

# Electron energy-gain spectroscopy of optical excitations in integrated photonic structures

Jan-Wilke Henke<sup>1,2</sup>, Dr. Yujia Yang<sup>3,4</sup>, F. Jasmin Kappert<sup>1,2</sup>, Dr. Arslan S. Raja<sup>3,4</sup>, Germaine Arend<sup>1,2</sup>, Guanhao Huang<sup>3,4</sup>, Dr. Armin Feist<sup>1,2</sup>, Zheru Qiu<sup>3,4</sup>, Rui Ning Wang<sup>3,4</sup>, Aleksandr Tusnin<sup>3,4</sup>, Dr. Alexey Tikan<sup>3,4</sup>, Prof. Dr. Tobias J. Kippenberg<sup>3,4</sup>, Prof. Dr. Claus Ropers<sup>1,2</sup>

<sup>1</sup>Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany,

<sup>2</sup>University of Göttingen, 4th Physical Institute, Göttingen, Germany, <sup>3</sup>Institute of Physics, Swiss Federal Institute of Technology Lausanne (EPFL), Lausanne, Switzerland, <sup>4</sup>Center for Quantum Science and Engineering, Swiss Federal Institute of Technology Lausanne (EPFL), Lausanne, Switzerland

## Background

Optical shaping of electron beams, e.g. in the form of longitudinal attosecond bunching that promises increased temporal resolution [1], significantly extends the range of experiments possible in transmission electron microscopes (TEM). Based on inelastic scattering with an optical field, the momentum and energy of the electron are modified by absorption or emission of photons [2]. This inelastic interaction can, in turn, be employed to investigate the nano-optical response of samples with high spatial resolution in photon-induced near-field microscopy (PINEM) [3]. However, due to the weak coupling of free electrons and photons, inelastic field probing and beam shaping techniques so far required intense optical pulses and short electron pulses available in ultrafast TEMs.

## Methods & Results

Here, we present the efficient modulation of a continuous electron beam by integrated photonics microresonators made from silicon nitride (Si<sub>3</sub>N<sub>4</sub>) that are optically pumped with a continuous-wave (CW) laser [4]. The fiber-coupled, chip-based resonator is placed inside a TEM, as illustrated in Figure 1a, such that the continuous electron beam can pass over the chip parallel to its surface before being analysed with an imaging spectrometer. Swift electrons interacting with the resonator's guided optical mode can absorb or emit photons from the laser field coupled to the resonator. This leads to the formation of electron energy sidebands spaced by the photon energy (~0.8eV, corresponding to ~1550nm) in the spectrum as shown in Figure 1b. The inelastic electron-light scattering is facilitated by the velocity matching of the electrons to the optical phase velocity as well as the high-Q resonant field enhancement. We characterise the latter by employing electron energy-gain spectroscopy (EEGS). To this end, the frequency of the CW pump laser is scanned across the cavity resonance at a low input power while electron spectra are recorded in parallel. We retrieve the laser detuning-dependent electron-light coupling strength (Fig. 1c) that exhibits a linewidth of 390 MHz

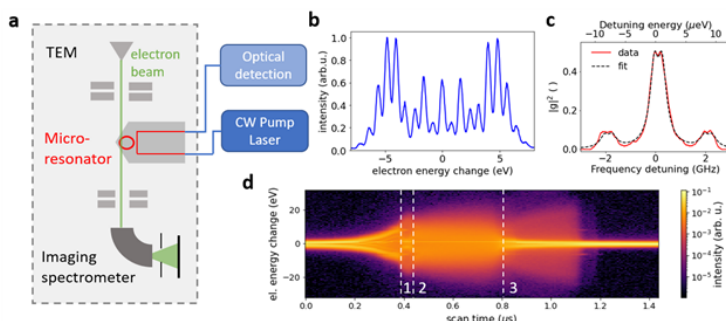
corresponding to a spectral feature of only  $3.1\mu\text{eV}$  width. From this EGS trace, we infer a cavity quality factor of  $7.7 \times 10^5$ .

Increasing the optical pump power coupled to the microring resonator, the inherent nonlinearity and anomalous dispersion cause the parametric generation of new optical frequencies via four-wave mixing. We observe the formation of various nonlinear optical intracavity states whose spectral and temporal properties strongly depend on the laser detuning from the cavity resonance frequency. When scanning the laser across the resonance at a power  $> 100\text{mW}$ , we can thus perform an EGS measurement on the nonlinear optical states by recording electron energy spectra in parallel [5]. The resulting electron spectral trace, shown in Figure 1d, exhibits prominent changes when entering different nonlinear optical states (marked by dashed white lines). For stable and chaotic intensity modulations (regions 1 and 2), resulting from the superposition of different optical wavelengths, averaging over different instantaneous interaction strengths leads to a smoothing of the electron spectra. However, the interaction of electrons with dissipative Kerr solitons (region 3), self-stable short optical pulses with a broad spectrum, yields a broad, low-intensity plateau and a strong central peak since only a fraction of electrons interacts with the high-intensity pulse and scatters to high energy changes.

## Conclusions

In conclusion, we characterise the inelastic interaction between electrons and the optical mode of an integrated photonics microresonator. By performing EGS on one of the cavity resonances, we achieve an unprecedented energy resolution that might be transferred to both the study of material excitations as well as the probing of quantum optical excitations with free electrons. The observed strong interaction of a continuous electron beam with a low-power CW laser, moreover, enables efficient longitudinal electron beam modulation with optical fields in a conventional TEM setup. Harnessing the toolbox of optical waveform shaping in integrated photonics, we employ the multicolour fields of optical frequency combs and their impact on the electron energy spectra upon interaction to further extend these beam-shaping capabilities.

**Graphic:**



**Figure 1:** a) Schematics of the experimental setup. The fiber-coupled microresonator chip is placed inside the transmission electron microscope (TEM) and pumped with a CW laser. The electrons pass over the resonator surface and interact with the optical mode. Changes to the electron energy distribution are measured using an imaging spectrometer. b) Electron energy spectrum after interaction with a resonator mode pumped with a CW laser. c) EEGS trace of a weakly pumped cavity mode recorded with a frequency-tuneable laser and exhibiting a linewidth of 390 MHz, corresponding to  $3.1\mu\text{eV}$ . d) At higher optical input power, the EEGS trace shows distinct changes to the electron spectrum when entering different nonlinear optical regimes (marked by white dashed lines).

**Keywords:**

EEGS, UTEM, Inelastic Electron-Light Scattering

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