Energy- and Time-Resolved Microscopy Using PEEM: Recent Developments and State-of-the-Art

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Abstract. Two novel methods of spectroscopic surface imaging are discussed, both based on photoemission electron microscopy PEEM. They are characterised by a simple electron-optical set up retaining a linear column. An imaging high-pass energy filter has been developed on the basis of lithographically-fabricated microgrids. Owing to a mesh size of only 7µm, no image distortions occur. The present energy resolution is 70 meV. The second approach employs time-of-flight energy dispersion and time-resolved detection using a Delayline Detector. In this case, the drift energy and the time resolution of the detector determine the energy resolution. The present time resolution is 180 ps, giving rise to an energy resolution in the 100 meV range.

1. Introduction
Spectroscopic imaging of surfaces with high lateral resolution is of utmost importance in many fields of research and technology. Several approaches have been made, mostly using dispersive energy analysers. Of particular interest are analysers that transport a whole image because of a higher collection efficiency as compared to the alternative approach using a microfocus being scanned across the surface. Imaging dispersive spectrometers pose the challenge that the image aberrations must be minimised. A fully aberration corrected instrument is operated at a high-brilliance Synchrotron radiation source (SMART project [1]). Another type of instrument being corrected for the leading spherical aberration term is optimised for laboratory-source applications (NanoESCA [2,3]). Typical instruments using hemispherical analysers without aberration correction require Synchrotron radiation sources as well [4]. The task of implementing an imaging dispersive analyser into a microscope column requires very demanding electron optics. In the present paper we present two alternative approaches that keep the experimental effort much lower by using an imaging high-pass-energy filter based on photofabricated microgrids or, alternatively, an imaging time-of-flight optics. Both methods retain a linear electron-optical column and thereby reduce the complexity of the set-up to a minimum.

2. High-pass energy filtered PEEM using silicon microgrids
Results obtained with the first version of imaging retarding field analyser have been published previously [5]. It employed metal grids with mesh 63x63 µm². To improve the analyser, photofabricated silicon microgrids with only 7 µm mesh were developed in a collaboration with the IMM Mainz. Since this mesh size is smaller than the bores of the micro channel plate image intensifier
(10 µm) the image artefacts of the first version, e.g. shadows of the grid and Moiré patterns of the two closely-spaced grids, were removed. Owing to field penetration through the grids the energy resolution was limited to 400meV. Using the microgrids and an additional pre-retardation optics with a telescopic beam, the energy resolution was improved to ΔE=73meV at an injection energy of 50eV as shown in figure 1. In order to obtain differential spectra, the integral intensity vs. energy curves of each pixel are numerically differentiated. The kinetic energy ranges from 0 to 1600eV with an analyser transmission of > 50% over the whole energy range. Images are free of shadows or distortions. The range of applications of this imaging analyser comprises spectromicroscopy, work function contrast imaging or local work function measurements with few meV sensitivity.

An example application of this analyser is shown in figure 2. A spectroscopic image series and an integral sum image were taken for a polycrystalline Cu sample with 4.9eV excitation (Hg lamp). Local spectroscopy reveals that two different characteristic spectra exist. The data have been processed using the spectral unmixing algorithm [6, 7]. The code performs a least squares fit of up to four reference spectra (here only two: green and red) to each pixel in the spectral image stack. It thus identifies the correlation of the image with the “finger print” of the related reference spectra. The weight of each reference spectrum is displayed in a separate false-colour image (spectral unmixed image).

**Figure 1.** Dependence of the energy resolution on the injection energy into the retarding field analyser. The energy resolution was measured at the Fermi-edge region of a differential spectrum taken for He I (21.2eV) excitation with a sample of Ag on Mo (@RT). It reveals an analyser energy resolution of 73 meV at 50eV injection energy (16%/84% criterion).

**Figure 2.** Series of spectroscopic images taken with the imaging retarding field analyser. The energy filtered image stack shows the surface of a polycrystalline Cu sample; Excitation: 4.9eV (Hg arc lamp).
3. Energy- and time-filtered PEEM using a time-of-flight optics with a Delayline Detector

This approach is based on the combination of PEEM with the well-established technique of Time-of-Flight (ToF) spectroscopy as discussed previously [8-10]. In our set-up we used a ToF-PEEM with dual detection. A channelplate / screen / CCD camera unit is mounted in forward direction, the Delayline Detector (DLD) assembly can be moved into and out of the beam by means of a UHV linear feedthrough. The working principle of the DLD is described in detail in [10]. In the case that an external time marker pulse exist (e.g. from a Synchrotron or laser source) the absolute time of the electron impact with respect to the time marker can be determined. The time coordinate $t$ can be exploited both for spectroscopic and time-resolved imaging.

The lateral resolution of the ToF-PEEM is about 20 nm and is not limited by the DLD. Time resolution for time-resolved detection is 180 ps ($\sigma$). Usable kinetic energies in the drift space range between 1200 eV for time-resolving experiments (practically no time dispersion) and 8 eV for high energy resolution (maximum time dispersion). The theoretical energy resolution resulting from the time resolution is 85 meV at 20 eV and 22 meV at 8 eV drift energy. Presently, we achieved a resolution of about 100 meV by measuring the Fermi edge of Cu at room temperature.

The energy resolution is achieved by the temporal dispersion in the low-energy drift space (length $l$, potential $U_l$). It is ultimately limited by the time resolution of the detector and the stability of the drift energy (e.g. a ripple of the voltage $U_l$). Time-to-energy conversion is made using the relation

$$E_{\text{kin}} + \Delta \Phi = eU_l \left( \frac{\Delta t}{t_{1,0}} \right)^2 - 1 \approx 2 \left( \frac{2e}{m_e} \right) \frac{1}{2} U_l^{3/2} l \Delta t$$

with $\Delta \Phi$ being the work function difference between the sample and the drift tube. $\Delta t$ is the difference in arrival time of an electron with kinetic starting energy $E_{\text{kin}}$ and an electron with zero starting energy arriving at $t_{1,0}$.

![Figure 3: Series of time slices (top), sum image and local time-of-flight spectra (bottom) of a structured Ag on Si-sample, taken with 400 nm picosecond-laser excitation. Note that the intensity in the “hot spot” is enhanced by a factor of 200.](image)

A typical result taken with this instrument is given in figure 3. A structured sample of Ag on Si was irradiated with 400 nm picosecond-laser radiation from a pulsed diode laser. The drift energy was
20 eV and the laser signal was used as time marker for the DLD. The images exhibit the microstructure and, in addition, a number of “hot-spots” as previously observed using femtosecond radiation from a TiSa laser [11]. The image series (top) shows time slices of the same sample area. The slices are extracted from the full 3D (x,y,t) data stack by setting a condition for the arrival time $\Delta t$ of the photoelectrons. The sum image is an integrated image over the time coordinate $t$. ToF spectra are taken for a smooth area of the Ag structure (marked by the large square) and a single “hot spot”, i.e. a point of high intensity (small square). The series of time slices reveals that the smooth areas show up at earlier arrival times than the “hot spots”. This reflects the fact that the low-energy cut-off of the “hot spot” spectra appear at smaller kinetic energies. The strong field enhancement due to plasmon excitation gives rise to a marked lowering of the workfunction threshold, comparable to the effect of a Schottky field emitter.

4. Conclusion

Two novel methods of spectroscopic imaging based on a photoelectron microscope have been presented. Both are characterised by a rather simple electron-optical set-up. In particular, the devices are implemented into a linear microscope column, thus making the alignment procedure as easy as for a standard non-energy-resolving instrument.

The first approach employs a pair of microgrids as high-pass energy filter in a retarding field arrangement. Owing to its small mesh size, no image artefacts are visible and the energy resolution is very good (presently 73 meV). The transmission is > 50 %, i.e. the intensity problem dispersive energy analysers have for high energy resolutions practically does not exist. Differential spectra and images are obtained by numerical differentiation of the intensity curve of each pixel. The imaging retarding field filter can be used with any CCD camera and works for pulsed as well as cw excitation sources.

The second approach is based on time-resolved image detection and thus requires a time structure of the exciting radiation. The image is acquired by means of a Delayline Detector that accumulates 3D (x,y,t) histogram arrays by detection of single counting events. All time slices with respect to the time marker of the excitation are acquired simultaneously. The full 3D histogram is comparable to the sequentially recorded image stack of the retarding field assembly. After acquisition, the 3D histogram array can be processed. Via time conditions one can define time slices corresponding to energy-filtered images in Time-of-Flight spectromicroscopy. The same approach gives “snapshots” in time-resolved observation of dynamic processes. Via conditions for the lateral coordinates x and y one can define microareas for microspectroscopy.

In conclusion, the microgrid analyser is very flexible with regard to the light source, i.e. it can be operated with a Hg lamp for threshold excitation, a helium lamp for VUV excitation or synchrotron radiation for soft X-ray excitation. The Delayline-Detector needs pulsed excitation sources (lasers, Synchrotron radiation). It develops its full potential for time-resolved observation of dynamic processes or in Time-of-Flight spectromicroscopy experiments.

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