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A collinear angle-resolved photoelectron spectrometer

O. Windelius [a,c,*], A. Aguilar [b], R.C. Bilodeau [b,d], A.M. Juarez [e], I. Rebolledo-Salgado [e], D.J. Pegg [f], J. Rohlén [c], T. Castel [g], J. Welander [c], D. Hanstorp [c]

[a] Department of Physics, Chalmers University of Technology, SE-41296 Gothenburg, Sweden
[b] Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[c] Department of Physics, University of Gothenburg, SE-41296 Gothenburg, Sweden
[d] Department of Physics, University of Connecticut, Storrs, CT 06269, USA
[e] Instituto de Ciencias Fisicas, Universidad Nacional Autonoma de Mexico, PO Box 48-3, Cuernavaca 62251, Mor, Mexico
[f] Department of Physics, University of Tennessee, Knoxville, TN 37996, USA
[g] École Nationale Supérieure d'Ingénieurs de Caen, ENSICAEN, 6 Boulevard Maréchal Juin, 14050 Caen, France

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ABSTRACT

In the present paper we describe a newly designed collinear photoelectron spectrometer for angular distribution measurements. We will henceforth refer to this instrument by the acronym **PEARLS** (**P**hoto**E**lectron **A**ngle-**R**esolved **L**inear **S**pectrometer). The design was motivated by the desire to collect electrons emitted from an extended linear source consisting of collinear photon and ion beams at a synchrotron radiation site. The electrons could be produced in either photoionization or photodetachment events. The primary advantage of a collinear beams geometry is that the effective interaction volume can be made much larger than that obtainable with a crossed beams geometry, which has been used in many earlier photoelectron spectroscopic studies. The present apparatus is capable of collecting electrons over a beam source length of 22 cm. The electrons are detected using Channel Electron Multipliers (CEMs). There are 4 detector planes placed perpendicular to the direction of the beam source, where each plane contains 4 CEMs. The use of all 4 detector planes with a total of 16 CEMs enhances the photoelectron signal, which is important at a synchrotron radiation site where the photon flux is typically low. If photoelectrons of different energies are emitted, the design allows for electrostatic energy analyzers to be placed in front of the CEMs. We have performed a photodetachment experiment to demonstrate the functionality of the **PEARLS** apparatus using a pulsed laser as the photon source. In particular, we have measured the angular distribution of photoelectrons ejected from Ag^- at two different photon energies.

1. Introduction

For many years, photoelectron spectroscopy has provided valuable information on the properties of atomic and molecular systems via the processes of photoionization and photodetachment [1–3]. In the present paper we will focus on the photodetachment of negative ions. The general lack of excited states with allowed dipole transitions prohibits conventional bound–bound state photon absorption spectroscopy. Essentially all information on the structure and dynamics of negative ions has been provided by the bound-free photodetachment process [3–5].

In photodetachment the energy and angular momentum of a photon is transferred to a negative ion, which subsequently breaks up into a neutral atom and a free electron. The initial energy and angular momentum is conserved and shared in the final state by the residual atom and the free electron. A known fraction of the energy transfer appears as kinetic energy in the free electron. Thus, measurements of photoelectron energies allows one to determine the binding energy of the electron in the negative ion prior to detachment [6,7]. The angular momentum transfer is manifested in the angular distribution of the detached photoelectrons [8,9]. The shape of a photoelectron angular distribution is characterized by an asymmetry parameter, β. Measurements of the photon-energy dependence of asymmetry parameters can provide information on the relative amplitudes and phases of the partial waves representing the detached electron in the final state.

Essentially all experimental investigations of photodetachment are accelerator based and have been conducted using mass-selected beams of negative ions that are essentially uni-

* Corresponding author at: Department of Physics, Chalmers University of Technology, SE-41296 Gothenburg, Sweden.
E-mail address: olle.windelius@donnergymnasiet.se (O. Windelius).

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directional and mono-energetic. Most studies to date have employed conventional lasers as the photon source. Such sources are able to provide photons over a wide range of wavelengths from the infrared to the ultraviolet. Free-electron lasers and synchrotron radiation sources are available for measurements requiring photon energies outside the range of conventional lasers [10,11]. The interaction geometry used to mate the ion and laser beams is chosen according to the type of experiment under consideration. For example, essentially all photoelectron angular distribution measurements have employed a crossed beams geometry in which a negative ion beam is crossed perpendicularly with a linearly polarized laser beam. Energy- and angle-resolved measurements can be made using various types of electron spectrometers [12,13]. The well-defined spatial interaction volume allows one to efficiently collect and detect photoelectrons. However, the small interaction volume associated with a crossed beams geometry results in a low rate of production of photoelectrons. Clearly, one could substantially increase the production rate by merging the laser and ion beams in a collinear interaction geometry. This advantage, however, is offset by the difficulty of collecting electrons from a relatively long linear source. The first attempt to solve the problem was made by Hanstorp et al. [14], who surrounded a collinear ion-laser interaction region with a cylindrical graphite tube. A line of holes were drilled along the length of the tube in a direction perpendicular to the axis of the collinear source. Photoelectrons ejected from the source passed through these holes on their way to a detector. Angular distributions were measured by rotating the polarization vector of the laser [15].

In the present paper we describe a new apparatus, **PEARLS** (**P**hoto**E**lectron **A**ngle-**R**esolved **L**inear **S**pectrometer), designed to study angular distributions of photoelectrons emitted from the collinear interaction of beams of positive or negative ions with a photon beam from a synchrotron radiation source. **PEARLS** was designed to permit angle-resolved measurements without the need to rotate the polarization vector of the synchrotron radiation (a difficult procedure usually leading to a substantial loss of flux). To our knowledge, the only previously reported measurement of an angular distribution involving a collinear source and synchrotron radiation was that of Al Moussalami et al. [16,17]. In this photoionization experiment the ejected electrons were collected from a relatively small volume within the merged beams source. It therefore suffered from a low production rate, similar to that in a crossed beams experiment.

The testing of the functionality of **PEARLS** at a synchrotron radiation facility was impractical due to the high demand for beamtime at such sites. Instead, we conducted offline tests of the ability of **PEARLS** to be used in angular distribution measurements by using lasers as the photon source. The test experiments, which were performed at Gothenburg University, involved the photodetachment of Ag^- and P^-. Negative ions were chosen for the commissioning of **PEARLS** since their small binding energies allow photoelectrons to be produced using visible laser sources available in our laboratory. However, the design will allow angular resolved photoelectron studies of both negative and positive ions of atoms, molecules, clusters as well as larger biomolecules.

2. Photoelectron angular distributions

The differential cross section for photodetachment, $\frac{d\sigma}{d\Omega}$, can be written, under the assumption that the target is unpolarized (as is the case for an ion beam) and within the dipole approximation, as [8]

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi}(1 + \beta P_2(\cos\theta)). \quad (1)$$

Here, σ is the total cross section and θ is the angle between the direction of the linear polarization of the photons and the momentum of the outgoing electrons. The angular part of the equation contains $P_2(\cos\theta)$, the second-order Legendre polynomial, and the asymmetry parameter β, which completely describes the angular distribution of the photoelectrons. The asymmetry parameter contains information on the relative amplitudes and phases of the partial waves that represent the free electron in the final state. These waves carry orbital angular momenta that differ from that of the bound electron as a result of absorbing a photon. In the case of an electric dipole transition, the change Δl in orbital angular momentum of the detached electron is $|\Delta l| = 1$. Thus, if an s-orbital electron is detached from a negative ion, the free electron will be represented by a pure p-wave. If, however, a p-orbital electron is detached, it will be represented by a superposition of s- and d-waves. Close to the detachment threshold the d-wave is suppressed due to the centrifugal barrier and so the s-wave dominates.

According to Eq. (1), the number of electrons emitted in any direction is proportional to the angular factor $1 + \beta P_2(\cos\theta)$. Therefore, in principle, the asymmetry parameter β can be determined by comparing the number of counts, C, registered in two orthogonal detectors placed parallel to and perpendicular to the direction of the polarization vector of the photons. However, if the ions are moving, one must consider transformations between the Ion Frame (IF) and the Lab Frame (LF). For example, in order for detached electrons to be emitted and detected in the LF at 90° with respect to the ion beam direction, they must be emitted in a backward direction in the IF (see Fig. 1). The angle, α, between the electron momenta in the two frames of reference is a function of the magnitudes of the ion velocity and the velocities of the electron in the IF and LF. This kinematic effect becomes decreasingly important as the angle θ between the directions of the detector and the polarization vector increases. In the limit of $\theta = 90°$, it vanishes altogether (See Fig. 1b).

One way to compensate for this frame transformation effect is to simulate the photoelectron angular distribution experiment using certain values of β as input, and compare the simulation output with the experimental results. It is reasonable to assume that the LF differential cross section to first order has the approximate form

$$\frac{d\tilde{\sigma}}{d\Omega} = \frac{\tilde{\sigma}}{4\pi}(1 + \tilde{\beta} P_2(\cos\psi)), \quad (2)$$

where $\tilde{\sigma}, \tilde{\beta}$ and the angle ψ are measured in the LF. Then, by setting $\psi = 0°$ and $\psi = 90°$ in Eq. (2) we obtain

$$C_{\parallel} \propto 1 + \tilde{\beta} P_2(1) = 1 + \tilde{\beta},$$

and

$$C_{\perp} \propto 1 + \tilde{\beta} P_2(0) = 1 - \frac{1}{2}\tilde{\beta},$$

respectively. If we then define

$$\tilde{Q} = \frac{C_{\perp}}{C_{\parallel}} \quad (3)$$

we get

$$\tilde{\beta} = \frac{1 - \tilde{Q}}{\tilde{Q} + \frac{1}{2}}. \quad (4)$$

The $\tilde{\beta}$ thus retrieved from the simulation can then be compared with the measured value.