

**Aberration correction in photoemission microscopy and applications in photonics  
and plasmonics**

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**Abstract**

We report the design, assembly, operation and application of an aberration-corrected photoemission microscope. The instrument used novel hyperbolic mirror-correctors with two and three electrodes that allowed simultaneous correction of spherical and chromatic aberrations. A spatial resolution of 5.4nm was obtained with this instrument in 2009, and 4.7nm in subsequent years. New imaging methodology was introduced involving interferometric imaging of light diffraction. This methodology was applied in nano-photonics and in the characterization of surface-plasmon polaritons. Photonic crystals and waveguides, optical antennas and new plasmonic devices such as routers, localizers and filters were designed and demonstrated using the new capabilities offered by the microscope.

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**Project titles and active periods**

- (1) Electron Mapping on the Nanoscale (6/2007-6/2010)
- (2) High-Resolution Photoemission Electron Microscopy (6/2010-6/2013)
- (3) High-Resolution Photoemission Electron Microscopy on Femtosecond Time Scales (6/2013-6/2017)

**Introduction**

Electron-optical aberration correction has been one of the grand challenges in electron microscopy since the 1940s. While theory provided significant insights into this challenge (1) experimental progress was much slower. A break-through was established around the year 2000, when improved TEM resolution could be demonstrated with the help of multi-pole correctors. In two independent papers (2, 3) a resolution improvement up to 40% was demonstrated based on spherical aberration correction in TEMs. Chromatic aberration was not addressed in these two developments. Instead chromatic aberration was minimized by energy filtering.

Independent of the multipole-based correction an alternative path towards aberration correction was demonstrated first theoretically (4), then experimentally (5) based on a curved mirror corrector by G. Rempfer in the 1990s. The experimental results were obtained in an optical bench, not a microscope, and showed that both spherical as well as chromatic aberration correction could be obtained simultaneously with a curved mirror. Despite the extremely promising results there was still a challenge to build an improved microscope.

This project connected to this challenge and targeted the demonstration of an aberration-corrected photoemission electron microscope (PEEM) using a hyperbolic mirror corrector. In the first portion of the project the research included the final assembly of an all home-built aberration-corrected instrument and obtaining best spatial resolution. The second portion of the project targeted exploratory work on semiconductor surfaces and nanostructures, and the design and construction of a second aberration-corrector with more versatility. Also, the integration of a femto-second laser for ultrafast imaging was planned, and new user-friendly software for the two correctors was to be developed. The third phase of the project addressed new methodology in applying PEEM to nano-optics and plasmonics in dielectric and metallic nanostructures.

In the following we will first briefly report the instrument development and subsequently the work on plasmonic and photonic nanostructures.

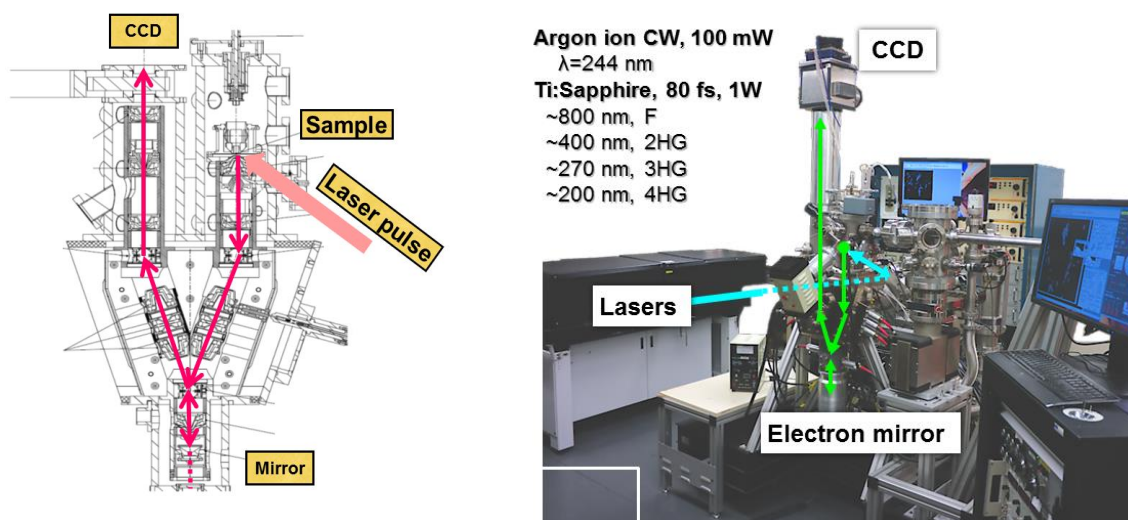


Fig. 1 a: Design of the aberration-corrected PEEM at PSU including beamline, mirror position and beam-splitter; b: Photo of the complete instrument with laser system and control units.

### Instrument development

Photoemission electron microscopes use photo-emitted electrons at comparatively low electron energies for electron imaging. In general, large chromatic and spherical aberrations occur in these instruments. Typically both types of aberrations are of the same size and importance, and useful aberration correction

schemes for these instruments must address both aberration types. Furthermore with the inherent low quantum efficiencies of the photoemission process, an energy filtering is not acceptable for reducing chromatic aberration (6). Therefore mirror correctors with their capability to address both aberrations simultaneously appear as the natural choice for photoemission microscopes. Indeed, following Rempfer's work (4, 5) mirror corrected PEEMs were built at BESSY in Germany, the ALS in Berkeley and at IBM Watson in New York. With the guidance and initiative of G. Rempfer, a group was also formed at Portland State University to build a corrected PEEM leading to the current project. R. Koenenkamp became the head of this group in 2003. He obtained initial funding from NSF in 2004 and subsequent funding from DOE-BES for a stand-alone aberration-corrected PEEM to be designed and built at PSU. The lay-out for this PEEM used a simple optical design with 11 electrostatic lenses, a vertical incidence, symmetric, hyperbolic mirror, and a small-deflection magnetic beam splitter (\*1-\*3) as shown in Fig. 1. Some of the optical elements and the complete instrument with load-lock, sample prep chamber and lasers is shown in part b of the figure.

The central initial goal in the project was to obtain improved spatial resolution. This naturally required extensive experimental characterization of the newly built instrument and detailed benchmarking with respect to the underlying design theory which used classical trajectory calculations and simulations. The experimental characterization utilized a reversed electron beam to measure the two aberration coefficients,  $C_s$  and  $C_c$ , for various settings of magnification, acceleration potential and electron starting energy. The experimental values were then used in the analytical model (\*2). It was soon found, however, that the experimental data were not sufficiently accurate. A more precise calibration of the voltage supplies was needed, shielding against magnetic stray fields had to be installed, measures against current arcing and leaking had to be taken, and internal vibration of the microscope had to be suppressed. Although time-consuming, these measures eventually led to the desired results. Using 244nm cw illumination from a frequency-doubled Ar<sup>+</sup> laser 5.5nm resolution could be established in 2009 on inorganic silver quantum dots on Cr substrate, and a little later 5.4 nm resolution was established on biological samples of sarcoplasmic reticulum, also on Cr covered glass substrates (\*3). These were the first results in electron optics demonstrating resolution improvement with a mirror corrector. A ~40% improvement compared to non-corrected resolution results was obtained. Following our work, the BESSY group in Berlin reported in 2013 a resolution of 18nm for x-ray light at BESSY (7), establishing also clear improvement over non-corrected imaging in synchrotron operation.

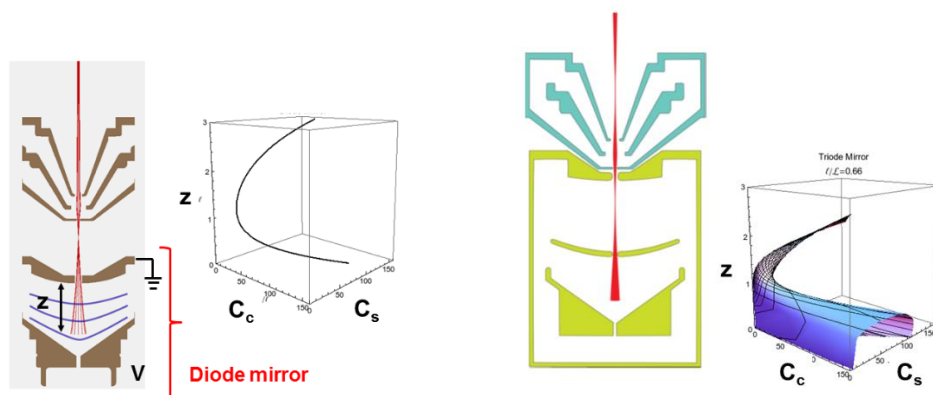


Fig. 2a: Diode mirror design and relationship of  $C_c$  and  $C_s$  as function of mirror length; b: Triode mirror and relationship of  $C_c$  and  $C_s$  as a function of mirror length.

Our 2009 results were obtained with the 2-electrode mirror shown in Fig. 2a. With this simple arrangement chromatic and spherical aberrations can be corrected simultaneously, but the final tuning between the two coefficients cannot be chosen independently. Both coefficients are coupled as illustrated in part b of the figure. Therefore microscopy with this type of corrector requires the same specific measurement situation in all experiments needing aberration correction. In PEEM in particular, magnification, photoelectron starting energies and acceleration voltage have to satisfy tightly defined limits for optimum results. As the electron starting energies depend on the photon energy in PEEM, this issue somewhat limits the range of photon energies in the use of the 2-electrode corrector. As the microscope at PSU was laid out only for near-threshold photoemission microscopy, these restrictions were not severe and entirely consistent with the envisioned future work.

For more flexibility and a fully independent adjustment between chromatic and spherical aberration a more versatile and more complex mirror was needed, and the design and implementation of such a mirror was part of the second project portion. This 3-electrode mirror is shown in Fig. 2c. It incorporates a second hyperbolically shaped electrode in the mirror field (\*7-\*9,\*25,\*34). The  $C_c$ - $C_s$  characteristics of this mirror, shown in Fig. 2d, form a 2-dimensional surface, opening a wider parameter space than given by the line in Fig. 2b. Within this space an independent control of  $C_s$  and  $C_c$  is possible. The tuning itself is computer-controlled. In a typical application both aberration coefficients can be varied within ranges of ~100%. While reasonable image quality could be obtained with this mirror in subsequent work, we were unable to reach the resolution values of the simpler 2-electrode mirror (\*3). We attribute these difficulties mostly to alignment problems with the mechanically somewhat weak structure. A more robust implementation of the same mirror has been made, but has not been extensively tested so far.

Table 1 gives an overview of our results with continuous and femtosecond pulse illumination in various wavelength regions. More details on these results can be found in ref. (\*26).

<b>Illumination</b>	<b>Resolution</b>
<b>dc, <math>\lambda=244\text{nm}</math> (Single photon mode)</b>	<b>4.9 nm</b>
80 fs, $\lambda=200\text{nm}$ (Single photon)	30 nm
80 fs, $\lambda=260\text{nm}$ (Single photon)	20 nm
80 fs, $\lambda=400\text{nm}$ (2-photon)	15 nm
80 fs, $\lambda=800\text{nm}$ (3-photon)	15 nm

Table 1: PEEM spatial resolution results for various illumination conditions including continuous and pulsed laser illumination at various wavelengths.

### Methodology work on PEEM in photonics and plasmonics

PEEM has been used in a wide spectrum of fields including magnetism, biology, catalysis, band structure analysis and others (6). It had long been recognized that in combination with pulsed lasers fast and ultrafast imaging can conveniently be performed. Pump-probe work has also been pursued in PEEM to

characterize electron and phonon dynamics. In work with pulsed lasers space-charge effects and surface charging often put limits on the usable intensity. As a consequence single-pulse imaging is not possible with nanosecond or shorter pulses, as the charging effects induce strong image deterioration. However, when reproducible dynamics are being probed and repetitive pulsing can be applied, ultrashort imaging frame times for micrographs can be obtained and pump-probe techniques can be applied to visualize the full dynamics of fast processes.

Besides imaging the dynamics of electron and phonon properties, there is also the possibility of imaging *photon* dynamics at surfaces. This is because the photoelectron yield in PEEM is also a function of the local light intensity. This in turn allows the direct evaluation of electric or magnetic field distributions at surfaces, as well as excitons, polariton and surface-plasmon distributions (8, 9). With the high spatial resolution of PEEM these field and intensity distributions can be explored at scales well below the light-optical diffraction limit. PEEM thus gives a convenient access to the optical near-field region. Moreover, while PEEM traditionally required ultra-violet light for the photoemission process, the high pulse intensities now available in femtosecond pulses open the way to non-linear photoemission processes and thereby extend the use of PEEM into the visible and infrared regions. With the use of pulsed light sources we thus have in PEEM a powerful and elegant method for work in nano-photonics across the whole visible and ultra-violet spectral ranges (\*23,\*24).

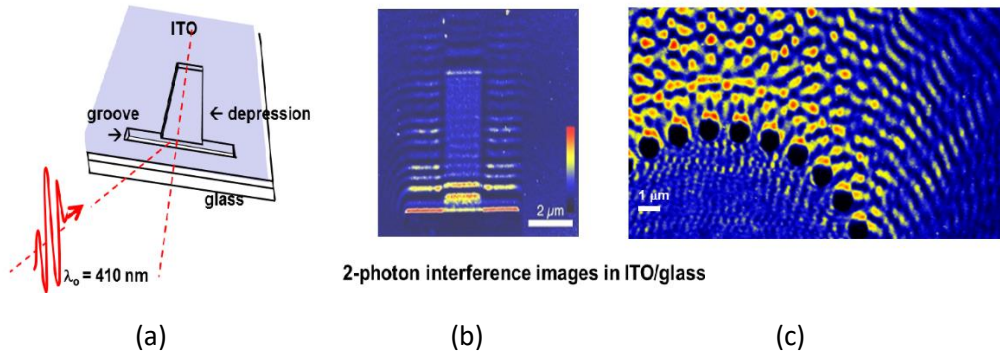


Fig. 3 a: Schematic arrangement for photonic measurements in PEEM. b: Simple diffraction pattern obtained in PEEM in a linear waveguide. More complex pattern obtained from an array structure.

In our work, a new imaging approach based on nanoscale diffraction patterns has been explored (\*14,\*19) that appears very promising in nano-photonics. Fig. 3 shows basic examples of such light diffraction patterns in thin transparent films on a glass substrate. The films act as waveguides and the shown patterns are due to the superposition of guided waves scattered in the transparent film and incident light falling directly on to the film. Diffraction near surfaces can elegantly be analyzed with the use of diffraction-integrals as developed by Kirchhoff, Fresnel and others. We showed that such an analysis provides a direct and quantitative insight into many optical properties, such as propagation speeds, phase shifts, loss factors etc. (\*13,\*15,\*19). Numerical calculations and simulations can further be used for understanding experimental results in great detail (\*19 -\*23).

We have used such interferometric approaches for the study of photon and surface-plasmon dynamics (\*4-\*6, \*11,\*12,\*22,\*28,\*31). While in the initial work we were interested in working out the experimental methodology and mathematical approaches, we later used these methods to explore photon and plasmon dynamics in novel nanostructures.

Using nano-meter sized transparent optical waveguides we evaluated in detail phase shifts at the waveguide surfaces and their effect on energy distributions at the boundaries of waveguides (\*27,\*30,\*35). We demonstrated a direct visual determination of the so-called Goos-Hanchen shift, a lateral shift occurring in total reflection (10) and a fundamental parameter allowing the determination of power flow in the evanescent region in waveguides and metamaterials ( ). We also used our PEEM for the characterization of photonic crystal waveguides. Using line-defects in thin-film photonic crystals, we demonstrated that interferometric imaging can directly determine the group index dispersion in the bandgap region of the photonic crystals (\*30). As the group index is a direct measure of the light propagation speed, the method offers a convenient way to determine signal and energy propagation speeds in photonic crystals. In particular it allows to characterize what is known as "slow" light, as indicated in Fig. 4.

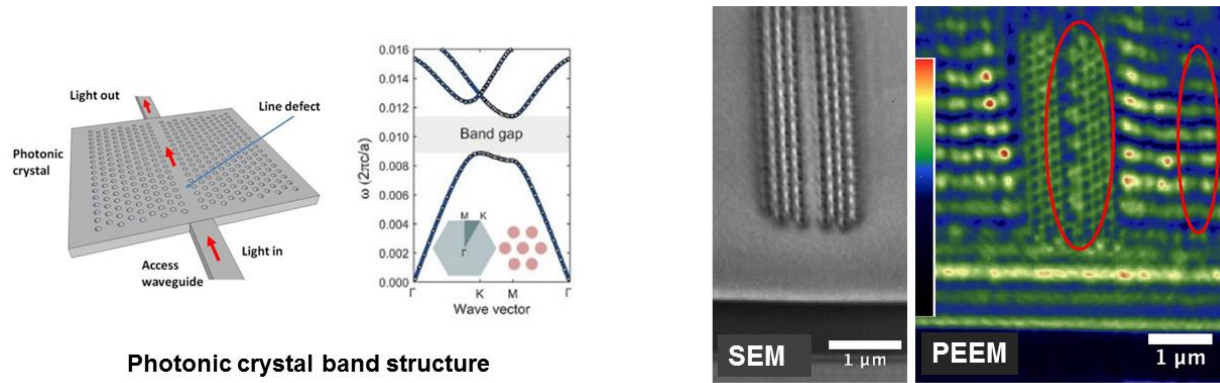


Fig. 4 a: Photonic crystal and waveguide design. b: dispersion curves in a hexagonal photonic crystal. c: SEM image of the planar photonic crystal, and d: PEEM image allowing the characterization of light propagation in the waveguide region.

In metallic plasmonic nanostructures the propagation of surface plasmon-polaritons is restricted to the surface, and photoemission microscopy and spectroscopy can be applied in similar ways as in thin film optical waveguides (8). In the third portion of the project we have addressed the visualization of plasmon propagation (\*28,\*31) and localization (\*4,\*5,\*22) in nanostructures such as optical antennas, waveguides and other basic devices.

Combining high resolution with the possibility of varying the light incidence angle (\*28), we were able to analyze the excitation dynamics and eigenmode structure of planar optical patch antennas across the entire visible wavelength region (\*26). The plasmonic eigenmodes in these antennas are strongly sensitive to surface properties and geometries. Particularly in small thin structures top and bottom planar plasmon modes as well as ring modes along the perimeter are known to interfere and hybridize, giving rise to design challenges and operation of these structures. In a detailed study with PEEM we were able to analyze some of the main properties of these antennas and discuss in detail the effects of asymmetries in the eigenmode distributions (\*28).



We have also utilized interferometric imaging to design and then operate new fundamental device structures such as surface-plasmon routers (\*11), plasmon resonance localizers (\*18) and filters (\*12). We were able to show in many cases that these devices can successfully operate at sub-micron size and that they are fully operational down to the scale of the excitation wavelengths. Furthermore, we showed that polarization tuning offers unique and efficient ways to control many of the device functions. Fig. 5 gives an illustration of this work where a polarization-controlled sub-micron surface-plasmon router structure is shown.

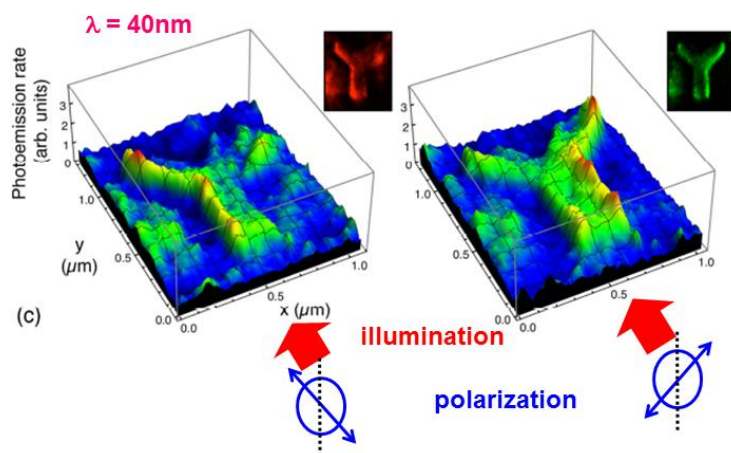


Fig. 5: Map of surface-plasmon intensity in a Y-structure milled into a single-crystalline gold platelet. The images show the photoelectron yield for two different polarization angles of the femtosecond excitation light. A routing effect in this 1μm device is clearly discernible.

## Conclusions

Our work demonstrated markedly improved spatial resolution in PEEM. Resolution improvements for continuous as well as pulsed excitation were obtained. Our results were the first to show spatial resolution improvement obtained in a mirror corrector system that compensates for spherical and chromatic aberrations at the same time. Work on inorganic as well as organic specimens showed resolution of ~5.5nm. Our best to date resolution is 4.7nm.

New applications of PEEM in photonics and plasmonics were explored. We could demonstrate imaging with nanometer resolution using light excitation throughout the visible region and with femtosecond frame times. Transparent nanostructures, photonic crystals and numerous fundamental plasmonic structures were characterized in detail. The work in plasmonics successfully explored resonant and non-resonant antenna structures, eigenmode distributions, hybridization and the use of polarization control for novel device structures and functions.

The insight from our work is twofold: The microscope development demonstrates that aberration-correction with mirrors is a viable option in electron optics that will be of particular interest for low

electron energies. This should be of principal interest for SEMs, low-voltage TEMs and for Focused Ion Beams.

Our work on methodology illustrates the applicability of PEEM in the visible and even infra-red spectral region based on multi-photon excitation in femtosecond time pulses. With spatial resolution of a few nanometers PEEM is a highly competitive tool in the nano-optics field as the imaging is direct, i.e. without a scanning process and without the need of a local probe. These advantages are of particular interest for work in the optical near-field regime in photonics and plasmonics.

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