Near-threshold photoelectron angular distributions from two-photon resonant ionisation of He and Ne atoms

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Abstract. We present an overview of two recent experiments in which photoelectron angular distributions from light-polarised He and Ne atoms are studied, with the emphasis on theoretical description of photoionisation. Polarised He (Ne) atoms are prepared in the 1s4p, 1s5p, or 1s6p (2p53d) states with linearly polarised synchrotron light and ionised with a tunable infrared laser. The asymmetry parameters extracted from the measured velocity map imaging spectra agree well with the calculated asymmetry parameters.

1. Introduction
Photoionisation of polarised atoms has proven to be a useful technique for probing details regarding photoionisation dynamics. This point was noted by Klar and Kleinpoppen [1] who pointed out that measurements of photoelectron angular distributions (PADs) from polarised atomic targets can provide “complete” information on the photoionisation process (the so called complete experiment). Of particular interest to us here are PADs from noble gas atoms prepared in polarised excited states for low kinetic energies of the photoelectrons (see [2] for a recent review). A way of preparing atoms in polarised atomic states is by photoexcitation with linearly polarised light. These polarised excited atoms are then ionised by absorbing the second photon – either from the same or from a different radiation source.

Here we review two recent experiments [3, 4], in which photoelectron angular distributions from light-polarised He and Ne atoms are measured, and present a theoretical framework for their description. We study angular distributions of photoelectrons ejected in two-photon ionisation (figure [1] of ground-state helium and neon atoms by simultaneous irradiation with synchrotron photons (linearly polarised, polarisation $\hat{e}_1$, energy $\omega_1$) and Ti:Sapphire laser photons (polarisation $\hat{e}_2 = \hat{e}_1$, energy $\omega_2$):

$$|g\rangle \xrightarrow{\omega_1} |\nu\rangle \xrightarrow{\omega_2} |k; \gamma\rangle,$$

(1)

where $|g\rangle$ denotes the ground state, $|\nu\rangle$ the chosen intermediate state, and $|k; \gamma\rangle$ the final continuum state. With $k$ we denote the wave vector of the photoelectron and with $\gamma$ additional...
quantum numbers of the final state. The energy of the synchrotron photons is chosen to lie on resonance with the intermediate state \((\omega_1 = E_\nu - E_g)\).

In the first experiment, we examine PADs from the ground state helium atoms

\[
\text{He } 1s^2 \overset{\omega_1}{\rightarrow} \text{He } 1snp \overset{\omega_2}{\rightarrow} \text{He}^+ 1s + e^-,
\]

where the singlet 1snp intermediate states with \(4 \leq n \leq 6\) are considered. We use LS coupling scheme for the description of bound and continuum states.

In the second experiment, we examine photoelectron angular distributions from the ground state neon atoms:

\[
\begin{align*}
\text{Ne } 2p^6 & \overset{\omega_1}{\rightarrow} \text{Ne } 2p^5 (2P_{1/2}) 3d \overset{\omega_2}{\rightarrow} \text{Ne}^+ (2P_{1/2}) + e^-, \\
\text{Ne } 2p^6 & \overset{\omega_1}{\rightarrow} \text{Ne } 2p^5 (2P_{3/2}) 3d \overset{\omega_2}{\rightarrow} \text{Ne}^+ (2P_{3/2}) + e^-,
\end{align*}
\]

where \(jK\) coupling notation \([5]\) is used for the intermediate \(2p^53d\) states with the angular momentum of the core (2p) electrons equal to \(J_c = 1/2\) and \(J_c = 3/2\) and the total angular momentum \(J = 1\), as described by (3a) and (3b), respectively.

Figure 1. Resonant two-photon ionisation of an atom in the ground state \(|g\rangle\). The intermediate state is denoted by \(|\nu\rangle\), and the energy of the first and the second photon with \(\omega_1\) and \(\omega_2\), respectively.

It has been assumed in (2), (3a), and (3b) that the dipole approximation is valid. Higher-order transition amplitudes (M1, E2, etc.) are expected to be negligible in comparison with the dipole amplitude in the first and in the second step for both He and Ne. Since, in addition, \(\hat{e}_1 = \hat{e}_2\) holds, the PADs are axially symmetric, and the differential cross section has the form

\[
\frac{d\sigma^{(2)}}{d\Omega} = \frac{\sigma^{(2)}_{\text{int}}}{4\pi} \left(1 + \beta_2 P_2(\cos \theta) + \beta_4 P_4(\cos \theta)\right),
\]

where \(\cos \theta = \hat{k} \cdot \hat{e}_1 = \hat{k} \cdot \hat{e}_2\), \(P_q\) denotes Lagrange polynomials of order \(q\), \(\sigma^{(2)}_{\text{int}}\) is the angle integrated generalised two-photon ionisation cross section, and \(\beta_2\) and \(\beta_4\) are the asymmetry parameters.

Both experiments were performed at the Gas Phase Photoemission beamline at Elettra synchrotron, Italy \([6]\). The details of the experimental setup are described in \([3, 7, 8]\) and the references therein. In short, the monochromatised synchrotron light is focused into the interaction region of a velocity map imaging (VMI) spectrometer, where it is aligned with a counter-propagating Ti:Sapphire laser beam tunable in the wavelength range of 670–1000 nm. For Ne, the laser can be used in a continuous-wave (CW) mode since the lifetime of the intermediate states is relatively long, and in the case of He, mode locking is used (pulse duration of 15 ps, 83.3 MHz repetition rate). The acquired images are inverted using the pBasex method \([9]\), which is used to reconstruct the original 3-dimensional distributions from the recorded 2-dimensional projections.
2. Photoelectron angular distributions

2.1. Helium

The measured electron yield is proportional to the generalised two-photon ionisation cross section (e.g., see [3]). Since the first step of the two-photon excitation is resonant, the generalised ionisation cross section can be written as:

\[
\frac{d\sigma^{(2)}}{d\Omega} \propto \sum_{\mu, \mu_s, m_s} |\langle \mathbf{k}; \lambda, \mu, \mu_s, m_s | D(e_2) | \nu \rangle|^2,
\]

(5)

where \(\lambda, \mu, \) and \(\mu_s\) are the angular momentum of the core electron, its projection on the \(z\) axis, and the projection of its spin, respectively, and \(m_s\) is the spin projection of the continuum electron. The dipole transition operator which describes the absorption of the infrared (IR) photon is denoted by \(D(e_2)\).

In \(LS\) coupling, the intermediate states are described well within the single-configuration approximation even for the lowest-lying member \((n = 2)\) of the \(1s\) series [10]. We may therefore use the uncoupling formula [5] to uncouple the “inner” electron part and the “outer” electron part of the wave function. This results in a relatively simple parametrisation for \(\beta_2\) and \(\beta_4\):

\[
\beta_2 = \frac{40 - 140X \cos \Delta}{28 + 35X^2}, \quad (6a)
\]

\[
\beta_4 = \frac{72}{28 + 35X^2}, \quad (6b)
\]

for the radial parts of the single-electron dipole matrix elements. It should be noted that unlike \(X\), phase difference \(\Delta\) is independent of the choice of the intermediate state. The notation used in \([6a]\) and \([6b]\) is the same as in [11].

Real configuration-interaction coupled Coulomb-Sturmian basis functions [12] have been used to calculate the bound and continuum states and the relevant dipole matrix elements. The values of \(X(\epsilon)\) just above the ionisation threshold have been checked to agree with the values obtained from extrapolated density of oscillator strength of bound \(1s\) states and \(1s\) states. The calculated ratios for \(n = 2\) and \(n = 3\) agree well with ratios extracted from the partial single-photon ionisation cross sections given in [13] for \(\epsilon \geq 0.5\) eV (figure 2). Near the threshold, the extracted values deviate slightly from our results, but the ratios obtained from the cross sections given in [14] for \(2 \leq n \leq 5\) – using the same approach as in [13] – which focus on the near-threshold region \((\epsilon \lesssim 0.11\) eV) may be seen to match the present calculations. Our measured and calculated values of \(X\) are shown in figure [3] and are seen to agree reasonably well for the \(1s\) and \(1s\) states. Furthermore, the measurements of Haber et al [11] are in good agreement with the theory for the \(1s\) state (filled triangles), whereas there is a minor discrepancy for the lower of the two points for the \(1s\) state (filled squares), for which the calculated value lies outside the experimental error bars.
The phase differences between the $1s\epsilon_s$ and $1s\epsilon_d$ waves have been obtained by extrapolating the energy dependence of the quantum defects $\kappa_0$ and $\kappa_2$ of the $1sn's$ and $1sn'd$ bound states across the threshold. The difference $\pi(\kappa_0 - \kappa_2)$ has been found to be negligible in comparison with the difference between the Coulomb shifts $\sigma_0 - \sigma_2$ in the investigated energy region. The

dependence of the phase difference $\Delta$ between the outgoing $1s\epsilon$s and $1s\epsilon$d waves on the kinetic energy of the photoelectron is shown in figure 4. The calculations are in good agreement with our experimental results and with the measurements presented in [11].

The asymmetry parameters close to the threshold do not depend strongly on the principal quantum number $n$ of the intermediate states considered here. While $\beta_2$ increases slightly with $n$ above the threshold ($\epsilon \lesssim 2$ eV), $\beta_4$ decreases with $n$ in this energy region. An example of a typical energy dependence of the asymmetry parameters is shown in figure 5 for the $1s5p$ intermediate state. Similar plots have been obtained also for the $1s4p$ and $1s6p$ states.
2.2. Neon

As for helium, the measured electron yields for two-photon ionisation of ground-state neon atoms are proportional to \[3\]:

\[
\frac{d\sigma^{(2)}}{d\Omega} \propto \sum_{M_c, m_s} |\langle \mathbf{k}; J_c, M_c, m_s | D(\hat{e}_2) | \nu \rangle|^2,
\]

(8)

where \(J_c\) is the angular momentum of the 2p electrons, \(M_c\) its projection on the \(z\) axis, and \(m_s\) the spin projection of the continuum electron. In the two sets of the measured PADs, the energy of the synchrotron photons is chosen to lie on resonance with the \(J_c = 1/2\) 2p\(^5\)3d \([3/2]_1\) state \(3a\) or the \(J_c = 3/2\) 2p\(^5\)3d \([3/2]_1\) state \(3b\). Calculations show that the intermediate states are described well within a single configuration approximation if \(jK\) coupling is used: \(K = J_c + \ell\), \(J = K + s\), where \(J\), \(\ell\), and \(s\) are the total angular momentum, orbital angular momentum of the continuum electron, and its spin, respectively.

The energy region of the final 2p\(^5\) continuum states consists of two sub-regions. The first sub-region, which lies between the 2p\(_{3/2}\) and 2p\(_{1/2}\) thresholds and is referred to as the autoionisation region, contains resonances which consist of \(J_c = 1/2\) discrete components (closed channels or configurations) mixed with \(J_c = 3/2\) continuum components (open channels). In the second sub-region, which lies above the 2p\(_{1/2}\) threshold, all the 2p\(^5\) channels are open. In our experiment, the energy of the IR photon is tuned to avoid double-resonance excitation in the first sub-region. This is possible since unlike the 2p\(^\ell\)ed resonances accessible with single-photon ionisation \([13]\), the accessible 2p\(^\ell\)ef and 2p\(^\ell\)ef resonances are narrow \([7]\). This fact allows for a simpler theoretical treatment in which the coupling between the \(J_c = 1/2\) and \(J_c = 3/2\) channels can be neglected. This has already been assumed in \([8]\), where it has been taken that the final state has a well defined momentum \(J_c\).

For convenience, we also use \(jK\) coupling to describe the final continuum states. The accessible final state with \(J_c = 3/2\) is written as a linear combination of the \(\epsilon\ell [K]J\), where 2p\(^5\) has been omitted. These channel functions will be denoted with indices \(\mathbf{A}\), \(\mathbf{B}\), \(\mathbf{C}\), \(\mathbf{G}\), and \(\mathbf{H}\), respectively. The \(J_c = 1/2\) state is written in terms of the \(\epsilon\ell [3/2]_1\), \(\epsilon\ell [1/2]_0\), and \(\epsilon\ell [5/2]_2\) channel wave functions, which we will denote with indices \(\mathbf{D}\), \(\mathbf{E}\), and \(\mathbf{F}\), respectively. With each of these channel functions, a dipole amplitude and a phase shift are associated. This results in a double set of expressions for \(\beta_2\) and \(\beta_4\), one for each of the two values of \(J_c\). The asymmetry parameters are written as quotients \(\beta_2 = A_2/A_0\) and \(\beta_4 = A_4/A_0\) \([2]\), where for \(J_c = 1/2\)

\[
A_0 = Q_{DD} + Q_{EE} + Q_{HH},
\]

(9a)

\[
A_2 = \frac{1}{4} Q_{DD} + \frac{\sqrt{2}}{2} Q_{EE} - \frac{\sqrt{6}}{14} Q_{HH} - \frac{\sqrt{3}}{2} Q_{HH} + \frac{2}{7} Q_{HH},
\]

(9b)

\[
A_4 = \frac{3}{14} Q_{HH} - \frac{3\sqrt{6}}{7} Q_{HH},
\]

(9c)

and for \(J_c = 3/2\)

\[
A_0 = Q_{AA} + Q_{BB} + Q_{CC} + Q_{GG} + Q_{EE},
\]

(10a)

\[
A_2 = \frac{1}{5} Q_{AA} + \frac{3}{10} Q_{BB} - \frac{3\sqrt{5}}{10} Q_{CC} + \frac{3}{70} Q_{GG} + \frac{3\sqrt{6}}{35} Q_{EE} - \frac{1}{5} Q_{BB} - \frac{\sqrt{5}}{10} Q_{EE} + \frac{30}{35} Q_{EE} + \frac{3\sqrt{6}}{35} Q_{EE} + \frac{11}{70} Q_{EE},
\]

(10b)

\[
A_4 = \frac{6}{7} Q_{GG} + \frac{3\sqrt{6}}{14} Q_{EE} + \frac{3\sqrt{6}}{14} Q_{EE} + \frac{3\sqrt{6}}{14} Q_{EE} - \frac{3}{28} Q_{EE}.
\]

(10c)
We have introduced $\Omega_{\ell\ell'} = D_\ell D_{\ell'} \cos(\pi \kappa_\ell - \pi \kappa_{\ell'} + \sigma_\ell - \sigma_{\ell'})$, where $\xi$ and $\xi'$ stand for the channel indices ($A$, $B$, etc.), $D_\ell$ and $D_{\ell'}$ for the real dipole transition amplitudes, $\pi \kappa_\ell$ and $\pi \kappa_{\ell'}$ for the short-range phase shifts, and $\sigma_\ell$ and $\sigma_{\ell'}$ for the Coulomb phase shifts. Since the excess energy considered here is low ($\epsilon \lesssim 100 \text{ meV}$), the dipole matrix elements and phase shifts are obtained by extrapolating density of oscillator strength and quantum defects of the $2p^5n'p$ and $2p^5n'f$ states across the ionisation thresholds. The reader is referred to [3] for details.

An approximation often used for parametrisation of photoionisation cross sections is to neglect the spin-orbit interaction in the continuum and to assume that the shapes of the orbitals and the phase differences do not depend on the total angular momentum $J$ of continuum channels (e.g., see [16]). If this is assumed in our case, the core-electron (“inner”) part and the asymptotic (“outer”) part of the wave function can be uncoupled. Simplified expressions are obtained if it is assumed that the phase shifts and the single-electron dipole matrix elements of various channels depend only on the orbital angular momentum $\ell$ of the continuum electron for a fixed intermediate state [3]. In the present case, however, this approach gives incorrect asymmetry parameters for $J_c = 3/2$ because the value of $\beta_4$ is constrained to zero. This means that in order to describe the PADs correctly, the dependence of the transition matrix elements on the channel quantum numbers should be considered.

The energy dependence of the asymmetry parameters given by (9a)–(10c) is shown in figure 6. An important observation is that the asymmetry parameters vary smoothly with $\epsilon$ also in the autoionisation region ($J_c = 3/2$ results). As can be seen, the calculated $\beta_4$ values for $J_c = 3/2$ are in very good agreement with the experiment. On the other hand, the values of $\beta_2$ extracted from the measured PADs deviate systematically from the calculated $\beta_2$ for $J_c = 1/2$. Nevertheless, generally speaking, the experimental and the calculated values agree well, especially for the angular momentum $J_c = 3/2$.

3. Conclusions

We have presented analyses of photoelectron angular distributions from polarised He and Ne atoms. These atoms are prepared in polarised excited states with linearly polarised synchrotron light and then ionised with a tunable Ti:Sapphire laser. We have considered photoionisation of He atoms in the singlet $1snp$ states with $4 \leq n \leq 6$ and Ne atoms in the $2p^5 \, (2P_J) \, 3d \ [3/2]_1$ states with $J_c = 1/2$ or $J_c = 3/2$.

It has been shown that for He, a simpler parametrisation of the asymmetry parameters may be used, where $\beta_2$ and $\beta_4$ are written in terms of the ratio of the single-electron dipole matrix elements of the accessible $1se$ and $1sd$ partial waves and the difference of the corresponding phase shifts. The parameters extracted from the measured data have been found to be in good agreement with our calculations.
For Ne, the measured and calculated asymmetry parameters agree reasonably well. It has been shown that in this case, it is not possible to reproduce the measured asymmetry parameters for the \(2p^5 (^{2}P_{3/2}) 3d \left[\frac{3}{2}\right]_1\) intermediate state if the spin-orbit interaction in the continuum is neglected and it is assumed that the single-electron dipole transition amplitudes and phase differences are independent of the total angular momentum of the channel wave functions.

Acknowledgments
This research was partly supported by the Italy-Slovenia joint research project “Dynamics at nanoscale” and by PRIN 2009W2W4YF and 2009SLKFEX. A. M. and M. Ž. acknowledge the support of the Slovenian Research Agency (research programme P1-0112).

References
[1] Klar H and Kleinpoppen H 1982 J. Phys. B 15 933–950
[2] Sukhorukov V L, Petrov I D, Schäfer M, Merkt F, Ruf M W and Hotop H 2012 J. Phys. B 45 092001
[3] O’Keeffe P, Bolognesi P, Mihelič A, Moise A, Richter R, Cautero G, Stebel L, Sergo R, Pravica L, Ovcharenko E, Decleva P and Avaldi L 2010 Phys. Rev. A 82 052522
[4] O’Keeffe P, Mihelič A, Bolognesi P, Žitnik M, Moise A, Richter R and Avaldi L 2012 Submitted to New J. Phys.
[5] Cowan R D 1981 The theory of atomic structure and spectra (Berkeley: University of California)
[6] Blyth R R et al 1999 J. Electron Spectrosc. Relat. Phenom. 101-103 959–64
[7] Moise A, Alagia M, Banchi L, Ferianis M, Prince K C and Richter R 2008 Nucl. Inst. Methods Phys. Res. A 588 502–8
[8] O’Keeffe P, Bolognesi P, Coreno M, Moise A, Richter R, Cautero G, Stebel L, Sergo R, Pravica L, Ovcharenko Y and Avaldi L 2011 Rev. Sci. Instrum. 82 033109
[9] Garcia G A, Nahon L and Powis I 2004 Rev. Sci. Instr. 75 4989–96
[10] Froese Fischer C 1977 The Hartree-Fock method for atoms (New York: John Wiley & Sons)
[11] Haber L H, Doughty B and Leone S R 2009 Phys. Rev. A 79 031401(R)
[12] Lagmago Kamta G, Piraux B and Scrinzi A 2001 Phys. Rev. A 63 040502(R)
[13] Chang T N and Fang T K 1995 Phys. Rev. A 52 2638–44
[14] Chang T N and Fang T K 1995 Phys. Rev. A 52 2052–56
[15] Chan W F, Cooper G, Guo X and Brion C E 1992 Phys. Rev. A 45(3) 1420–33
[16] Schohl S, Cherepkov N A, Petrov I D, Sukhorukov V L, Baier S and Hotop H 1998 J. Phys. B 31 3363–85