Simultaneous Nanoscale Excitation and Emission Mapping by Cathodoluminescence

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ABSTRACT: Free-electron-based spectroscopies can reveal the nanoscale optical properties of semiconductor materials and nanophotonic devices with a spatial resolution far beyond the diffraction limit of light. However, the retrieved spatial information is constrained to the excitation space defined by the electron beam position, while information on the delocalization associated with the spatial extension of the probed optical modes in the specimen has so far been missing, despite its relevance in ruling the optical properties of nanostructures. In this study, we demonstrate a cathodoluminescence method that can access both excitation and emission spaces at the nanoscale, illustrating the power of such a simultaneous excitation and emission mapping technique by revealing a subwavelength emission position modulation as well as by visualizing electromagnetic energy transport in nanoplasmonic systems. Besides the fundamental interest of these results, our technique grants us access into previously inaccessible nanoscale optical properties.

KEYWORDS: cathodoluminescence, scanning transmission electron microscopy, super-resolution, transition radiation, plasmonics, nanowire, nanohole

Over the last decades, nanoscopic optical characterization using free electron beams (e-beams) has attracted considerable attention in various research fields, such as nanophotonics and materials science, while fundamental physics benefits as well from the ability of e-beams to access optical states beyond the diffraction limit of light. Optical measurement techniques based on free electrons are classified into electron energy-loss spectroscopy (EELS) and cathodoluminescence (CL), which are typically performed by using a scanning transmission electron microscope (STEM) or a scanning electron microscope (SEM) to obtain spectrally and spatially resolved optical information with a resolution down to a few nanometers or less. Optical information in EELS measurements is extracted by recording the energy loss experienced by the incident electrons, while photons generated upon e-beam excitation are collected and analyzed in CL measurements. The nominal high spatial resolution in both approaches arises from the small size of the e-beam at the position of the specimen combined with the precision in the excitation position, although the optical properties of the sampled nanostructures are oftentimes dominated by relatively delocalized emission or scattering processes involving multiple optical modes and their coupling to radiation, rather than by the actual excitation. In particular, the most interesting piece of information in CL lies in the processes taking place in between excitation and emission, producing spatiotemporal energy flow across different modes of the specimen. Indeed, the excitation and emission positions do not match in general in optical systems such as semiconductors due to carrier diffusion, topologically protected edge-states capable of transporting electromagnetic energy before out-coupling to radiation, cavities combined with localized emitters, and many others. Although the CL approach is advantageous with respect to EELS for the characterization of emission, it does not directly reveal the position from which photons are emerging. This leaves us with a longstanding problem in the analysis of semiconductor optical devices, where excited carriers or excitons recombine and emit photons (incoherent emission processes) after some spatial diffusion that reduces the effective spatial resolution in the CL measurement. Also, in nanophotonic antennas or

Received: October 7, 2022
Accepted: November 17, 2022
Published: November 22, 2022
waveguides dominated by coherent processes, the determination of the emission position is important to analyze and engineer electromagnetic energy transport and conversion. To monitor carrier diffusion in semiconductors, a method that combines near-field probing and electron beam excitation has been proposed, which nevertheless suffers limitations in detection angle or polarization selectivity due to the near-field probing nature as well as the disturbance of the environment by the probe itself.

In this study, we demonstrate a simultaneous excitation and emission mapping (SEEM) approach based on CL and using a collimation parabolic mirror as an imaging element to project the three-dimensional (3D) information associated with the emission position on an arbitrarily chosen two-dimensional (2D) plane (Figure 1a). To select the projection plane of the emission space, angle-resolved CL detection is employed. The current method extends in a nontrivial way previous CL capabilities, such as the evaluation of dispersion relations as well as the determination of emission directionality and interference.

RESULTS AND DISCUSSION

Emission Imaging in STEM-CL. The CL-emission imaging system is built in a STEM-CL setup equipped with an angle-resolved spectrum mapping system, where a parabolic mirror is inserted at the sample position of the STEM instrument to collimate the light radiated from the sample (Figure 1). The emitted light, transferred to the optical system through a polarizer and an angle selection mask, is subsequently focused on a camera for emission imaging (Figure 1b). Simultaneous spectral measurement is also available by means of a beam splitter (see Supporting Information (SI) for details on the setup). The collected CL signal is given by the overlap between the excitation region and the optical collection spot, which is back-projected from the detection optics (schematically illustrated in Figure S1 in SI). Since each pixel on the camera can be optically traced onto a back-projected spot on the sample plane, the emission image on the camera corresponds to the optical spot scan on the specimen. The “excitation region” extends beyond the e-beam position due to the finite extension of the modes sampled in the specimen (i.e., wave propagation or diffusion). We use two different modes of operation, namely: (1) “emission imaging” corresponding to scanning the optical spot over the specimen for a fixed selected e-beam spot position (see Figure 1c); and (2) “emission-spot-decomposed CL mapping”, in which the e-beam is scanned while the optical spot is fixed at a selected position (see Figure 1d).
Using the parabolic mirror as the image forming element, the optical axis for the image formation can be selected by the angle-resolving mask placed on the angular space plane. Without the angle-resolving mask, the optical axis corresponds to the $x$ axis with a detection solid angle of around $3\pi$ steradians, and the imaging plane of the camera corresponds to the $y$-$z$ plane of the sample space (see SI for these imaging conditions). Under such conditions, a sample shift along the $x$ direction produces a defocus of the emission image. To obtain an emission image corresponding to a $x$-$y$-plane projection (i.e., the STEM imaging plane), the optical axis should be along the $z$ axis, implying that only radiation emitted along the upward direction should be collected. Figure 2 shows an emission image demonstration (mode (1), see Figure 1c) with upward detection ($\theta = 0^\circ$), revealing a shift in the emission spot when physically displacing the sample ($a \sim 2 \mu m$ ZnS:Pb particle), as schematically illustrated in panels a, d, and g. In this measurement, a single emission image is acquired by integrating the signal while scanning the e-beam over the entire area of the STEM image. Thus, the imaged emission spot corresponds in fact to the image of the entire particle. As the specimen is moved along the $x$ and $y$ axis by $\pm 2 \mu m$, the emission spot shifts accordingly in the emission images (Figure 2c,d), faithfully mimicking the sample shift in the STEM images (Figure 2b,e). With a $z$ specimen shift, which corresponds to defocus in the STEM image (Figure 2h), the emission image in Figure 2i is also defocused. These sets of results show that the projection plane of the three-dimensional emission position space can be arbitrarily chosen by selecting the emission angle as, for example, the $x$-$y$ or $y$-$z$ plane (see also the SI).

**Emission-Spot-Decomposed CL Mapping.** Since an emission image (as a function of the position of the optical spot) is obtained for each e-beam position, a four-dimensional (4D) data set is obtained by synchronizing the emission image acquisition with a raster scan of the e-beam. The data representation of the two operation modes (Figure 1c,d) is possible by postprocessing the 4D data set (see SI for details). In this section, we present an example of mode (2) (see Figure 1d). The results obtained by applying mode (1) (see Figure 1c) to the same data set are shown in SI. The angle-resolving mask is inserted such that the imaging plane corresponds to the $x$-$y$ sample plane by selecting the upward emission from ZnS:Pb particles, as illustrated in Figure 3a. Figure 3c shows an integrated emission image over the entire e-beam scan area (Figure 3b), showing three spatially separated emission spots in the integrated emission image corresponding to the three particles in the STEM bright-field image (Figure 3c). Figure 3d shows emission-spot-decomposed CL maps constructed by integrating over e-beam excitation positions around selected particles. The signal integration area of the excitation position is indicated by the dashed rectangles (I)-(III) in Figure 3c. The emission-spot-decomposed CL map integrating all the three emission spots in Figure 3d-(I) shows clear particle shapes that are in good correspondence with the STEM bright-field image. Figure 3d-(II) shows the emission-spot-decomposed CL map integrated over the upper right emission spot, as indicated by the rectangular region (II) in Figure 3c. Only the upper right particle is imaged in the CL map corresponding to the selected emission spot. Similarly, as shown in Figure 3d-(III), the emission-spot-decomposed CL map corresponding to the region (III) containing the lower spot in Figure 3c shows the CL particle image of only the lower particle. Thus, the emission-spot-decomposed CL mapping can directly visualize the correlation between the excitation and emission positions. For this particular instance of separated particles, the correlation is complete, in contrast to connected regions in nanowires and perforated films, in which the correlation between excitation and emission reveals information about intermediate processes, as we discuss below. As a reverse analysis, the emission image of a certain excitation can also be extracted from the 4D data set of the SEEM measurement (see details in SI).

**Beating the Diffraction Limit in the Evaluation of the Emission Spot Shift.** Although the spatial resolution to separate two emission spots is limited by Rayleigh’s criterion, the position of a single spot can be determined well below the diffraction limit of light from the location of the intensity maximum, in analogy to optical super-resolution techniques such as PALM. Since the actual profile of the point spread function (PSF) of the emission spot slightly changes with position due to the parabolic mirror optics, we adopt a center-of-mass (COM) evaluation to determine the emission spot position instead of the PSF fitting used in the PALM method.
To examine this evaluation method, we measure transition radiation from a planar metal layer.\textsuperscript{22} Since transition radiation can be attributed to a perpendicular dipole excited exactly at the position of the e-beam, the emission position can be precisely controlled with the resolution of the electron microscope. To efficiently collect the transition radiation, we insert the angle-resolving pinhole mask at a detection angle of $\theta = 45^\circ$ and set the solid angle range to 1.8 sr, as schematically shown by the red hatched area in the angular space in Figure 4a. Since emitter shifts along the $x$ and $z$ directions are both projected on the vertical axis in the emission position image under this observation conditions (see Figure S4 in SI), we here evaluate only the emission spot shift in the horizontal direction corresponding to the $y$ direction in the sample space. Figure 4b shows the emission image of a representative excitation position, where the intensity spreads approximately over 500 nm due to light diffraction. The calculated COM position is marked as a blue dot. For three representative horizontal scans, the COM position of each excitation point is plotted on the emission image integrated over the three scans, as shown in Figure 4c. To summarize the evaluation, we plot the emission spot shift in Figure 4d for an e-beam scan area of 1.6 $\mu$m $\times$ 1.6 $\mu$m as a function of the excitation position shift (in steps of 80 nm in $x$ position). The distance between two emission spots is evaluated for all possible combinations in each horizontal scanline. The horizontal axis in Figure 4d corresponds to the shift in the excitation position by the e-beam, and the vertical axis stands for the average $y$-shift of the emission spot as resolved from the COM. The error (standard deviation of the measurement) increases as the excitation position shift becomes larger, which indicates that the shape of the spot deforms for large position shifts (see Figure S6 in SI). For a shift in excitation position of up to 1 $\mu$m, the error in the evaluation of the emission position is $\leq$50 nm, well below the diffraction limit of light. For smaller emission position shifts, the accuracy can reach down to $\sim$10 nm, as shown in Figure 4e. We thus conclude that super-resolution beyond the light diffraction limit can be achieved in SEEM on the determination of the CL emission position. While we evaluated here the small limit of the emission spot shift, knowing the maximum measurable range can also be relevant. With a parabolic mirror having a focal length of 1.5 mm, we could reasonably evaluate the spot shift up to around 5 $\mu$m. However, further characterization is still needed to estimate practical operation ranges, possibly including image processing or dynamic optical alignment.

**Shift of the Emission in a Plasmonic Nanowire.** We now evaluate slight shifts of the emission spot in the presence of optical modes in a plasmonic nanowire as a result of the delocalized nature of such modes. When a fast electron passes through or near a metal nanowire, an electric dipole is excited at the surface near the e-beam spot, giving rise to a localized emission source, in a similar manner as transition radiation on a flat metal surface. Simultaneously with this localized dipole, surface plasmon modes of the wire are also excited (in response to such dipole), resulting in nodes/antinodes along the wire axis, which can perturb the resulting optical emission, which is no longer concentrated at the e-beam position. We explore this effect by examining a 1.5 $\mu$m long silver nanowire of 95 nm in diameter with its major axis oriented parallel to the $y$ direction (see Figure 5a,b). As illustrated in Figure 5a, by using the angle-resolving mask and a polarizer, only $p$ polarized emission around $\theta = 0^\circ$ with a detection solid angle of 1.14 sr is acquired to selectively observe the local polarization along the short axis of the wire parallel to the support film (a 20 nm thick free-standing elastic carbon membrane). We choose the wavelength range as 550–700 nm, in which the wire plasmon mode is most clearly observed. The mask area is schematically illustrated as a red circle in Figure 5a. In a conventional CL map, as shown in Figure 5c, a high-intensity contrast with multiple antinodes is visible along the upper and lower edges of the wire, indicating the presence of a wire mode.\textsuperscript{23–25} The high-intensity hotspot location appears alternatively at the upper and lower edges.
along the wire axis. This zigzag contrast is due to the local polarization across the short axis interfering with the charge distribution of the wire mode as well as to the detection asymmetry originating from the circular aperture inserted around θ = 0°, which covers a larger solid angle in the x-positive direction. Indeed, a boundary-element method (BEM) simulation (Figure 5g) for the same wire geometry corroborates the emergence of a clear standing wave pattern along the wire when no angle or polarization selection is applied. The zigzag hotspot distribution is also reproduced with the polarization- and angle-selection under the experimental conditions, as shown in Figure 5h (see details of the BEM calculations in Methods).

To examine how this interplay between the local dipole and the plasmon mode influences the emission position, we evaluate the shift of the emission spot position with respect to the excitation e-beam position. Three excitation spots indicated in Figure 5b are chosen as representative positions, and the corresponding three emission images are collected, as shown in Figure 5d. The emission spot seems to follow the change in the excitation position, indicating that localized emission from the electron beam position is dominant. The emission-spot-decomposed CL map using the corresponding emission position locations also supports the primary contribution of localized emission at the e-beam position (Figure 5e, operation mode (2), see Figure 1d). To more quantitatively corroborate the correlation between the shifts in the emission spot and e-beam excitation position, we perform a COM analysis of the emission spot similar to the previous discussion in Figure 4. In Figure 5f, the COM position is plotted as a function of the excitation position along the y axis (long axis of the wire). The COM position plot reveals modulated features, resulting in deviations from a linear relation. This modulation period roughly corresponds to the antinode period, indicating some perturbation related to the standing waves associated with plasmon modes propagating along the wire. The simulated COM analysis shown in Figure 5i further confirms this modulation of the spot position. The COM positions and the corresponding spot shift positions are indicated by the dashed lines in Figure 5c for the experiment and in Figure 5h for the simulation. The spot position tends to be shifted toward the antinode positions of the wire mode, well supporting the experimental results. We note that the observed deviation from perfect rod image symmetry along the horizontal direction in the experiment in Figure 5c is due to imperfections of the actual rod shape. Thus, using the SEEM method, we reveal that the CL emission position from the plasmonic nanowire excited by an e-beam is perturbated by the presence of plasmon modes and that the emission position tends to be attracted toward the antinode positions due to field propagation associated with those plasmons.

**Emission from Metal Holes Coupled to Surface-Plasmon Polaritons**. The second model system with (in principle) nonleaky wave propagation is a metal thin film perforated with holes, which supports both propagating surface-plasmon polaritons (SPPs) and localized surface-plasmon modes at the holes.26,27 The local modes can be excited by the e-beam directly hitting the hole edge as well as through scattering of SPPs induced by the e-beam on a flat silver area distant from the hole location.16,28 Since the e-beam incident on a flat silver surface generates transition radiation together with SPPs, two spatially separated emitters corresponding to the e-beam position and the hole are present. We use a slit-type mask with p polarization to detect both the transition radiation and the local mode, as illustrated in Figure 6a. The diameter of the observed silver hole is 260 nm, as shown in the STEM dark-field image in Figure 6b. Figure 6c shows the emission images integrated over the e-beam excitation regions (1)−(3) in Figure 6b. The emission image of region (1), selecting only the hole area, shows a single emission spot. Note that the elongation of the emission spot (horizontal direction in panel c-(1)) is due to the shape of the slit mask (see the details in SI). Since the emission-spot-decomposed CL map of the region (1) in panel d shows the field distribution of the dipole with the strong field along the x-axis edge of the hole, we understand that this center emission spot is dominantly formed by the localized mode of the hole. When the e-beam hits the flat silver area away from the hole, causing both transition radiation and emission of the local mode through SPPs, these two emission spots exhibit...
Figure 5. Emission imaging of a silver nanowire. (a) Schematic illustration of the measurement. The pinhole mask is inserted to select upward radiation with $p$ polarization at a detection angle $(\theta, \varphi) = (0^\circ, 0^\circ)$ with a solid angle of 1.14 sr, which is indicated by the red area in the emission angle space. The wavelength range is chosen as 550–700 nm. (b) STEM bright-field image of the silver nanowire (1.5 μm length and 95 nm diameter) and (c) corresponding CL map. The direction of $p$ polarization is schematically shown by the yellow arrow in panels a and c. (d) Superimposed emission images (operation mode (1), see Figure 1c) with the emission signals integrated over e-beam positions in the left edge (red), center (green), and right edge (blue) of the wire (see color-coordinated rectangles indicating the e-beam position regions in panel b). (e) Emission-spot-decomposed CL photon maps (operation mode (2), see Figure 1d) with the emission signals integrated over the emission image in the areas (I)–(III) in panel d. (f) Relationship between the measured shifts in excitation position and emission COM from the left edge in the $y$ (horizontal) direction. The emission spot at each excitation position along $y$ is evaluated along the upper edge of the wire. (g) Simulated unpolarized CL map with the signal integrated over all angles. (h) Angle- and polarization-selected CL map under the same conditions as in the experiment. (i) Simulated COM shift as a function of e-beam position. The e-beam is scanned along the upper edge of the wire, as in the experiment of panel f. The wavelength for the simulation is 631.8 nm.

Figure 6. Emission imaging of a single silver hole. (a) Schematic illustration of a silver hole measurement with $p$ polarization. The collected radiation is limited by the slit mask to the upward direction along the $x$-$z$ plane, as indicated by a red stripe in the emission angle space. (b) STEM dark-field image of the probed silver nanohole with a diameter of 260 nm. (c) Emission images integrated over e-beam positions in the areas indicated by the rectangles (1)–(3) in panel (b). (d) Emission-spot-decomposed CL photon maps integrated over emission position space in the rectangles (I)–(III) in panel c. The direction of polarization is schematically shown by the yellow arrow in panel d-(I).
interesting interferences. Figures 6c-(2) and 6c-(3) show the emission images corresponding to the beam excitation on the flat film areas 0.5 μm away from the hole, as indicated by the (2) and (3) squares in panel b. As shown in Figure 6c-(2), the emission image corresponding to excitation from the top side yields a single emission spot shifted to the upper side compared to the direct excitation of the hole in Figure 6c-(1). In the corresponding emission-spot-decomposed CL photon map in Figure 6d-(II), where only the emission from the upper part is selected, as indicated in Figure 6c-(2), the contribution of transition radiation is clearly observed in the form of strong intensity over the flat silver film region. In contrast, the emission image from the lower part of the silver film (region (3) in panel b) gives two separate emission spots, as shown in Figure 6c-(3). The upper emission spot shares the same position as the direct nanohole radiation in panel c-(1), while the lower emission spot corresponds to the position of the transition radiation from the silver thin film. Indeed, in the emission-spot-decomposed CL photon map, using this lower spot (Figure 6d-(III)), the bottom part of the flat silver film is brightened, which is opposite to the map of Figure 6d-(II), constructed from the upper part of the emission spot. The difference in the spot shape depending on the upper (2) or lower (3) side excitations is related to the phase difference of the two emission sources, i.e. hole and TR, due to the asymmetry in the detection direction with respect to the y-z plane. The relative phase of emission from the hole with respect to transition radiation from the e-beam position is different depending on whether SPPs reach the hole from the upper or lower sides of the film (i.e., from regions at different distances with respect to the mirror). Thus, the two emission spots appear to interfere constructively or destructively for lower or upper side excitation, respectively. Such interfering spot formation can be reproduced in the emission spot simulation assuming two separated emission sources corresponding to the hole and the transition radiation with different phases in SI.

When two holes are adjacent within the attenuation distance of SPPs, the local modes of the two holes interact through the propagating SPPs, which emulates a system consisting of a receiver and a transmitter connected by a waveguide transporting the electromagnetic energy. We here demonstrate SEEM measurements of such a two hole-system, where a pair of silver holes are aligned along the y direction (horizontal direction in the image), as shown in Figure 7a. The holes are 500 nm in diameter and are separated by 2 μm, which should be long enough to avoid near-field interaction of local modes. We choose to measure s-polarized emission to eliminate the contribution of transition radiation, and the same angular region as in the single hole measurements (Figure 6) is selected. We take larger size holes to increase the hole-scattering signals relative to the single hole considered in Figure 6. Quadrupole components can be relevant for large holes, particularly at short wavelengths (see details of the influence of the quadrupole mode in SI). Under panchromatic imaging conditions, without wavelength selection in order to obtain a sufficient signal-to-noise ratio, we observe dominant dipole components similar to the smaller hole in Figure 6: The wavelength-integrated emission image from the left hole (Figure 7b-(1)) shows one emission spot corresponding to the local dipole mode excited at the left hole. The emission image from the right hole (Figure 7b-(3)) also has one spot, which is shifted along the y direction from the left hole spot (Figure 7b-(1)). When the e-beam excites the area between the holes (region (ii) in Figure 7a), two emission spots are observed due to the excitation of both holes through SPPs, as shown in Figure 7b-(2). The emission-spot-decomposed CL map including both emission spots, corresponding to integration over e-beam positions within the area (I) in Figure 7b-(2), shows patterns that are consistent with electric field distributions of horizontally polarized dipoles of both holes, as shown in Figure 7c-(I). In order to visualize energy transport from the left hole to the right one via SPPs, we extract the emission-spot-decomposed CL map (operation mode (2), see Figure 1d) of the emission position region (II) in Figure 7b-(3). As shown in the map in Figure 7c-(II), the local mode of the right hole produces a dominant feature, while the dipole-like intensity distribution at the left hole is also visible. This means that emission from the right hole occurs upon left-hole excitation, directly showing evidence of signal transfer across holes mediated by SPPs. Thus, the SEEM method allows for a visual evaluation of energy transport between distant nanostructures.

CONCLUSIONS

We have developed a nano-optical imaging method to access the emission position space, which allows simultaneous 4D information acquisition including the 2D excitation mapping by the e-beam and the 2D optical emission image corresponding to each excitation position. The projection plane of the 3D emission position space can be arbitrarily selected by the detection angle. By applying a center-of-mass analysis to the emission image, the resolution in the position of emission is pushed well below the diffraction limit of light. We have applied this SEEM method to determine the spatial shift of the emission spot in a silver nanowire and revealed that the emission spot is modulated periodically as the e-beam is raster scanned and hits the nodes and antinodes of the plasmon modes in the nanowire. Using holes on a silver membrane, the spatial transport of the electromagnetic energy propagated by
SPPs has been directly visualized by identifying both the excitation and the emission positions.

The introduced SEEM method constitutes a powerful approach for spatially analyzing the whole process of photon generation, from excitation to actual emission. While we have presented application examples of coherent emission processes, the SEEM approach can also be applied to the evaluation of collective relaxation processes associated with incoherent emission, such as in semiconductors with carrier or exciton diffusion with a characteristic range from a few hundred nanometers to microns. The effective spatial resolution in CL, which is limited by carrier diffusion, can be improved in the SEEM approach, for example, by selectively extracting the radiation emitted at the desired location to consider the effect of carrier diffusion. In consequence, the developed SEEM method enables a nanoscopic comprehensive analysis of excitation and emission that holds potential to unveil previously inaccessible information in optical materials and devices, stepping into the next stage of nano-optical imaging.

METHODS

Cathodoluminescence Measurements. A modified STEM (JEM-2100F, JEOL, Japan) with a Cs-corrector is used at an acceleration voltage of 80 kV. The electron probe current is about 1 nA with a 20 mrad illumination half angle, which results in a 1 nm probe size. An aluminum parabolic mirror is mounted between the pole pieces of the objective lens, so that light emission from the sample at the focal point of the mirror is collected with a collection solid angle of more than 3 sr. The emission from the sample is collimated by the parabolic mirror, and guided out of the STEM column through the vacuum window. After traveling through a polarizer (s and p polarization), the emitted light is split into two paths by a beam splitter: to the camera for SEEM analysis (optical path (I) in Figure 1b); and to the spectrometer for angle-resolved spectroscopic CL (the optical path (II)). In the latter, the emission is focused on the spectrometer plane. The detection angle can be selected by a mask inserted on the emission angle planes of the both light paths (II,III). More details of the basic CL system can be found in previous publications. In the emission imaging system, the real-space emission position reflected by the beam splitter is focused on the camera plane. For a wavelength range selection in emission, a bandpass filter is inserted in the optical path.

Sample Fabrication. Silver nanowires (Silver nanowire A50SL (Ethanol dispersion)/ Filgen) are dispensed on a TEM grid with an elastic carbon supporting membrane. A nanowire with a diameter of 95 nm is selected and observed in the experiment. Silver holes are prepared using colloidal lithography and film transfer, where a free-standing carbon thin membrane with holes of diameters of 260 and 500 nm is fabricated and transferred to a TEM grid. Silver and SiO2 films are deposited via sputtering on holey carbon films with thicknesses of 50 and 15 nm, respectively. Since one side of the hole film is covered with a carbon film, SPP propagation is limited to the silver/SiO2 interface.

Boundary-Element Method (BEM) Simulation. Simulations are carried out using an adapted version of the BEM that exploits the axial symmetry of the nanowire, such that the sample boundary is one-dimensional and the number of parametrization points is radically reduced. The electric far-field amplitude associated with the CL emission produced by the e-beam excitation is separately calculated for each azimuthal angular number m, with a global dependence on azimuthal angle φ and w encapsulated in an overall factor eimφ. We achieve convergence using 700 parametrization points and |m| ≤ 4. The COM analysis of the simulated field is performed by calculating the optical spot imaged through the parabolic mirror, as described in the Supporting Information.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsnano.2c09973.

Details of the measurement procedure, projection of the emission position space without angle selection, excitation-position-decomposed emission image, calculation of the emission image on the camera, simulated projection-plane selection of the emission position, shape of the emission spot produced by sample position shifts, CL spectrum of the single hole with a diameter of 260 nm, emission spot shape with a slit mask, interfering emission spots from transition radiation and hole, interference of dipole and quadrupole (PDF)

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The manuscript was written through contributions from all authors. T.M. and T.S. conceived the research. T.M., S.O., and T.S. carried out the experiment. T.M., F.J.G.A., and T.S. performed the analysis and calculation. All authors have given approval to the final version of the manuscript.

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We thank Dr. Naoki Yamamoto for his valuable advice. The authors acknowledge the financial support from JSPS PD2(20J14821), JSPS Kakenhi (21H01782, 22H05033), JST FOREST (JPMJFR213J), the European Research Council (Advanced Grant No. 789104-eNANO), the Spanish MI-CINN (PID2020-112625 GB-I00 and Severo Ochoa CEX2019-000910-S), the Catalan CERCA Program, and Fundaciós Cellex and Mir-Puig.

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