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Application of a time-of-flight spectrometer with delay-line detector for time- and angle-resolved two-photon photoemission

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ABSTRACT

We describe the design and operation of a time-of-flight electron spectrometer which is capable of simultaneously acquiring the energy and momentum distribution of low-energy photoelectrons in two dimensions parallel to the surface. We discuss its capabilities and limitations in particular for time- and angle-resolved two-photon photoemission (2PPE) with pulsed lasers. The performance of the spectrometer is demonstrated by presenting 2PPE data on the momentum-dependent electron dynamics of the first ($n = 1$) image-potential state on Cu(0 0 1). The data reveal a weak but systematic dependence of the decay dynamics on sample azimuth with one-fold symmetry which we attribute to a small residual step density on the nominal flat surface.

1. Introduction

Time- and angle-resolved two-photon photoemission (2PPE) is a very powerful experimental tool for the investigation of electron dynamics at solid surfaces, as it makes it possible to monitor the temporal evolution of energy and parallel momentum of optically excited electrons in initially unoccupied electronic states [1]. For this pump-probe technique, a short pump laser pulse excites electrons in initially unoccupied states while a subsequent probe pulse with variable time delay is used for photoemission of the excited electrons. One of the strengths of this technique is based on the fact that the parallel momentum of electrons is conserved for photoemission from surfaces with translational symmetry, which makes it possible to deduce the energy and parallel momentum of the electrons in the transiently excited state by measuring their kinetic energies and emission angles after photoemission. This can be accomplished by different types of electron analyzers.

Many 2PPE setups use low-order optical harmonics of femtosecond laser oscillators with repetition rates of ~100 MHz for the generation of pump and probe pulses. In combination with electrostatic electron analyzers and electron counting, the high repetition rate of these lasers makes it possible to achieve a superior dynamic range and signal-to-noise ratio even at low pulse energies, which are necessary to avoid space charge effects. Electrostatic electron analyzers are often equipped with multiple channeltron detectors which are mounted along the energy-dispersive detection direction. Even if this already introduces a certain degree of parallel detection, it is typically used to enhance the signal-to-noise ratio when scanning the kinetic energy. The mapping of the angular distribution of the photoelectrons with these spectrometers requires the sequential acquisition of spectra at different discrete tilt and azimuthal rotations of the sample. As in the case of angle-resolved photoemission spectroscopy (ARPES) of occupied states, time- and angle-resolved 2PPE has strongly benefited from the introduction of display-type hemispherical electrostatic analyzers [2] after angular-resolved lens modes were developed also for low electron energies [3]. The parallel detection of a certain quasi-continuous energy and momentum range makes it possible to simultaneously investigate momentum-dependent electron dynamics within electronic bands without sample tilt at least along one direction parallel to the surface [4].

Amplified laser systems in combination with optical parametric frequency generation are frequently used for 2PPE when a broader and continuous tunability of the photon energy is required

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[5,6]. The lower repetition rate of these laser systems makes it possible to alternatively use time-of-flight (TOF) electron spectrometers with high transmission, which can also reach an energy resolution of down to a few meV for low-energy electrons [7]. In these spectrometers, channelplates are typical used as fast electron detectors within a limited acceptance angle. Parallel detection at several emission angles with a time-of-flight electron spectrometer was introduced very early by Haight et al., who used an array of 64 individual anodes, each of them representing a single electron detector [8]. The elaborated parallel detection electronics and signal processing, however, prevented a wide spread use of this concept.

The sensitive detection of charged particles with high temporal and spatial resolution became more popular with the introduction of delay line detectors [9–13], which were initially developed for cold target recoil ion momentum spectroscopy (COLTRIMS) [14]. In these detectors, the position of a charge cloud generated by a MCP is determined by its travelling time on a meander-shaped copper wire which serves as anode. The meander form results in a low effective propagation speed perpendicular to the wire, which strongly enhanced the accuracy of the determined position. A minimum of two such delay lines are required for the reconstruction of the position in two dimensions. The multi-hit capability of theses detectors was improved by the use of three delay lines in a hexanode arrangement [12,13]. Recent developments take up again the design principle of Haight et al. and consist of a large number of parallel delay lines in order to considerably increase the multi-hit capability [15]. Delay line detectors are now used also in different types of TOF electron spectrometers for the investigation of photoemission from surfaces [16–21]. Commercially available TOF electron spectrometers with delay line detectors include electrostatic lenses which increase the usable energy range [19,20]. The reconstruction of the emission angle or parallel momentum from the spatial position on the detector in these spectrometers, however, requires an advanced simulation of the electron trajectories through the lens system [22].

Here, we use the simpler approach of a field-free drift tube which has the advantage of a simple linear relation between lateral position and parallel momentum. A similar TOF spectrometer equipped with a hexanode delay line detector for ARPES with pulsed lasers has been described in Ref. [17] where momentum-resolved spectra have been presented for a single direction parallel to the surface. In this paper we focus on the capabilities of a TOF spectrometer with delay line detector for time- and angle-resolved 2PPE. First, we describe the design and operation of our spectrometer and discuss the restrictions on the usable event rate with respect to the signal-to-noise ratio and linear range at a given laser repetition rate. In the second part, we demonstrate the performance of the spectrometer for 2PPE by presenting two-dimensional data on the momentum-dependent electron dynamics of the first ($n = 1$) image-potential state on Cu(0 0 1).

2. Time-of-flight spectrometer

2.1. Basic layout

The basic layout of the TOF spectrometer is depicted in Fig. 1. It consists of a grounded field-free cylindrical drift tube with a conical nose, both made of vacuum-compatible aluminium. The nose has an aperture of 10 mm in diameter which is covered by a fine mesh for electrical shielding and for providing a flat electric potential with respect to the sample. All parts of the drift tube were covered with graphite in order to achieve a homogeneous work function. Time-of-flight detection of the photoelectrons in the two directions perpendicular to the spectrometer axis is realized

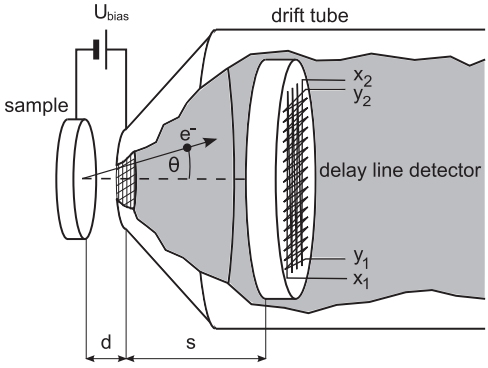

Fig. 1. Schematic layout of the time-of-flight spectrometer which consists of a cylindrical drift tube with conical nose and a delay line detector. The flight length s within the field-free drift tube can be varied between 37.5 mm and 240 mm.

by a commercially available two-dimensional delay-line detector (DLD40) with an active diameter of 38 mm provided by RoentDek Handels GmbH, Germany. The detector is covered by a grounded and graphite-covered mesh, which closes the field-free region of the drift tube. The delay line detector can be translated within the drift tube in order to vary the length of the field-free region s between 37.5 mm and 240 mm. This makes it possible to increase the acceptance angle up to $\sim\pm 25^\circ$ for small s or to alternatively enhance the energy resolution for large s. In addition, the whole spectrometer can be translated by up to 50 mm in order to place the entrance grid of the flight tube in a typical distance of $d = 3-5$ mm to the sample. Magnetic shielding is realized by a single μ-metal liner of the UHV-chamber in combination with three pairs of external coils for a coarse compensation of the earth magnetic field.

2.2. Position sensitive TOF detection

The delay line detector consists of a stack of two microchannel plates (MCP) in Chevron configuration with an active diameter of 38 mm which amplify single photoelectrons into a charge cloud behind the MCP stack. The front side of the MCP stack is biased by +200 V in order to increase the detection sensitivity of the low-energy photoelectrons. The reference signal for the determination of the electron position as well as its flight time is generated by capacitive coupling from the MCP stack and defines the arrival time of the electrons at the MCP t_{MCP}. This voltage pulse is fed trough a separate electrical feedthrough, a fast preamplifier (FAMP1+, RoentDek), and a constant fraction discriminator (CFD1b, RoentDek). It is then used to trigger an eight-channel time-to-digital converter (TDC8HP PCI card, RoentDek) which digitizes all timing signals with a time resolution of better than 100 ps (25 ps per bin).

The two-dimensional position (x, y) of the charge cloud is measured by its propagation time on two perpendicularly oriented delay lines. Each delay line consists of a pair of a signal and a reference copper wire, which are installed in a meandering pattern and the ends of which are connected to an electrical feedthrough. The signal wire is biased by +50 V with respect to the reference in order to generate a current difference between signal and reference wire at each end. After conversion of these current differences into short voltage pulses, the four signals of the delay lines are processed by fast amplifiers and constant-fraction discriminators which are combined onto a single board (ATR19, RoentDek). These four pulses define the times $t_{x_1}, t_{x_2}, t_{y_1}, t_{y_2}$ from which the position x and y of the incident electron with respect to the centre of the detector can be determined by

$$x = c_{\text{eff}}(t_{x_1} - t_{x_2}),$$
$$y = c_{\text{eff}}(t_{y_1} - t_{y_2}). \quad (1)$$