# Probing nonlinear optical processes with electron energy-gain spectroscopy

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Ultrafast transmission electron microscopy enables the investigation of optical properties on the nanoscale via inelastic scattering between free electrons and light [1,2]. Combining integrated photonic structures with electron microscopy allows for a significant boost of the underlying interaction strength [3], facilitating the exploration of free-electron quantum optics [4-6]. At the same time, electrons can be employed to characterize the electric near-field and the inherent nonlinear optical properties of the microresonators [7], which find application frequency metrology to optical sensing.

Here, we investigate the characteristics of the nonlinear dynamics inside a chip-based, high-Q silicon nitride microresonator using inelastic scattering of free electrons at the resonator's optical near-field [8]. We measure the electron energy distribution after the interaction using a dispersive spectrometer. Benefiting from the strong field enhancement in the resonator structure and electron-light velocity phase matching, significant interaction strength can be achieved for a continuous electron beam interacting with a continuous-wave laser. When scanning the pump laser frequency at low optical input power across one of the cavity resonances in an electron energy-gain spectroscopy (EEGS) measurement, we can determine the resonance linewidth from the wavelength-dependent electron spectra with the characteristic double-lobed shape of coherent interaction [3]. At higher optical pump powers, four-wave mixing leads to the transient generation of nonlinear optical comb states comprised of multiple frequencies. The pronounced differences between the various optical states, ranging from stable intensity modulations to chaotic modulation instabilities, result in characteristic changes to the electron spectrum. The most prominent features arise from electrons interacting with dissipative Kerr solitons [7], self-stable short pulses with a broad optical spectrum. This results in two main features: a low-intensity spectral plateau of electrons scattered to high energy changes at the high-intensity pulse and a sharp spectral peak for unscattered electrons.

Thus, our work extends the interaction of free electrons and light to the multicolor regime by employing optical microcombs. This enables new schemes in electron beam modulation and manipulation, while also demonstrating the capabilities of electron-based probing of complex optical fields. In the future, these advanced capabilities may be extended to the sensing of different quantum states of light.

## References

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