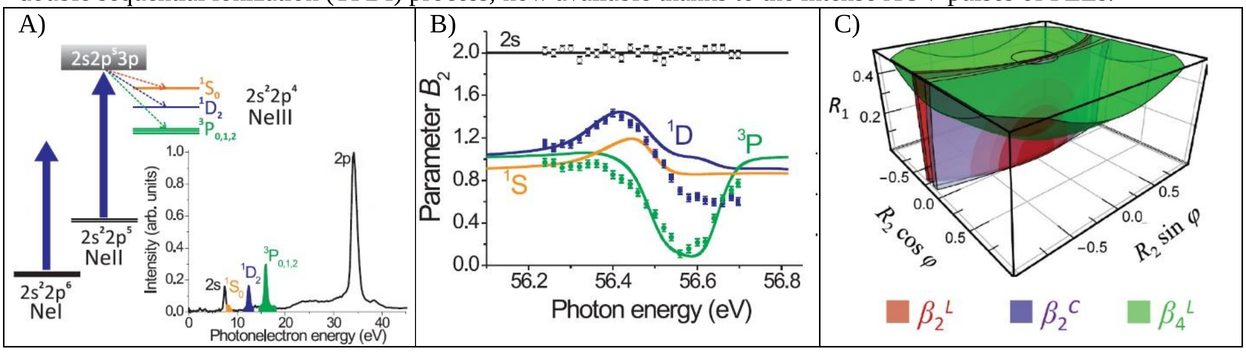

Fig. 1 a) Energy level scheme for singly and doubly ionized neon and photoelectron spectrum. The three final ionic states for the double ionized neon $^3P_{0,1,2}$, $^1D_2$, $^1S_0$ and the corresponding photoelectron peaks are indicated by green, blue and orange, respectively. B) Experimental, squares, and calculated, lines, β_2 parameter around the resonance of the auto-ionizing states for the linear case. Colors as in previous figure. C) Complete experiment on PI of Ne^+ $^2p^5$ ^2P to the Ne^{++} $^2p^4$ ^3P state at the photon energy 53eV. Allowed sets of parameters *R1, R2cosφ, R2senφ* extracted from the values of $\beta_2(C)$ (blue), $\beta_2(L)$ (red), $\beta_4(L)$ (green) for the ^3P peak.

PADs were acquired with a velocity map imaging spectrometer (VMI) for photon energies between 40 and 60 eV, and with a fine scan in the 56.2-56.8eV range, corresponding to the energy of the auto-ionizing states of Ne II (Fig. 1a). In a 2-photon process, PADs can be described on the base of the Legendre polynomials by the relation: $I(\theta) = I_0/4\pi[1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)]$, where θ is the angle between the axis of symmetry and the emission direction of the photoelectrons. The validity of the theoretical model is verified by the agreement with the β_N parameters (Fig. 1b), and the ratio of ionization of the different peaks. In the Cooper-Zare model, [3], the dynamics of each PI step is described by two absolute values of the single electron ($2p$-εs and $2p$-εd) ionization amplitudes $d_{L=s,d}^{(j=1,2)} = |d_L^{(j)}|\exp(i\varphi_L^{(j)})$ and their relative phase $\varphi^{(j)} = \varphi_s^{(j)} - \varphi_d^{(j)}$. The polarization of Ne^+ is described by $R_1 = |d_s^1/d_d^1|$, and the PAD of the second PI by $R_2 = |d_s^{(2)}/d_d^{(2)}|$ and $\varphi^{(2)} = \varphi_s^{(2)} - \varphi_d^{(2)}$. These parameters can be linked to the experimental data by the β_2 and β_4, coefficients for linear and circular polarized cases, thus determining a restricted allowed space of the PI amplitudes (Fig. 1c). The intersection of the allowed spaces defines the solution. It is finite due to the error bars, and would restrict to a point in case of absolutely precise measurements.

References

[1]E. Allaria, et al. Highlz coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet. Nature Photonics. 6, 699-704 (2012).

[2] B. Bederson. The "Perfect" scattering experiment. I Comm. Atom. Mol. Phys, 1, 41 (1969).

[3] J. Cooper, R.N. Zare. Angular distribution of Photoelectrons. J. Chem. Phys. 48, 942-943 (1968).