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Electron trajectory simulations of time-of-flight spectrometers for core level high-energy photoelectron spectroscopy at pulsed X-ray sources

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ABSTRACT

The advent of Free Electron Lasers (FELs), able to provide short (2–100 fs) and intense (10^{33} photons/s/mm^2/mrad^2/0.1%bandwidth) pulses of light also in the hard X-ray regime ($\hbar\omega$> 2000 eV), opens new possibilities to study the ultrafast dynamics of processes, exploiting the capability of Hard X-ray Photoelectron Spectroscopy (HAXPES) to measure core-level spectra of elements with bulk sensitivity. In order to detect the intense bursts of high kinetic energy electrons generated by the X-ray pulses with an energy resolution comparable to the existing category of electron analyzers, a new class of spectrometers must be designed. We present a characterization of two different TOF spectrometers, namely one based on a retarding cylindrical lens and another one based on the spherical reflector geometry. SIMION® software has been used in order to evaluate electron trajectories of high kinetic energy electrons (5000–10,000 eV) and extract transmission properties, angular acceptance and energy resolution. It resulted that while the linear system is able to accept a larger solid angle (~50 msr), the spherical mirror offers a better resolving power (around 71,000). Both analyzers are capable of a transmission above 90% within range of kinetic energies wide enough to measure the full line-shape of a core photoionization peak. Furthermore, we proved that both instruments are able to discriminate between two consecutive electron bunches having a temporal separation inferior than 220 ns, which is the distance between two consecutive photon pulses at the European X-ray Free Electron Laser (EXFEL), which is currently under construction in Hamburg.

1. Introduction

Progress in the design and realization of new types of electron spectrometers has allowed to make significant steps forward in many aspects of solid state physics. For example, techniques like photoelectron spectroscopy (PES) [1] have taken full advantage of the development of systems like hemispherical deflector analyzers (HDA) over the course of the years. More recently, the quest for increasing bulk sensitivity has led to the advent of so-called hard X-Ray PES (HAXPES), where the electron energy to be detected is sensibly increased with respect to standard PES to values ranging from 5 to 10 keV [2]. As a consequence, the existing class of electron analyzers had to be upgraded [3,4] in order to obtain bulk sensitive spectra with statistics and energy resolution comparable with standard PES experiments [5].

On the other hand, performing PES experiments at pulsed X-ray sources, such as X-Ray Free Electron Lasers (XFELs), able to convey pulses with a duration <1 ps, it is possible to extract dynamical information at time scales typical of electronic processes [6]. Time-of-flight (TOF) spectrometers seem to be naturally suited for use at pulsed sources, as they intrinsically require a time reference. The working principle rests on measuring the flight time t of particles between a target and a detector, which is a function of the particles kinetic energy, $t(E)$. In the simplest form of drift tube, by knowing the distance between the target and the detector, the electron velocity and so the electron energy can be retrieved. However, in a simple drift tube, all the electrons which are photoemitted within the analyzer accepted solid angle are able to reach the detector, which could cause detector saturation, especially at sources like XFELs, which deliver high intensity photon pulses on a sample. Furthermore, for drift tubes of conventional size, the energy resolution is severely degraded at high kinetic energies (see Section 2).

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In the present work we investigate through simulations with the SIMION® software [7] two different types of retarding TOF analyzers suitable for high energy photoemission experiments at a pulsed X-ray sources, namely a linear lens and a spherical mirror. Retardation of electrons has the effect of improving the energy resolution while chromaticity in the linear lens and dispersive power in the spherical reflector, allow these analyzers to select only a part of the kinetic energy spectrum (i.e. the analyzers act as a band-pass filter); this energy window will correspond to a well defined time-of-flight window. This last aspect is important at high intensity sources like the European X-Ray Free Electron Laser (EXFEL), to avoid detector saturation. The EXFEL, which is currently under construction at Hamburg, should deliver about 27,000 pulses/s with a duration of 2–100 fs in the hard X-ray regime [8,9]. We have to note that the quoted 27,000 pulses/s does not imply that the repetition rate is 27 kHz, due to the particular time structure of the electron bunches in the linear accelerator of EXFEL. Indeed in one second ten electron bunch trains are produced, each with a total duration of 600 μs (so the time separation between trains is 99.4 ms): in each train there are 2700 bunches, each with a duration of 2–100 fs and a separation of about 220 ns. So in a single train, the actual repetition rate is about 4.5 MHz. This will be an improvement with respect to the low repetition rate of already existing XFELs like SACLA (10 Hz, with a maximum of 60 Hz [10]) and the Linac Coherent Light Source (LCLS, 120 Hz [11]). In order to be able to assign to each detected photoelectron bunch the photon pulse which has generated it, the interval of time-of-flights of the electrons which arrive on the detector must be shorter than the time separation between two consecutive photon pulses. This can be done by properly choosing the width of the acquired energy window (which corresponds to a suitable time window).

The paper is structured as it follows: in Sections 2 and 3 the case of a linear TOF spectrometer based on an electron lens and of a TOF spectrometer based on the time-energy dispersion properties of a spherical electron mirror will be presented, respectively. In Section 4 the results for the two systems will be discussed and compared.

2. Linear time-of-flight spectrometer

2.1. Spectrometer design

Linear TOF spectrometers based on a drift tube have always attracted much attention thanks to their intrinsic mechanical simplicity, but have found limited use at intermediate and high electron energies due to the poor energy resolution achievable. In a TOF system, the absolute energy resolution ΔE can be estimated as the product of the time resolution Δt and of the so-called differential energy resolution, $F(E) = (dt/dE)^{-1}$:

$$\Delta E = |F(E)|\Delta t, \tag{1}$$

In the case of a drift tube

$$\frac{dt}{dE} = -\left(\frac{m}{8}\right)^{1/2} \frac{L}{E^{3/2}} \Rightarrow F(E) = -\left(\frac{8}{m}\right)^{1/2} \frac{E^{3/2}}{L}, \tag{2}$$

where m is a the electron mass and L the flight distance (i. e., the distance between the electron source and the detector). Eq. (2) is obtained by inverting the expression for the classic kinetic energy, with the electron velocity $v = L/t$. One can see that the time-energy dispersion (TED) dt/dE decreases with the electron energy and so the energy resolution degrades at higher kinetic energies, because ΔE increases with the energy itself. To give some orders of magnitude, to achieve an energy resolution of 100 meV at 10, 000 eV kinetic energy, if we consider an uncertainty on the flight time of $\Delta t \simeq 0.1$ ns (a typical value for delay line detectors) then we obtain, using Eqs. (2) and (1), $L \simeq 1.2$ km. It becomes immediately clear that for high energy electrons, in the 5–10 keV range, the use of a drift tube for photoelectron spectroscopy is not practical. Relativistic effects were not included in this simple calculations, because at 10,000 eV the difference between relativistic and classical velocity is still smaller (for kinetic energy $E = 10$ keV, $v_{cla} = 0.198c$ and $v_{rel} = 0.195c$) than other sources of uncertainty intrinsic to the simulation process which we used to characterize the spectrometer. Furthermore we have to take into account that the time resolution Δt is not affected only by the time resolution of the detector (ΔT_1). Indeed this is not correct, because electrons with the same kinetic energy but different emission angles will follow trajectories with different lengths, which in turn means different transit times. So, a bunch of monochromatic electrons will have a spread in time of flight (ΔT_2), which must be taken into account when evaluating the time resolution and then the energy resolution. In first approximation the overall time resolution Δt can be written as

$$\Delta t = \sqrt{{\Delta T_1}^2 + {\Delta T_2}^2}. \tag{3}$$

In order to realize a TOF spectrometer with high transmission and resolving power, two systems based on a cylindrical retarding electron lens, instead of a drift tube have been recently proposed, namely ArTOF [12–14] and THEMIS [15]. These systems are capable to focus a wide angular distribution on a detector; furthermore for a given electron energy and take-off angles, there will be an unique position on the focal plane at the end of the lens. By using a time-resolved 2D delay-line position sensitive detector, both the arrival time and the $x-y$ impact coordinates of the electrons can be measured. This allows to calculate the trajectory and to retrieve not only the kinetic energy, but also the in-plane emission angles of the electrons.

The main advantage of these lenses is that by properly biasing the electrodes, the transit times can be made a few orders of magnitude longer with respect to a simple drift tube with the same length. Clearly this requires a retardation of the electrons to lower kinetic energies, which means that, due to the chromaticity of the lenses, only a limited set of kinetic energies can be acquired in parallel: in order to probe a wider kinetic energy range it will be necessary to scan the applied voltages, such as in the case of a hemispherical analyzer.

In the present work we extend the investigation of the use of an advanced lens system such the ones described in refs. [12–15] in order to detect high energy electrons (more than 5 keV). Starting from the geometry of those systems we develop an instrument with good energy resolution even at high energy by strongly decelerating the electrons (from several keV to few eV), keeping at the same time a good angular acceptance and an as high as possible transmission. A configuration with six electrodes with inner diameter of 10 cm, gap between the electrodes of 1 cm and a total length of 100 cm (see Fig. 1(a)) allowed obtaining an instrument with practically manageable dimensions given the constrain to work at high energy.

The potentials of the electrodes have been fixed through an iterative process of simulation using the SIMION® software, in which various potential configurations were proved, evaluating for each one the transmission curve, until we found a configuration which allowed to retain, in the accepted solid angle (about 50 msr), a transmission of 100% in the widest possible kinetic energy range, which resulted to be as wide as about 110 eV around a central energy of around 10,000 eV. In particular, Fig. 1(a) shows some electron trajectories from a 0.5 mm circular source placed at 40 mm from the lens entrance at energy of 10,260 eV, the center of the energy band at which the electrodes potential were optimised. One can note that the trajectories exit almost parallel from the lens, within a circular radius of about 15 mm. Fig. 1(b) and (c) displays electron trajectories in the end part of the instrument, for energies