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Phase-locked photon-electron interaction without a laser

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Abstract – Ultrafast electron-photon spectroscopy in electron microscopes commonly requires ultrafast laser setups. Photoemission from an engineered electron source is used to generate pulsed electrons, interacting with a sample that is excited by the ultrafast laser pulse at a specified time delay. Thus, developing an ultrafast electron microscope demands the exploitation of extrinsic laser excitations and complex synchronization schemes.

Here, we present an inverse approach based on cathodoluminescence spectroscopy to introduce internal radiation sources in an electron microscope. Our method is based on a sequential interaction of the electron beam with an electron-driven photon source (EDPHS) and the investigated sample. An electron-driven photon source in an electron microscope generates phase-locked photons that are mutually coherent with the near-field distribution of the swift electron. Due to their different velocities, one can readily change the delay between the photons and electrons arriving at the sample by changing the distance between the EDPHS and the sample. We demonstrate the mutual coherence between the radiations from the EDPHS and the sample by performing interferometry with a combined system of an EDPHS and a WSe2 flake. We assert the mutual frequency and momentum-dependent correlation of the EDPHS and sample radiation, and determine experimentally the degree of mutual coherence of up to 27%. This level of mutual coherence allows us to perform spectral interferometry with an electron microscope.

Our method has the advantage of being simple, compact and operating with continuous electron beams. It will open the door to local electron-photon correlation spectroscopy of quantum materials, single photon systems, and coherent exciton-polaritonic samples with nanometric resolution.
With the advent of ultrafast electron microscopy\textsuperscript{1,2}, visualizing the photoinduced dynamics in materials such as of magnetic vortices\textsuperscript{3} and chemical reactions\textsuperscript{4} has become possible at unprecedented spatial resolution. Particularly the ability to track the ultrafast dynamics of localized and propagating plasmons\textsuperscript{5} in nano-optical systems as well as phonon polaritons in quantum materials\textsuperscript{6} has recently boosted the application of ultrafast electron microscopy in the form of photon-induced near-field electron microscopy (PINEM)\textsuperscript{7}. Moreover, PINEM has evolved into a unique tool for tailoring the quantum-path interferences in an extremely controllable system of single-electron wavepackets interacting with optical near fields, prepared in either classical or quantum states\textsuperscript{8-12}. Combining real- and reciprocal-space information with elastic and inelastic processes in diffraction and electron energy-loss spectroscopy, full information about the fundamental aspects of electron-photon interactions is obtained, either in transmission or scanning electron microscopy\textsuperscript{13-16}.

In a PINEM setup, pulsed electron beams are commonly generated by virtue of the photoemission process: An ultrafast laser pulse is used to excite the apex of a sharp tip or other forms of cathodes, generating an electron pulse with a specific degree of spatial coherence, which depends on the electron source. A second laser pulse is then used to coherently induce a polarization in the sample at a certain delay with respect to the electron pulse. The stimulated interaction of the electron pulse with the laser-induced near-field excitations leads to the predominantly longitudinal modulation of the electron beams. The near-field zone of the sample hence mediates the transfer of energy and momentum from the coherent laser beam to the sample, where the strength of the photon-electron interactions is controlled by the synchronicity between the near-field excitation and the moving electron wavepacket\textsuperscript{17-20}.

Yet, a visionary application of ultrafast electron microscopy is to coherently control the material electronic excitations. Due to the high spatial resolution of electron-beam-based characterization techniques, electron beams could be used to coherently drive individual quantum systems to higher states\textsuperscript{21} or to probe strong-coupling effects\textsuperscript{22} or atomic Floquet dynamics\textsuperscript{23} in two-level or multi-level quantum systems. Combined with mutually coherent radiation sources, quantum walks on quantized states of a quantum system, such as quantum dots, defect centers, and excitonic systems, could be coherently controlled, by initiating a set of quantum-interference paths. Coherent control methods thus require a drastic improvement of the ultrafast electron microscopy setups, to enhance the mutual coherence between the photons and electrons, such that spectral phases can be retrieved. The latter is crucial for example for retrieving quantum interference effects.

To improve the mutual coherence between photon and electron excitations in an electron microscope, we here propose and experimentally realize a proof-of-concept experiment for an inverse approach, based on intrinsic radiation emitted from the electron beam, rather than extrinsic laser radiation, to generate photons that are phase-locked to the near-field distribution of the swift electron. In our setup, an electron beam excites a nanostructured electron-driven photon source (EDPHS)\textsuperscript{24-26}, which generates well-collimated photon pulses (Fig. 1a and b), as shown by the angle-resolved cathodoluminescence (CL) pattern (Fig. 1c). The EDPHS consists of an array of nanopinholes in a 40 nm gold film deposited on a Si$_3$N$_4$ membrane created by focused ion-beam milling, with the hole radii gradually varying from 25 nm (holes in the inner rim) to 150 nm (holes in the outer rim). This allows for the generation of broadband photonic radiation (Fig. 1d)\textsuperscript{27,28}. The radially propagating surface plasmon polaritons induced by the impacting electrons scatter off the nanopinholes and radiate into the far-field in the form of a TM$_x$-polarized Gaussian wave (see Supplementary Note 1 for a full characterization of the EDPHS radiation).
Fig. 1. Spectral interferometry with an electron beam. (a) An electron moving at a kinetic energy of 30 keV interacts with an electron-driven photon source (EDPHS) that generates photons with a collimated Gaussian spatial profile. The delay $\tau$ between photon and electron beams arriving at the sample is controlled by the distance $L$ between the sample and the EDPHS. The energy-momentum distribution of the total scattered field from the sample will be detected and analyzed to specify the mutual correlation between the EDPHS and the sample radiation, as specified in the text. (b) SEM image of the combination of the EDPHS and sample at the distance of $L = 2 \mu m$. (c) Angle-resolved CL pattern of the EDPHS structure. (d) CL spectra of the EDPHS and the WSe$_2$ flake integrated over the whole momentum space. (e) Schematic illustration of three-dimensional data cubes (CL intensities versus the wave numbers ($k_x,k_y$) and wavelength ($\lambda$)) at selected delays between the EDPHS and the sample. The shown data at the forefront are angle-resolved CL maps of the total field at the filtered wavelength of 800 nm ± 10 nm. (f) CL spectra of the total field at $\theta = 45^\circ \pm 2^\circ$ and $\varphi = 100^\circ \pm 2^\circ$. Both (e) and (f) maps are acquired at indicated delay values of $\tau_1 = 129$ fs ($L = 19 \mu m$) and $\tau_2 = 143$ fs ($L = 21 \mu m$).

Due to the difference between the electron group velocity ($v$) and the vacuum speed of light ($c$), the delay between the electrons and photons arriving at the sample can be precisely controlled using a piezo stage, inserted inside the sample chamber of a scanning electron microscope (SEM). Notably, our setup has 6 degrees of freedom, allowing to move both sample and EDPHS structures independently by using two nano-positioning stages (see Supplementary note 2 and Fig. S2). Specifically, the delay can be described as $\tau = L(v^{-1} - c^{-1})$, where $L$ is the distance between the EDPHS and the sample. Given an electron kinetic energy of 30 keV in our experiments ($v = 0.328 c$), the delay can be varied in steps of 6.8 attoseconds by changing the distance $L$ in steps of 1 nm. Moreover, given the dynamic range of 6 mm for the piezo stage, the delay can be tuned within the range of 0 fs (corresponding to zero distance or touching point) to 40.8 ps.
For our proof-of-concept experiment, we use thin exfoliated WSe$_2$ flakes (80 nm thickness) placed on top of a holey carbon transmission electron microscopy grid. Fig. 1b shows the SEM image of the WSe$_2$ flake positioned blow the EDPHS, at a distance of $L = 2 \mu$m from the EDPHS. Belonging to the class of semiconducting transition-metal dichalcogenides, WSe$_2$ hosts energetically different A and B excitons at room temperature due to spin-orbit coupling, at energies of 1.68 eV ($\lambda = 738$ nm) and 2.05 eV ($\lambda = 605$ nm), respectively. It has previously been demonstrated that the excitons can strongly couple to the photonic modes of thin transition-metal dichalcogenide flakes$^{29,30}$, where this phenomenon is generally referred to as the self-hybridization effect$^{30}$ (Fig. S4). This coupling results in an energy splitting and opening of a bandgap, apparent in the dispersion diagram of the guided waves (Supplementary Fig. S5e), as well as the creation of lower polariton (LP) and upper polariton (UP) branches that become apparent in the CL spectrum (Fig. 1d)$^{31,32}$. Besides optics-based characterization techniques, electron beams can be used to probe the propagation dynamics and resulting spatial coherence of exciton polaritons in thin WSe$_2$ flakes using CL spectroscopy$^{33}$. Indeed, electron-beam spectroscopy has been applied intensively to investigate the interaction between excitons and photons or plasmons in hybrid structures$^{22,34,35}$. Here, we develop a CL-based technique that allows us to fully determine the amplitude and phase of the aforementioned excitations. More details about the exciton polariton excitations and probing them using CL can be found in Supplementary Note S3.

Using CL, the interference between the transition radiation with the scattering of the exciton polaritons from the edges of the flakes can be used to determine the phase constant of the exciton polaritons, i.e., the change in the phase per unit length of propagation of exciton polaritons. When a moving electron approaches the surface of a flake, an image charge is induced inside the flake that together with the electron forms a time varying dipole. Its annihilation, when the electron crosses the surface, causes ultrabroadband transition radiation. The strong exciton-photon coupling and resulting energy splitting are apparent from the CL spectra of the WSe$_2$ flake (see Supplementary Note S3 for more details about the CL of the WSe$_2$ flakes). Thus, the electron beam can excite both the LP and UP branches, whereas the EDPHS radiation can excite most efficiently the LP branch (Fig. 1d). The overall detected CL signal, is the superposition of the electron-beam induced and EDPHS-induced scattered field from the sample (Fig. 1a, bottom). The degree of mutual coherence between the elements of this superposition, is inferred from the results of an interferometry technique outlined here, and discussed step by step below.

The total momentum-resolved CL spectrum is thus rewritten as$^{36}$

$$\Gamma_{\text{CL}}(k_\parallel, \omega) = (4\pi \hbar k_0)^{-1} \left[ |\vec{E}_{\text{EDPHS}}(k_\parallel, \omega)|^2 + |\vec{E}_{\text{cl}}(k_\parallel, \omega)|^2 + \vec{E}_{\text{cl}}(k_\parallel, \omega) \cdot \vec{E}^*_{\text{EDPHS}}(k_\parallel, \omega) e^{i\omega t} + \vec{E}_{\text{cl}}(k_\parallel, \omega) \cdot \vec{E}_{\text{EDPHS}}(k_\parallel, \omega) e^{-i\omega t} \right]$$

where $\hbar$ is the reduced Planck’s constant, $k_0 = \omega/c$ is the free-space wave number of the light, $k_\parallel = k_0 \sin \theta = \left((k_0^2 + k_N^2)^{1/2} \right)$ is the parallel wavenumber, $\vec{E}_{\text{cl}}(k_\parallel, \omega)$ and $\vec{E}_{\text{EDPHS}}(k_\parallel, \omega)$ are the electron-induced and EDPHS-induced electric field components detected in the far field. Hence, by changing the distance between the EDPHS and the sample, the observed interference fringes in both frequency and momentum space can be controlled. The visibility of the interference fringes allows for the determination of the degree of mutual coherence between the EDPHS and electron-induced radiations. In addition, spectral interferometry is widely used to characterize the broadening of the ultrafast laser pulses. There,
due to the beaming characteristic of the laser pulses, one could only measure the spectra along the longitudinal direction ($\theta = 0$)$^{37}$. Here, despite the beaming characteristic of the EDPHS radiation, transition radiation and electron-induced radiation in general, scatter to relatively high polar angles. In contrast to collimated laser beams, the coherent electron-induced radiation pattern normally is a dipolar one$^{38}$ so that angle-resolved spectroscopy is required to fully capture the mutual coherence between EDPHS and sample radiation. Hence, the interference patterns in both momentum and energy space are characterized, and a three-dimensional energy-momentum data cube is acquired, in dependence of the delay $\tau$ between EDPHS and sample radiations. For example, by spectrally filtering the total radiation at $\lambda = 800 \text{ nm} \pm 10 \text{ nm}$, the interference maps in the momentum space were observed and analyzed (Fig. 1e). Similarly, by filtering the angular distribution of the detected CL signal around $\theta = 45^\circ \pm 2^\circ$ and $\varphi = 100^\circ \pm 2^\circ$, corresponding to $k_{\|} = 0.707 k_0$ and the azimuthal direction normal to the edge of the flake, the spectral interference fringes can be examined (Fig. 1f). Particularly, we notice that when the WSe$_2$ flake is excited with both the electron beam and the EDPHS radiation, the overall CL spectrum differs from an incoherent superposition of EDPHS and sample CL spectra. More importantly, the total CL angle-resolved maps and spectra vary with the distance $L$ between the EDPHS and the sample (Fig. 1e and f), and delay-dependent $k$-space or spectral interferences are observed. Noteworthy, such interference patterns are only observed when the EDPH radiation as well as the electron-induced polarization inside the sample show coherent radiation properties. Therefore, polariton excitations such as plasmon polaritons in the EDPHS and exciton polaritons of the sample could be used for examining the functionality of our approach.

Since the photon generation process relies on the electron-induced surface plasmon polaritons inside the EDPHS, we anticipate that the EDPHS radiation has a high degree of mutual coherence with respect to the evanescent field accompanying the electron. Direct proof of this hypothesis is performed by using subsequent interactions of the electron beam with two similar EDPHS structures, as shown in Supplementary Note S1 and the figures therein. In order to explore the mutual coherence of the EDPHS and the sample radiation, we analyze the dependence of the angle-resolved CL patterns on $L$, by choosing a WSe$_2$ flake as the sample (Fig. 2a). The mutual correlation function between the EDPHS radiation and the sample radiation is a function of both wavelength and momentum, as stated above. First, we analyze the correlation between the EDPHS and sample radiation by filtering the overall radiation at the carrier wavelength of the EDPHS radiation (i.e., $\lambda = 800 \text{ nm}$) and analyzing the angle-resolved radiation pattern. When only the electron beam excites the sample, specific interference fringes in the angle-resolved CL pattern are observed, due to the interference between the transition radiation and exciton polaritons (Fig. 2b) (see Supplementary Note S3). A drastic alteration of the interference fringes is observed when the EDPHS radiation and the electron beam simultaneously excite the sample. The momentum-distance interference fringes, are observed within the distance range of $L = 22 \mu\text{m}$ to $L = 40 \mu\text{m}$, (Fig. 2c, Fig. 2d, region $R_1$), and these interference fringes are altered by including the EDPHS radiation, which is determined by the temporal coherence of the generated EDPHS radiation and decoherence phenomena, due to the interaction of the superimposed EDPHS and the sample radiation with the environment. This latter effect is precisely the reason why the visibility of the interference fringes is diminished by further increasing the distance $L$ above 40 $\mu\text{m}$. Performing the measurements in finer steps, we are able to resolve the interference fringes versus the transverse angular momentum and distance $L$, demonstrating the high degree of mutual coherence between the EDPHS and the sample radiation. The interference fringes – within the fully coherent range - can be simulated using classical electromagnetism.
Fig. 2. Angle-resolved cathodoluminescence maps at the filtered wavelength of $\lambda = 800$ nm versus the delay $\tau$ between the EDPHS and the sample. (a) SEM image of the WSe$_2$ flake. The EDPHS beam size and the electron impact position are both depicted on the image. Angle-resolved CL maps of (b) the WSe$_2$ flake and (c) the combination of the WSe$_2$ flake and EDPHS at indicated distances between them ($k_x = k_0 \sin \theta \cos \varphi$ and $k_y = k_0 \sin \theta \sin \varphi$, where $\theta$ and $\varphi$ are the polar and azimuthal angles with respect to the sample plane). Electrons traverse the flake at a distance of 800 nm from the edge of the flake. (d) Measured $L-k$ CL map acquired at the azimuthal angle range marked in (c) with a purple triangle. Here, $k_{\parallel}^2 = k_x^2 + k_y^2$. The mutual spatial coherence between the EDPHS and CL radiation is demonstrated by the visibility of interference fringes, in the region $R_1$. $R_2$ denotes the region where the visibility of the interference fringes vanishes. All angle-resolved CL maps are taken at a wavelength of 800 nm. Dashed lines in the left panel indicate the distances at which the full angle-resolved maps were acquired as shown in (c) and at which the CL intensity-$k$ line plots are shown in (d). For complete visualization of the interference maps, see Supplementary Movie 1.

considering a realistic system of the EDPHS radiation and the sample (Fig. 3) (see Supplementary Notes S3 and S4 for details of the simulation method). In the simulation, a rectangular WSe$_2$ flake is considered that is sequentially excited by a swift electron at the kinetic energy of 30 keV and the EDPHS radiation (the previously performed simulation results, which includes the interaction of the electron beam with our EDPHS structure, is used (see Fig. S1)). The EDPHS-induced and electron-induced polarizations are superimposed at the corresponding delays (Fig. 3a and b) and the far-field radiation is obtained by projecting the field distributions from the near-field to the far field, using free-space Green’s functions. The delay between the electron-induced and EDPHS-induced polarization affects the total diffraction angle of the field. Instead of the superposition of two waves with the same propagation direction (as for the combination of two identical EDPHS structure (Supplementary Fig. S5)), a directional beam and dipolar field profile are superimposed. The latter radiation is caused by the transition radiation mechanism as an example. Thus, the interference patterns are highly momentum-dependent. The agreement between the
simulation and experimental results suggests that the superposition of the EDPHS and electron-induced scattered field from the sample underpin the experimentally observed interference patterns.

To better understand this effect, we propose a model to reconstruct the interference patterns using geometrical optics (Fig. 3d; lower right panel). For this, we consider possible beam paths that contribute to the far-field patterns as (i) the EDPHS radiation that is directly transmitted through the film, (ii) the EDPHS radiation that is scattered off the edge of the flake, (iii) the transition radiation, and (iv) the excitation of the exciton polaritons and their scattering from the edges. First, we notice that EDPHS excitation cannot directly excite the exciton polaritons, due to the momentum mismatch.

Fig. 3. Modeling the experimentally observed $k$-space interference fringes. (a) Simulated near-field distributions induced by a moving electron at the kinetic energy of 30 keV (top), and the EDPHS radiation (bottom), and (b) their superposition at two depicted delay times between the EDPHS radiation and the coming electron beam, at the $x$-$y$ plane located 5 nm above the sample (top row), and the $y$-$z$ cross section, cutting parallel to the electron beam trajectory at the electron beam impact position (bottom row). (c) (Left) Experimental results compared to the (middle) simulated CL intensity versus in the momentum – distance space (distance between the EDPHS and the sample, $L$, and the transverse momentum parallel to the shorter symmetry axis of the flake), compared to the analytical results (right) obtained using the (d) proposed model that considers the interference between a direct transmission of the EDPHS radiation through the film and its scattering from the edge, the transition radiation (TR), and the scattering of the excited exciton polaritons (EPs) from the edges. Contributions of the EDPHS and the electron-induced radiation are shown by green and red wavy arrows, respectively.
between the exciton polariton dispersion and that of the free-space light. The scattering of the EDPHS radiation from the edges results in the excitation of exciton polaritons and forms a standing wave pattern inside the film that also contributes to the scattered light from the edges (see Fig. 3a for a visualization of the standing-wave pattern and its scattering from the edges). In addition, the electron beam directly excites the exciton polaritons as well and also causes transition radiation (see Supplementary Note S3). The interferences between these four beam paths form the interference pattern depicted in Fig. 3d in the lower left panel, matching the simulated and experimental results. Minor disagreements are due to the fact that in the model, only scattering from two edges are included, whereas the experimental and simulation results include more scattering edges.

The model thus reproduces the measured pattern, further confirming the high degree of mutual coherence between the EDPHS and the sample radiation. Comparing the experimental results with the results of classical electromagnetic simulations, a degree of coherence of 27% is inferred. Increasing the spatial distance to \( L > 40 \, \mu\text{m} \), leads to the deterioration of the visibility of the interference fringes (Fig. 2d, Region R3). This behavior is a peculiar example of the decoherence phenomena, where the interaction of the individual components of the radiation field with the environment suppress the coherence of the EDPHS and the electron-induced polarization in the sample (see Supplementary Note S4 and Supplementary Fig. S6 for more information).

The high degree of coherence between the EDPHS and the sample radiation, within the aforementioned distance range, is also spectrally analyzed and used for spectral interferometry, as we demonstrate in the following. The angle(momentum)-resolved CL spectra of the combined EDPHS and sample radiation shows as well a clear interference map, at higher momentum ranges between \( k_{||} = 0.7 \, k_0 \) and \( k_{||} = 0.8 \, k_0 \) (Fig. 4a). The LP branch is prominently excited, as the EDPHS radiation peaks at 800 nm. Moreover, by changing the delay (distance between the EDPHS and sample), both the modulation frequency as well as the visibility of the interferences fringes are altered (Fig. 4b). The dependency of the CL signal on \( k_y \), using the technique compared here, i.e., filtering the signal along the azimuthal degree of freedom using a mechanical slit, is compared to the signal obtained beforehand, where the angle-resolved patterns were obtained at the filtered wavelength of \( \lambda = 800 \, \text{nm} \) (Fig. 4c). Obviously, good agreement is obtained. These spectral interference fringes are thus used to retrieve the spectral phase in the following.

To proceed, we first rewrite eq. (1) into three components as

\[
\Gamma_{\text{CL}}(\omega) = |I_0(\omega)| \left[ 1 + |\sigma(\omega)|^2 + \sigma(\omega)e^{i\omega \tau} + \sigma^*(\omega)e^{-i\omega \tau} \right]
\]

at a fixed \( k_{||} \) value, where \( \sigma(\omega) = E_{z, \text{el}}(\omega)/E_{z, \text{EDPHS}}(\omega) \) is the ratio of the EDPHS and the sample \( z \)-components of the electric field radiated to the far field, and \( |I_0(\omega)| = (4\pi \hbar k_0)^{-1} |\tilde{E}_{\text{EDPHS}}(\omega)|^2 \). The first term, i.e., \( \Gamma_0(\omega) = |I_0(\omega)| \left[ 1 + |\sigma(\omega)|^2 \right] \), does not have any dependence on the delay \( \tau \), whereas the last two terms, defined as \( \Gamma_+(\omega) = |I_0(\omega)| \sigma(\omega)e^{i\omega \tau} \) and \( \Gamma_-(\omega) = |I_0(\omega)| \sigma^*(\omega)e^{-i\omega \tau} \), clearly depend on the delay. Thus, taking the Fourier transform of the overall CL spectrum, one can transfer all the components into the time domain, with \( \Gamma_0(t) = \mathcal{F}\{\Gamma_0(\omega)\} \) and \( \Gamma_\pm(t) = \mathcal{F}\{\Gamma_\pm(\omega)\} \) centering at \( t = 0 \), and \( t = \pm \tau \), respectively. Now we perform this for the CL signal at the distances of \( L = 20 \, \mu\text{m}, L = 22 \, \mu\text{m}, \)
and $L = 30 \, \mu m$, all at $k_\parallel = 0.77 \, k_0$, where the time-dependent CL signal is obtained. Three dominant peaks are observed as expected, namely, the DC term at $t = 0$, and the AC terms at $t = \pm 136 \, fs$ for $L = 20 \, \mu m$, $t = \pm 150 \, fs$ for $L = 22 \, \mu m$, and $L = \pm 204 \, fs$ for $L = 30 \, \mu m$, respectively. The occurrence of the DC term is clearly due to the delay-independent CL intensities corresponding to the individual EDPHS and sample CL signals ($\Gamma_0(t)$; the first two terms on the RHS of equation (1)). In addition, the AC terms that occur exactly at $\tau = L \left( v_\text{el}^{-1} - c^{-1} \right)$, are due to the last terms on the RHS of eq. (1). Assuming that the CL intensities from the EDPHS and the sample are at the same level of strength, the degree of mutual coherence is obtained as $\text{CL}_{AC}/\text{CL}_{DC}$, which corresponds to exactly 27%, as inferred from the comparison of the simulated and measured results before. The ratio of the CL intensities of the EDPHS and the sample is experimentally confirmed by taking the CL spectra of individual components under the same experimental conditions (see Fig. 1d). The broadening of the DC signal is also acquired by taking the bandwidth of the DC peak, which corresponds to 5.2 fs FWHM. Thus, the temporal broadening of the

Fig. 4. Spectral interference fringes. (a) Momentum-resolved CL spectra at the distance of $L = 22 \, \mu m$ (delay $\tau = 150 \, fs$), for an electron traversing the WSe$_2$ flake at a distance of 2 $\mu m$ from the edge. (b) CL intensity acquired at the wavenumber of $k_\parallel = 0.77 \, k_0$ at depicted distances. (c) CL intensity versus the lateral momentum, where the results obtained by filtering along the azimuthal direction via a mechanical slit and then selecting the spectral content at $\lambda = 800 \, nm$, are compared to the results obtained by spectrally filtering the radiation at $\lambda = 800 \, nm$, and then selecting the azimuthal range $\varphi = 90^\circ \pm 2$. (d) Fourier-transformed CL intensity at depicted $L$ values, with three peaks at $t = 0$ (DC term) and $t = \pm \tau$, at depicted distances between the sample and the EDPHS. $\delta \tau_1 = 14 \, fs$ and $\delta \tau_2 = 55 \, fs$ correspond to $\delta L = 2 \, \mu m$ and $\delta L = 8 \, \mu m$, respectively. Retrieved relative electric-field amplitude and phase with respect to the EDPHS for (e) $k_\parallel = 0.77 \, k_0$, using the CL signal acquired for $L = 22 \, \mu m$, and (f) $k_\parallel = 0.7 \, k_0$, using the CL signal acquired for different $L$ values.
EDPHS and sample radiation are both approximately 5.2 fs. Taking the inverse Fourier transform of only the AC signal and by filtering the time-dependent signal around the AC peak, the spectral amplitude and phase are both retrieved (Fig. 4c). For doing this, we note that

$$|\Gamma_0(\omega)|/|\Gamma_\pm(\omega)| = \left(1 + |\sigma(\omega)|^2\right)/|\sigma(\omega)|$$

(3)

allowing us to compute the relative electric-field amplitude as

$$|\sigma(\omega)| = \frac{|\Gamma_0(\omega)|}{2|\Gamma_\pm(\omega)|} - \sqrt{\left(\frac{|\Gamma_0(\omega)|}{2|\Gamma_\pm(\omega)|}\right)^2 - 1}.\quad (4)$$

Moreover, the relative phase is obtained by simply retrieving the phase of $\Gamma_\pm(\omega)$, as $|I_0(\omega)|$ is a real-valued quantity. Moreover, since the CL spectrum of only the EDPHS radiation is easily obtained at the first stage, the electric field amplitude of only the sample radiation can be retrieved. However, only the differential CL phase between the sample and EDPHS radiation can be acquired with this technique, since no information about the phase of the EDPHS radiation is at hand. The proposed phase-retrieval algorithm should not depend on the delay between the reference and the signal, as far as the AC and DC spectral components are completely distinguishable. This fact can be used as a benchmark for obtaining the accuracy of the proposed technique. Retrieving the intensity and phase at different $L$-values, the fluctuations in the obtained results are negligible for $L = 20 \, \mu$m and $L = 22 \, \mu$m. However, a maximum difference of the phase value in the order of 20% is obtained, when comparing the values for $L = 20 \, \mu$m and $L = 30 \, \mu$m, which provides an estimate for the accuracy of our spectral interferometry technique.

Thus, the proposed algorithm can be used to retrieve the amplitude ($|\sigma(\omega)|$) and the phase ($\alpha(\omega)$) of the momentum-energy maps (Fig. 5). The accuracy of the acquired maps depend on the visibility of the interference fringes, thus are reliable for $0.5k_0 \leq k_\parallel \leq 0.8k_0$. The retrieved amplitude (Fig. 5a) shows a smooth shift of the LP and UP polariton branches toward shorter wavelengths upon increasing the transverse momentum. This behavior is expected from the dispersion of exciton polaritons (strong exciton-photon interactions), for both LP and UP branches. In contrast, the retrieved phase shows fluctuating behavior versus transverse momentum and a less obvious fluctuation versus wavelength. Considering the lowest-order scattered rays, (see Supplementary Note 4), the relative phase can be described as $\alpha_{n=0}(\omega) = k_\parallel l_1 - \beta(\omega)l_1 - \varphi_{T_2}(\omega)$, which is a smooth function of $k_\parallel$. Thus, we notice that the fast fluctuation behavior of the relative phase is due to the inclusion of higher order scattering terms.

The correlative photon-electron spectroscopy based on EDPHS thus allows for phase-stable spectral interferometry by improving the mutual coherence between the arriving photons and electrons at the sample. Moreover, the compactness of the design allows to minimize decoherence in the photon-electron interaction because the photon-generation process happens at a distance of only a few micrometers above the sample. The scheme thus maximizes the mutual coherence between the electrons and photons. Intriguingly, advanced nanofabrication techniques could be used to design EDPHS structures with tailored photon emission properties. Generating vortex light or even temporally shaped optical pulses is possible by the control of multiscattering events and engineering of defect centers in both the lateral and longitudinal directions. The approach thus opens new directions in understanding the momentum-
spectral correlations in polaritonic materials and correlated electron systems such as transition-metal dichalcogenides. Merging this method with advanced holographic techniques allows for unravelling a variety of information about the charge and energy transfer dynamics at ultimately attosecond time resolution. Moreover, this setup has the potential for exploring fundamental aspects of electron-photon interactions and addressing key questions such as entanglement between generated photons from different scattering events, which could be addressed by combining this with an electron-beam analyzer and spectrometer in an SEM.

Fig. 5. Momentum-wavelength map of the relative EDPHS- and electron-beam-excitations amplitude and phase. Retrieved (a) amplitude, and (b) phase.

Methods.

Cathodoluminescence Spectroscopy – Experimental data were collected in a high-resolution SEM using a field emission microscope (Zeiss SIGMA) equipped with a CL detector (Delmic B.V). SEM images here are obtained using the secondary electron detector in our SEM setup. We used an acceleration voltage of 30 kV and a beam current of 1 to 14 nA (based on the experiment) to excite both EDPHS and specimen throughout the measurements. Despite using a high current, we did not observe a significant radiation damage. An off-axis aluminum-coated parabolic mirror with a hole for the electron beam (diameter: 600 µm) was installed directly below the pole piece (and below the sample) to collect the generated CL radiation. A nanopositioner stage was installed inside the chamber, and a nanorobotic arm was used to accurately position the EDPHS above the sample stage. The acceptance angle of the CL radiation was 1.49 sr and the dwell time of the spectroscopic measurements differs from 50 to 400 ms in different experiments, to account for long experimental time and the signal to noise ratio. Collected CL radiation was directed to the CCD camera for further analysis. Angle-resolved maps were obtained by exposing the sample to electron beam irradiation with the spot size of 50 nm for 10s at each step, whereas energy-
momentum CL measurements were acquired by a single exposure (integration time approximatively 150 s) to the electron beam. For mapping the energy-momentum maps, CL radiation was directed through a one-dimensional slit opening, where a diffraction grating dispersed it on a two-dimensional CCD array, with the momentum components defined by the slit mapped in the vertical direction, yielding a $I(k,\lambda)$ map. The obtained bare CCD image was mapped onto the energy-momentum space, considering the parabolic shape of the mirror, the magnification of the optical path, and the camera pixel size$^{7,8}$.

Numerical Simulations – In order to numerically explore the mutual coherence and interference effects, we performed several simulations using the finite-difference time-domain method. Particularly, we used this technique to simulate the spatio-temporal distribution of the EDPHS radiation. The obtained EDPHS radiation were then superimposed with the electron-beam excitations to excite the sample in a sequential way by altering the delay between the electron and the EDPHS radiation. To perform the simulations, the home-build numerical code described elsewhere and the supercomputing cluster of the Kiel university have been used. Full details about the parameters and simulation times are presented in the Supplementary Notes 1 and 4.

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Author Contributions:

M.T. Performed the experiments, designed and realized the nano-positioner stage, and analyzed the data together with N.T. M.H. fabricated the EDPHS structure. K.R. produced the WSe$_2$ crystals in his group. N.T. conceived the data, designed the experimental configuration, and performed the simulations. N.T. wrote the manuscript with contributions from all coauthors.

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