

## Visualization of Optical Wave Propagation in Femtosecond Photoemission Electron Microscopy

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Photoemission electron microscopy (PEEM) combines in a unique way photon probing and electron imaging. The imaging electrons are generated in a photoelectric process by illuminating the sample with ultra-violet or x-ray light. After generation these electrons are accelerated and introduced into an electron-optical system to produce a microscopic image of the specimen surface. The advantages of PEEM are manifold:

The avoidance of electron beam exposure makes PEEM a much gentler method than standard electron microscopy. This advantage is important when fragile organic or biological structures are studied. PEEM is also highly surface sensitive, since the photoelectrons typically escape the sample from a depth of only a few nm. As a consequence PEEM naturally probes nano-volumes – even without extensive sample prep. The photon-based excitation process allows a large arsenal of optical spectroscopies to be utilized in the microscope. Specific initial states can be selectively probed by adjusting photon energy, momentum and polarization selection. This allows a detailed mapping of electron energy and momentum. Ground state as well as excited state properties can be studied.

As photon pulses can be made very short, PEEM also allows high time-resolution. Femtosecond imaging can be obtained in PEEM with visible light excitation [1]. As short photon pulses allow extremely high excitation rates, non-linear optical and electronic processes can be visualized and studied.

Here we utilize femtosecond pulsing in an aberration-corrected photoemission microscope to visualize the propagation dynamics of optical waves in micrometer scale optical waveguides. By applying an interferometric technique [2] we show that a quantitative description of the wave propagation can be obtained. We show that optical scattering processes, absorption losses, phase shifts and mode formation can be observed on sub-micron length scales. We use a 100nm thick indium-tin-oxide film on glass substrate as the optical waveguide and 410nm light for probing. At this wavelength the ITO is nearly transparent and the wave propagation can be observed over several 10 micrometers. The spatial resolution in these experiments is approximately 20nm, i.e.  $\lambda/40$ .

The light pulses are coupled into the ITO film through vertical holes that are milled into the planar film with a focused ion beam. We use a linear groove in the ITO to excite planar propagating waves, and we use various assemblies of circular holes to generate more complicated wave patterns. The optical waves in the ITO layer interfere with a coherent portion of the laser pulse that travels through vacuum. Its propagation direction is at an angle of 60 degrees from the optical waveguide surface normal as shown in Figure 1.

The resulting interferogram can directly be observed in PEEM, since the electron emission rate is a function of the photon densities and field strengths at the film surface. The interferogram thus produces a micrograph that can be quantitatively evaluated. Using spatial Fourier analysis, the dominant modes in

the waveguide can accurately be determined. Typically we find 6-10 separate modes corresponding to the various polarizations, propagation directions and frequencies. We find forward moving and reflected modes, and – at high pulse intensities – coupled modes and higher harmonic modes.

When plasmonic materials are brought into the vicinity of the waveguide we are also capable of imaging a plasmonic response and an interaction between guided photonic modes in transparent waveguides and plasmonic modes in metals. The capability of directly imaging optical and plasmonic dynamics with a high spatial resolution makes PEEM [3, 4] an interesting tool in the study of integrated photonic circuits.

#### Experimental details:

The microscopy was performed in a 20 kV PEEM. The microscope was equipped with a home-built electron mirror for chromatic and spherical aberration correction [5]. A frequency-doubled Ti:sapphire laser at a wavelength of 410 nm with a pulse length of 80-100 fs at 80 MHz was used as a light source. Laser polarization was set using a tunable waveplate. ITO films with typical resistances of  $15 \Omega/\square$  and optical transparency of 85% in the visible region were obtained from SPIE Supplies Inc. These films were structured with an FEI dual beam focused ion beam machine.

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[3] R. Könenkamp et al. *Ultramicroscopy*, **110** (2010) p. 899

[4] RC Word et al., *Surf. Sci.* **607** (2013) p. 148.

[5] R. Könenkamp, et al. *Appl. Phys. Lett.* **101** (2012) p. 141114.

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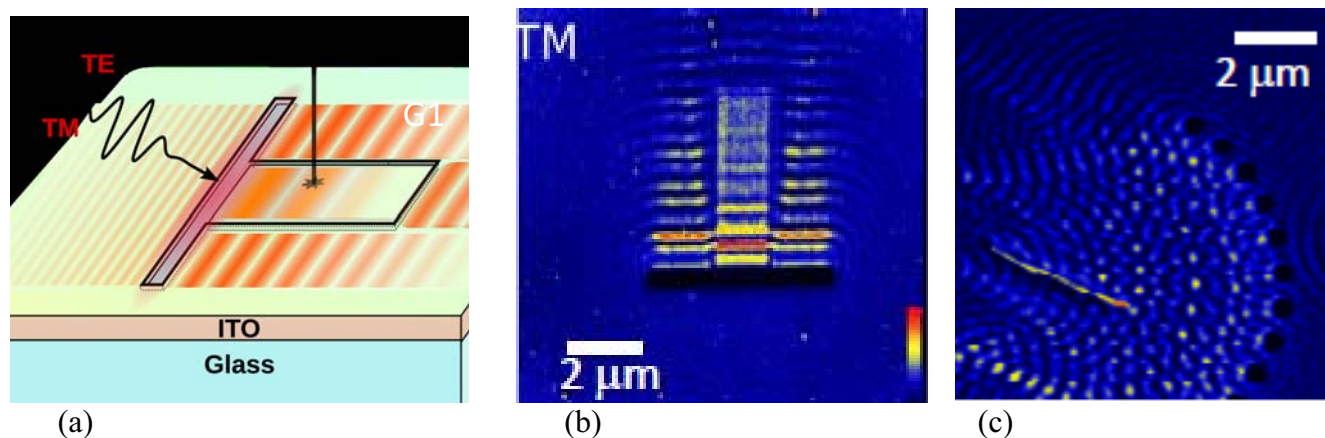


Figure 1. (a) Illumination and electron emission scheme for interferometric PEEM on a photonic ITO waveguide. (b) Experimental results showing wave propagation in the ITO waveguide excited by diffractive coupling in a vertical slit (black). The laser beam direction is from bottom to top. (c) Gold nanowire lying in the wave field of a circular hole assembly (black) in an ITO waveguide. Plasmonic excitation of the nanowire can be inferred from a detailed analysis.