# Complete Momentum and Energy Resolved TOF Electron Spectrometer for Time-Resolved Photoemission Spectroscopy

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**Abstract**

Over the last decade, high-resolution angle-resolved photoemission spectroscopy (ARPES) has emerged as a tool of choice for studying the electronic structure of solids, in particular, strongly correlated complex materials such as cuprate superconductors. In this paper we present the design of a novel time-of-flight based electron analyzer with capability of 2D in momentum space (kx and ky) and all energies (calculated from time of flight) in the third dimension. This analyzer will utilize an improved version of a 2D delay line detector capable of imaging with <35 microns (700 x 700 pixels) spatial resolution and better than 120ps FWHM timing resolution. Electron optics concepts and optimization procedure are considered for achieving an energy resolution less than 1meV and an angular resolution better than 0.1 deg.

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## I. Introduction

The measurement of the energy spectrum of emitted electrons as a function of emission angle is the goal of Angle-Resolved Photoemission Spectroscopy (ARPES), which provides a mapping of the electronic structure of a solid and the symmetry properties of the lattice. The most widely used instrument for ARPES is the hemispherical Electron Energy Analyzer (EEA) [1]. This technique requires the mechanical scanning of the sample at least in one direction which that significantly reduces the accuracy of measured spectrum due to the degradation of the sample properties during the scan. To solve this problem we developed an Imaging Time-of-Flight (TOF) electron spectrometer (**Fig.1**) with Microchannel Plate (MCP) and Cross Delay Lines (XDL) electron detector having 2D spatial (700x700 lines) and temporal (better than 120ps FWHM) resolution [2]. The kinetic energy of electrons is determined by their precise arrival time at the detector. The advantage of a TOF-EEA is a significantly wider window of simultaneously detected energies and the stigmatic properties of the electron optics that avoid analyzer transmission calibration in two perpendicular directions. One of the first designs of a TOF-EEA for surface-science applications was introduced by Bachrach et al. [3], and was later further developed for gas-phase samples [4].

## II. Design concept of electron optical systems

The electron optical scheme of a TOF ARPES analyzer consists of several major components: the electrostatic Immersion Objective Lens forming diffraction pattern of photoelectrons in the plane of variable aperture; the telescopic doublet of Einzel lenses which project the diffraction plane of EOS on XDL detector with magnification in the range M = 0.6-1.8; the correction module consisting of two octupole deflectors and a stigmator. This lens system was optimized for angular resolution better than 0.1 degree and energy resolution < 5meV for 0.5 mm wide electron emission spot on the sample and the 30 degrees angle of divergence . The major constrain on image quality of static type EOS is the energy spread in the photoelectron flow (i.e. system's mean chromatic aberration). In a TOF EOS with a position sensitive detector this problem doesn't exist because the collected electrons are sorted by their arrival time on the detector or, in other words, by energy. For a detector with a temporal resolution <120ps and the pass energy of the analyzer of 10eV it corresponds to the electron's energy spread in the range of few millivolts. At the same time the image distortions due to chromatic aberration (<10 um) are significantly less than spatial resolution of the detector.

### III. Image Formation in Momentum Space

Immersion objective lens (Fig.1) allows shifting the electron kinetic energy $\varepsilon_0$ by accelerating or decelerating the beam to a particular value specified by $\varepsilon_{pass} = \varepsilon_0 - U_0 + U_1$ which is considered as a pass energy of the analyzer, where $U_0$ and $U_1$ are potentials of the first and the last electrodes of the lens. Object lens was designed to create a photoelectron diffraction pattern in the plane of variable aperture for all possible values of the immersion parameter $K_{im} = \varepsilon_0 / \varepsilon_{pass}$ . The telescopic doublet

of the Transport and Projective lenses (Fig.1) translate this image onto the plane of the XDL detector. In such a system, only geometrical (spherical, coma, astigmatism) aberrations take effect and some of them are minimized by an appropriate lens design. The spherical temporal aberration and astigmatism are corrected during the image data processing.

### IV. Energy Resolution and Temporal Aberrations

The time propagation of charged particles with initial phase coordinates $r_0^{'}$ and $r_0$ , energy $\varepsilon_0$ and energy variation $\Delta\varepsilon_0$ through an axial symmetrical EOS with potential distribution U(z) along optical axis Z could be expressed as:

$$t(\varepsilon_0 \mid r'_0, r_0, \Delta\varepsilon_0) = \int_0^z F(r'_0, r_0)[U(z) + \varepsilon_0 + \Delta\varepsilon_0)]^{-1/2} dz \quad (1)$$

By denoting $< t \mid i, j, k >= \partial t / \partial r_0^{'i} \partial r_0^j \partial \Delta\varepsilon_0^k$ and assuming $r_0^{'}$, $r_0$ and $\Delta\varepsilon_0$ are small parameters, than the propagation time can be expressed in the form of a power series:

$$t(\varepsilon_0 \mid r'_0, r_0, \Delta\varepsilon_0) = t_0(\varepsilon_0) + \Delta t_\varepsilon(\Delta\varepsilon_0) + \Delta t_{rr'}(r'_0, r_0) + \Delta t_{rr'\varepsilon}(r'_0, r_0, \Delta\varepsilon_0) \quad (2)$$

In Eq, ( 2 ) - $t_0 =< t \mid 000 >$ - flight time of a particle along the optical axis;

$\Delta t_\varepsilon(\Delta\varepsilon_0) =< t \mid 001 > \Delta\varepsilon_0 + < t \mid 002 > \Delta\varepsilon_0^2$ - second-order variation of the flight time due to the energy spread. It also defines the energy dispersive properties of EOS.

$\Delta t_{rr'}(r'_0, r_0) = R_0^t Q R_0$ - the temporal-spherical second-order aberration and

$\Delta t_{rr'\varepsilon}(r'_0, r_0, \Delta\varepsilon_0) = R_0^t G R_0 \Delta\varepsilon_0$ - the third-order aberration, which defines the variation of the energy dispersive properties of EOS on the detector plane. The **Q** and **G** are matrixes of aberration coefficients

$$Q = \begin{pmatrix} <t|200> & <t|110> \\ <t|110> & <t|020> \end{pmatrix}; \qquad G = \begin{pmatrix} <t|201> & <t|111> \\ <t|111> & <t|021> \end{pmatrix}$$

and $R_0 = \begin{pmatrix} r'_0 \\ r_0 \end{pmatrix}$ is the vector of the initial phase coordinate.

## V. TOF_EEA design and reconstruction of the energy spectrum

Translation of time-of-flight into the energy spectrum could be done directly by solving Eq.1 as $\varepsilon_0 = t^{-1}(z \mid r'_0, r_0, \Delta\varepsilon_0)$ . More efficient way to find the kinetic energy of electron as function of flight time $\varepsilon_0(t)$ derived from solution of the second order polynomial equations ( 2 ). The optimization of analyzer's EOS is done with a criterion of maximum energy resolution and the beam transmission rate. For convenience let's introduce the normalized flight time as $\tau = t / t_{pass}$ - where t and $t_{pass}$ are flight times of particles with kinetic energy $\varepsilon_0$ and $\varepsilon_{pass}$ accordingly. From **Fig.2 it** follows that the flight time is dependent not only on the immersion parameter of EOS but also on the emission angle of electrons and has to be taken in consideration during the calculation of

the photoelectron energy spectrum. Spatial energy resolution at the detector plane is shown on **Fig. 3** as function of electrons emission angle $r_0^{'}$.

### VI. Experiment setup

For testing the analyzer we developed a pulsed (70 ps) electron gun with a back-illuminated photocathode ( **Fig.4**). The energy spread of the photoelectrons was measured to be 0.4eV. A pinhole mask with 1 mm round holes was installed at the entrance to the simplified setup of the analyzer (fixed aperture and no correction elements in the scheme) (**Fig.4).** An additional test was done on the ALS beam line 12.0.1. A full spectrum up to 60eV was measured from cooled (T=15 K) gold sample (**Fig.5**).

### VIII. Conclusion.

The tests led to a conclusion that the analyzer's angular resolution is better than 0.2 degree. The performance of newly developed TOF electron spectrometer is in good agreement with the simulation results. The new spectrometer is capable of quantitative ARPES experiment with a submilivolt energy resolution, opening new frontiers in the photochemistry and catalysis research. Also we are greatly appreciated support from DOE (No. DE-AC02-05CH11231)

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**Figures:**

**Fig. 1.** Electron Optical Scheme of TOF ARPES analyzer:

1. Objective electrostatic axial symmetrical lens
2. Variable iris aperture, radius = 0.25- 2”
3. Transmission and Projective lenses
4. Correction elements- two deflectors and stigmator
5. Cross Delay Lines 2D+T electron detector

**Fig. 2**. Electrons normalized flight time $\tau = t / t_{pass}$ as a function of the emission angle for different emersion parameters ( Kim) of TOF analyzer.

**Fig.3.** Energy resolution of the analyzer as a function of the emission angle of electrons for different emersion parameters ( Kim) of analyzer.

**Fig. 4**. The testing setup of TOF ARPES analyzer with a pulsed (FWHM - 70 ps ) photocathode electron gun.

**Fig.5.** TOF spectrum and a histogram images obtained from a gold sample on the specified peaks and overall spectrum range.