Controlling free electrons with optical whispering-gallery modes

Free-electron beams are versatile probes of microscopic structure and composition\(^1,2\), and have revolutionized atomic-scale imaging in several fields, from solid-state physics to structural biology\(^3\). Over the past decade, the manipulation and interaction of electrons with optical fields have enabled considerable progress in imaging methods\(^4\), near-field electron acceleration\(^5\,6\), and four-dimensional microscopy techniques with high temporal and spatial resolution\(^7\). However, electron beams typically couple only weakly to optical excitations, and emerging applications in electron control and sensing\(^8\,10\) require large enhancements using tailored fields and interactions. Here we couple a free-electron beam to a travelling-wave resonant cavity mode. The enhanced interaction with the optical whispering-gallery modes of dielectric microresonators induces a strong phase modulation on co-propagating electrons, which leads to a spectral broadening of 700 electronvolts, corresponding to the absorption and emission of hundreds of photons. By mapping the near-field interaction with ultrashort electron pulses in space and time, we trace the lifetime of the microresonator following a femtosecond excitation and observe the spectral response of the cavity. The natural matching of free electrons to these quintessential optical modes could enable the application of integrated photonics technology in electron microscopy, with broad implications for attosecond structuring, probing quantum emitters and possible electron–light entanglement.

The acquisition of laser-driven electron energy gain spectra (EEGS) yields spatial and spectral mode maps in nanoplasmonics\(^19\,22\) and photonic crystals\(^23\). Near-field electron microscopy, with broad implications for attosecond structuring, probing quantum emitters and possible electron–light entanglement, motivates the use of these structures for optical modes could enable the application of integrated photonics technology in electron microscopy, with broad implications for attosecond structuring, probing quantum emitters and possible electron–light entanglement.
In this study, we focus on two scenarios using microspheres with diameters of 2 μm and 5 μm. The WGMs in the smaller spheres allow us to reach an extremely strong and coherent modulation of the electron wavefunction, which is manifest in the emergence of electron energy sidebands of 220 eV and 700 eV for highly chirped picosecond and moderately chirped femtosecond optical pulses, respectively. For the larger spheres, the smaller free spectral range allows us to analyse the coupling of multiple WGMs to electrons and to trace the optical cavity ring-down time. These results establish the WGM as a key element in future approaches to coherently manipulate free electrons. Moreover, we expect that WGM-enhanced electron spectroscopy has the potential of ultimately facilitating controlled coupling to individual quantum systems.

The experiments are conducted in an ultrafast TEM (UTEM) featuring electron pulses as short as 200 fs (see illustration in Fig. 1a and details in Methods). These electron pulses are passed near the surface of optically illuminated silica microspheres. The WGMs are excited by optical pump pulses with a centre wavelength of 800 nm. The similar velocities of the electron and WGM phases allow them to efficiently exchange energy through the evanescent light field that permeates vacuum (Fig. 1a inset).

Figure 1b displays a spatial map of the electron–WGM interaction strength, with the colour code representing the width of the electron energy spectrum. The spectrum substantially broadens near the surface of the sphere and, to a lesser extent, near the metallic edges of the support structure (see Methods). The interaction strength is evident along wide sections of the sphere circumference, and is suppressed only in regions that are approximately perpendicular to the incident horizontal polarization of the light. Because the excited transverse profile of a single WGM spans approximately one wavelength, we infer from the wider azimuthal distribution of the interaction map that the driving laser excites multiple modes.

We observe very similar halo-shaped interaction maps on multiple individual spheres and assemblies, with overall magnitudes depending on the specific geometrical arrangement and in-coupling conditions. An azimuthally selective excitation of a WGM is achieved in structures with substantial geometrical anisotropy, such as the doublet displayed in Fig. 2a. Here, a pair of 2-μm-diameter spheres is partially merged along an axis perpendicular to the edge of the supporting copper grid (see Fig. 2a). For this structure, the interaction strength is maximized when the pump excites a WGM with transverse magnetic polarization (TM), that is, for polarization along the doublet axis. The electron energy gain and loss via the PINEM are driven by the polarization component parallel to the electron path. The spatial dependence of the resulting electron bandwidth in Fig. 2b shows very good agreement with the expected PINEM interaction for a single TM-polarized WGM, as simulated in Fig. 2c. Specifically, both the azimuthal extent of about one wavelength and the exponential radial decay with a characteristic length of $L_{\text{decay}} = 0.1 \mu m$ are reproduced. The experimental evaluation of the decay length is based on a line scan, for which the spectral bandwidth was estimated at each point as a function of its distance from the surface (orange circles in Fig. 2d). It should be noted that the radial dependence of the measured coupling strength is a combination of the mode decay and the phase-matching condition. The circular propagation of the WGM implies that the linear phase velocity increases with the radius; thus, at larger radii the WGM field acquires an increasing velocity mismatch to the electrons. This radially dependent phase matching reduces the observed decay length to 100 nm from a mode decay length of 260 nm. The individual curves in Fig. 2d are spectra recorded for a set of fixed distances, which visualize the broadening of the spectrum near the sphere; the spectra are scaled vertically for clarity. Close to the sphere’s surface, the spectral bandwidth extends to a remarkable 220 eV while keeping the double-lobed shape that is characteristic of

**Fig. 1** Experimental scheme. a, A laser-driven WGM circulating a microsphere drives stimulated gain and loss in a traversing electron, resulting in an exceptionally broad energy spectrum spanning hundreds of sidebands. Inset, efficient transfer of energy and momentum is enabled by matching the velocities of the electron, $v_e$, and the wave in the optical cavity, $v_p$, within the evanescent modal field. b, Spatial map of interaction strength for two neighbouring spheres with linearly polarized excitation in the UTEM, quantified in terms of electron spectral bandwidth. c, Scanning electron micrograph of the two spheres on the support structure.

**Fig. 2** Electron spectral broadening induced by WGMs. a, An annular dark-field image of a microsphere doublet comprising a pair of 2-μm-diameter spheres that are optically pumped from the top (into the page plane). b, Measured electron bandwidth as a function of position. c, Simulated electron bandwidth from the coherent interaction with a single WGM. a.u., arbitrary units. d, Electron spectra measured at distances of 0 nm, 30 nm, 60 nm, 100 nm, 170 nm, 220 nm, 290 nm and 380 nm from the surface of the sphere (shifted accordingly and scaled vertically for clarity). The electron bandwidth reaches 200 eV at the surface (bottom curve; see text). The scanning line is annotated on the right inset. The decay length of the exponential fit (solid lines) of the measured interaction strength (red circles), $L_{\text{decay}}$, agrees with the calculated length scale (see Fig. 2c). Pumping the sphere with a shorter pulse extends the electron bandwidth to 700 eV (left inset), albeit at a reduced purity.
This case, increased values of $\Delta g$ is highly relevant in any coherent use of the modulated electron $\Delta g$ of the electron beam, as well as from the spatial and temporal profiles, $\Delta \text{photon energy}$. In practice, however, one has to account for some variations of the coupling strength across the electron energy, and thus the phase modulation imprinted on the electron wavefunction is uniform and deterministic\(^\text{24}\). Replacing the SF6 bar with a shorter BK7\(^\text{bar}\) results in an optical pump duration of 400 fs (see Methods for details). Thus, the strength of the optical field is increased, albeit at the cost of some variation of the coupling strength across the electron pulse duration. The top-left inset of Fig. 2d shows an electron spectrum with a considerably wider bandwidth of 700 eV, far beyond what is typically observed in PINEM\(^\text{24,30}\). By comparing this measured energy gain (350 eV) with the calculated WGM field, we estimate a peak acceleration of 1.4 GeV m\(^{-1}\).

To quantify the strength and temporal uniformity of the interaction, we fit the experimental data to the expected PINEM spectrum\(^\text{24,41}\). The interaction parameter, $g$, also referred to as the coupling or Rabi parameter\(^\text{24}\), provides the transition amplitude between adjacent electron energy states, and thus quantifies the spectral distribution of the electron beam after the inelastic interaction with the laser field. For a uniform interaction strength, the electrons are in a pure state, fully determined by the Rabi parameter. The probability $P_1$ for a total gain or loss of $k$ quanta of the photon energy in such a state is $P_1 = |j_k(2g)|^2$, where $j_k$ is the Bessel function of the first kind\(^\text{24,30}\). In the limit of $g \gg 1$, the electron energy width (or bandwidth) scales as $4|g|$, times the optical photon energy. In practice, however, one has to account for some variations, $\Delta g$, of the interaction strength that originate from the finite size of the electron beam, as well as from the spatial and temporal profile of the laser field. These averaging effects can be described by an incoherent sum of spectra calculated with a normally distributed coupling parameter with a standard deviation of $\Delta g$. The relative uncertainty, $\Delta g/g$, quantifies the deviation from a pure electron state. This parameter is highly relevant in any coherent use of the modulated electron beam, such as the formation of attosecond electron pulse trains\(^\text{27-29}\). In this case, increased values of $\Delta g/g$ will result in unwanted broadening of the attosecond electron pulses. For the measurements in Fig. 2d, we evaluated the relative uncertainty of the interaction strength for the 220- and 700-eV-wide spectra as $\Delta g/g = 0.09$ and $\Delta g/g = 0.33$, respectively.

To characterize the resonance properties in the interaction with the WGMs, we utilize larger spheres with a diameter of 5 μm (Fig. 3a), for which the optical bandwidth of our laser covers several discrete cavity modes. We first study the temporal decay of the cavity field by measuring the electron spectrum as a function of time delay between the electron pulses and 50-fs optical pulse pumps (Fig. 3b). The simulated cavity response for a sphere with a diameter of 4.765 μm (dashed black line) agrees with the experiment, from which we identify the mode polarizations (TM or TE) and indices $\ell$. The colour maps present the PINEM-relevant azimuthal component, $E_\phi$, of the identified modes.

For the spectral mode analysis, we pump the microsphere with a strongly chirped pulse, mapping optical frequencies to arrival times (see details in Methods). The delay-dependent electron spectrum (Fig. 3c) now shows three broadened regions corresponding to distinct WGM indices. By comparing with the computed time-dependent cavity response for the chirped pulse (dashed line), we find that the strong features represent two TM modes with vacuum wavelength $\lambda_{\text{TM},22} = 804$ nm and $\lambda_{\text{TM},23} = 773$ nm. The weaker feature is attributed to a transverse electric (TE) mode at $\lambda_{\text{TE},23} = 790$ nm, which results from a lower field along the electron trajectory (see colour-coded mode profiles of the relevant out-of-plane field component, $E_\phi$). Aside from reproducing the general features of the measured spectra, the calculation also predicts small temporal oscillations, because the frequency sweep rate in the chirped pulse is only moderately slower than the cavity.

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**Fig. 3** Spectral and temporal properties of the interaction between free electrons and WGMs. **a.** Annular dark-field micrograph of spheres with diameters of 2 μm and 5 μm supported by a thin carbon film. The yellow circle marks the measurement position. **b.** Left, colour map of the electron spectrum (logarithmic scale) measured after a loading of WGMs by a 50-fs-long optical pulse at time $t = 0$ (see scheme at top). At later times, the electron is driven by light stored in the cavity. Right, the exponential decays of $g$ and $\Delta g$ (symbols) evaluated from each spectrum separately, and fits with a cavity lifetime of 260 fs (dashed lines) corresponding to a quality factor of $Q = 305$. **c.** WGM selectivity probed by the electron’s spectral response. Using a frequency sweep (chirped pulse; see scheme at top), three modes are identified by the increased electron bandwidth as the sweep crosses their resonance frequencies. The simulated cavity response for a sphere with a diameter of 4.765 μm (dashed black line) agrees with the experiment, from which we identify the mode polarizations (TM or TE) and indices $\ell$. The colour maps present the PINEM-relevant azimuthal component, $E_\phi$, of the identified modes.
ring-down. Stretching the optical pulse further would eliminate these oscillations. Similar oscillations are indeed discernible in some areas of the experimental spectrogram between $t = 0$ and $t = 1$ ps.

In conclusion, our work combines resonant cavity enhancement with electron–light phase matching to strongly drive electron–light transitions, resulting in a coherent electron spectral broadening spanning hundreds of photon orders. This is achieved by using WGM microresonators, prototypical dielectric structures with numerous applications in spectroscopy and sensing. Although the current study focuses on low quality factors and free-space excitation, it can readily be extended to chip-based microresonators with quality factors greater than 10$^7$ (ref. 42) and using phase-matched, high-ideality and fibre-pigtailed nanophotonic waveguides for excitation. Importantly, such an approach could also reach the strong-coupling regime, promoting the possibility for electron–photon entanglement.$^6$ Moreover, attosecond optical modulation of the phase and density of continuous electron beams in standard electron microscopes appears in reach. Such high-current electron beams dressed by light could transfer optical polarizations on nanometre-sized focal spots, possibly acquiring high-resolution spectroscopic information from resonator-coupled atoms$^{30,40,43}$, molecules$^{36}$ and nanoparticles$^{44}$. In a similar vein, continuous-wave probing of the phase response of resonators could enable all-optical real-time detection of electrons by a dispersive phase shift, in analogy to previous all-optical molecule detection.$^1$ The ability of microresonators to generate optical dissipative solitons may enable the coupling of electrons to tightly localized fields with lengths of only a few optical cycles$^{39}$, relevant for time-gated interaction. Generally, cavity-enhanced and phase-matched near-field electron interactions could allow a merging of electron microscopes and photonic chip-based microresonators, with far-reaching consequences in local quantum control and sensing.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-020-2320-y.

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The pump laser is an amplified Ti:Sapphire system (Coherent RegA) that provides pulses centred at a wavelength of 800 nm, with a bandwidth of 34 nm full-width at half-maximum (FWHM), a repetition rate of 600 kHz and an average power of 150 mW entering the TEM. For experiments with a highly chirped pumping laser pulse (3.5 ps FWHM), the beam passes through a 19-cm-long bar made of Schott dense flat glass, SF6, resulting in a group velocity dispersion (GDD) of approximately $3.8 \times 10^4$ fs$^2$. For the moderately chirped pulse (400 fs FWHM), we used a 10-cm-long bar of BK7 glass that provided a GDD of approximately $4.5 \times 10^4$ fs$^2$. In practice, we calculated the full dispersion relation of our pulse in the glass bars using the dispersion coefficients in ref. [4]. For short pulses (50 fs; see Fig. 3b), no glass bar was used in the beam path.

The optical beam is nearly co-propagating with the electron beam (6° off axis) and focuses down to a characteristic mode size of 10 μm FWHM. The timing between the electron pulse and the laser is controlled by a delay stage. The experiment (see illustration in Fig. 1a) investigates the electron interaction with WGMs circling inside silica microspheres with diameters of 2 μm and 5 μm (SSD5003 and SSD5000, respectively, Bangs Laboratories). For the sample preparation, the spheres are immersed in ethanol and are randomly distributed by drop-casting on a lacy-carbon support (ultrathin carbon/lacy support on 400 mesh, Ted Pella). The experiments use scanning TEM for a systematic acquisition of the electron spectrum and spectral bandwidth (Gatan Enfimium spectrometer) and an annular dark-field detector.

### Simulation details of the PINEM map in Fig. 2c

The calculation uses electrons accelerated to 200 keV that interact locally with a WGM in a 2-μm-diameter sphere having a vacuum wavelength of $\lambda = 806$ nm, which was found to be the mode closest to the laser wavelength using the WGMode toolbox. Furthermore, the WGMode toolbox was used to calculate the $E_x$ component of the electric field on the equatorial plane, from which the PINEM-relevant field in space and time was calculated by adding the temporal and azimuthal phase $e^{-i\omega t}$, where $\omega$ is the angular frequency of the mode and $\phi$ is the azimuthal angle. The contribution to the PINEM signal at each coordinate $(x, y)$ was integrated over the electron trajectory, $t(z)$, as $E_{\text{PINEM}}(x, y, z) = \int_{-\infty}^{t_{\text{max}}} E_x(x, y, z(t)) dt$, where $t_{\text{max}}$ is the electron charge. This dimensionless integral is the electron energy gain/loss, normalized by twice the photon energy. The effect of other components of the field is negligible. Although a large path is integrated numerically, the main contribution is from distances close to the sphere’s surface, near the maximal field amplitude, $E_{\text{max}}$. At this proximity, the field travels approximately as a phasor, $E_x = E_{\text{max}} e^{-i\omega t - i\phi R / \ell}$, where $R$ is the sphere’s radius. For a perfect phase matching, the electron’s velocity fits the WGM phase velocity at the surface, $v_{\text{el}} = v_{\text{WGM}}$. Near the equator, this simplified phase velocity term can be compared to phase-matching experiments with flat vacuum–dielectric interfaces and vacuum–grating interfaces and electron–nanoparticle interactions. The role of an imperfect phase matching is similar to energy conversion processes in nonlinear and extreme nonlinear optics. Accurate agreement is not observed, and the evaluation of the abovementioned modal decay length of $260$ nm was retrieved by an exponential fit to the electric field magnitude ($|E|$) for mode number $\ell = 8$ near the sphere’s surface. Farther from the sphere, the field is accurately described by the Hansen solutions of the spherical vectorial wave equation. The solution was calculated using the WGMode toolbox for 2-μm-diameter spheres with a refractive index of $n = 1.4533$ (ref. [4]). Figure 2c can be produced from the $E_x$ field component using a MATLAB script (see ‘Data availability’).

### Calculation of the expected spectral response of the WGM in Fig. 3c

The only fitting parameter for the expected WGM-driven electron spectrum (dashed line in Fig. 3c) is the sphere’s diameter, for which the best fit was found to be 4.765 μm. This value is only 2% smaller than the measured diameter. To evaluate the resonator response, we used the tabulated refractive index for fused silica, $n = 1.4533$ (ref. [4]) and the experimentally measured lifetime of 260 fs (see Fig. 3b). The measurement results in Fig. 3b suggest a uniform temporal decay for all the participating modes. This is reasonable because the decay is dominated by scattering, which is faster than the optical leakage rate.

The wavelength of the resonances was calculated using the WGMode package, and the relative strength of each resonance was based on the maximal field components, $E_x$, of the different modes. For the TM modes, $E_x$ is maximal at the centre, whereas for the TE mode, $E_x$ peaks at a slightly shifted position compared with the plane of circumference (see red and blue colour maps in Fig. 3c). The temporal dispersion of the pump is calculated using the measured bandwidth of our laser (34 nm) and the full chromatic dispersion in the long SF6-glass bar (see experimental details above). The arrival time of the central pumping wavelength ($\lambda = 800$ nm) was calibrated on the basis of PINEM experiments on metallic surfaces and was determined as time zero.

### Data availability

The data supporting the findings of this study are available within the paper and at the Open Science Framework repository at https://osf.io/5da5g/?view_only=779f3a157219431bb3e48bc3d4fd7747. Source data for Figs. 1–3 are provided with the paper.

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Acknowledgements We acknowledge the joint effort of the UTEM team in Göttingen and especially the support of M. Möller, H. J. Gaida, T. Danz and T. Domroese. We thank J. Liu for productive discussions. O.K. gratefully acknowledges funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement number 752533. T.R.H. acknowledges the support of a postdoctoral fellowship from the Alexander von Humboldt Foundation and its sponsor, the German Federal Ministry for Education and Research. This work was funded by the Deutsche Forschungsgemeinschaft (DFG) through the Collaborative Research Center ‘Atomische Skal Control of Energy Conversion’ (DFG-SFB 1073, project A05) and the Priority Program ‘Quantum Dynamics in Tailored Intense Fields’ (DFG-SPG 1840).

Author contributions O.K. conceived the experiment, C.R. directed the study, O.K. and H.L.-M. conducted the experiment with contributions from A.F. and T.R.H.; O.K., H.L.-M., G.S. and M.S. prepared the samples. O.K. analysed the data with contributions from A.F., O.K. and C.R. wrote the manuscript with contributions from T.K., H.L.-M., A.F. and M.S. and based on discussions with all authors.

Competing interests The authors declare no competing interests.

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Peer review information Nature thanks Nahid Talebi and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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