Supporting Information to

Far-field radiation of three-dimensional plasmonic gold tapers near apices

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Figure S1. Simulated CL spectra (a–b) and experimental EEL spectra (c) of the taper with 13° opening angle as a function of distance from the apex along its shaft. (a) is calculated using the Drude plus 2 critical point functions of the dielectric function of gold and (b) only with the Drude model. The red dashed rectangle specifies the spectral range of the experimental CL measurements.

The simulated CL spectra in Figure S1a and S1b are calculated through summation of the Poynting vectors along the six orthogonal directions for each electron impact. In this way, both the radiated power and the guided power along taper are included. The non-radiative losses are absent in the CL spectra when compared with the corresponding experimental EEL spectra (Figure S1c). First, the simulated CL spectra in Figure S1a were computed by fitting the experimental permittivity of gold to the Drude model plus two critical point functions, as pointed out in ref. [1, 2]. In this case, the mentioned model was used to incorporate both the response of the conduction electrons and
the influence of the $d$-band transitions. The behavior of the conduction electrons, like surface plasmons, was represented by the Drude model. In Figure S1a, the multiple peaks display an intensity discontinuation around 1.5 eV to 1.75 eV and a slight spectral shift above 1.75 eV compared to the smooth tendency in the EELS spectra.

Since the $5d–6sp$ interband and the $6sp$ intraband transitions of gold happen within the same spectral range (red dashed rectangle) as well, band transitions may offer a channel for non-radiative decay in addition to the decay of surface plasmon, and thus cause the difference between the CL and EELS spectra.

To reveal the influence of band transitions, we selectively switched off the band transitions by numerically setting the oscillator strengths for our permittivity model to zero, so that only the Drude term for the matter’s response was left. The corresponding CL spectra are shown in Figure S1b. They display almost the same features as the previous CL spectra as shown in Figure S1a, including the well-defined resonances below 1.5 eV and the broadband signals above 1.5 eV. The major difference is that the CL intensity in the spectral range above 1.5 eV increases when considering only the Drude term, which can be explained as less absorption due to the lack of inter-/intra-band transitions. This result indicates that the discrepancy between the CL and EELS is not because of radiation caused by inter-/intra-band transitions. It supports also that the plasmon-induced radiations plays the dominant role in the acquired CL spectra.

Concerning the physical reasons, CL is sensitive to the far-field phase difference, which is not accessed by EELS. Therefore, far-field interference effects can be reflected in the CL spectra and may result in the above-mentioned observation. To further investigate the underlying reason, we simulated the CL spectra of a $13^\circ$ taper without an apex, so that the reflection of the $m = 0$ mode at the apex and the radiation of it at the apex were omitted. This was achieved by inserting absorbing boundaries across the taper, so the electromagnetic waves were absorbed by the boundaries and not reflected by the apex. The corresponding results are shown in Figure S2 and the resonance discontinuation is still visible around 1.5 eV. Therefore, the far-field interference between the radiations at different locations along the taper should be highly responsible for the cleft in the CL spectra.

To understand the influence of the local radius on the radiative behavior of plasmonic modes, the energy–momentum dispersion of the $m = 0$ and $m = 1$ plasmonic modes along infinite gold fibers were computed. In this analytic treatment, a fast electron travelling near the fiber surface excites the plasmonic modes with different azimuthal orders, which propagate along the tapers ($z$ direction) in the picture of energy and momentum conservation (inset at the top of Figure S3). The linear momentum of the excited $m = 0$ and $m = 1$ modes along the propagation direction $k_z$ was calculated versus the energy (equivalent to the corresponding energy loss of the exciting electron). The dispersion relation of the modes at different fiber radii are presented in Figure S3a and S3b. Dispersions on the left of the vacuum light line represent the radiative behavior, which corresponds to a CL signal. As seen in Figure S3a, the $m = 0$ mode is excited at all three radii of 40, 60, and 90.
nm. However within the CL detection range (highlighted by the red dashed boxes), it couples hardly to radiation. In contrast, the $m = 1$ mode strongly couples to radiation within the detection range as shown in Figure S3b. It is almost not excited at the small radius of 40 nm. However, it becomes excited and predominantly radiative at the radius of 60 nm. For a larger radius of 90 nm, the $m = 1$ mode tends to propagate evanescently between 1.5 and 2.5 eV and the radiation is redshifted to lower energies. This indicates that there exists a critical radius at which the $m = 1$ mode couples to radiation. Therefore, the radiation of the $m = 1$ mode should be responsible for the CL signals observed at the local radius around 60 nm. These analytical treatments have been discussed in detail in ref. [4].

**Figure S2.** Simulated CL spectra (b) of the 13° taper without an apex (a) in order to omit both the reflection of plasmons and their radiations at the apex. The impact locations of electrons denoted by red dots along the shaft in (a). Perfect absorption layers are positioned at both truncations of the taper.
Figure S3. Momentum-resolved energy loss spectra of (a) $m = 0$ mode and (b) $m = 1$ mode on infinite gold fibers with the radius of 40, 60 and 90 nm, respectively. The inset above shows the calculation scheme in which a fast electron excites surface plasmonic modes of an infinite gold fiber propagating along the $z$ direction. The linear momentum $k_z$ of individual modes is plotted versus the energy loss of the exciting electron. The rectangles with red dashed lines specify the detection range of the CL measurements. The dispersion of light in vacuum are depicted as white dashed lines in each plot. The bulk plasmon of gold in the simulations is at around 2.5 eV.

In Figure S4, calculated radiation along different directions (+z, -x, +x, and +y directions) of the 13° taper with the exciting electron beam along the shaft is demonstrated. The radiation along the +z direction emerges above a critical radius. However, the intensity along the +z direction (∼ 3 ×
is roughly only 20% compared to the intensity along other directions (~$15 \times 10^{-4}$). Therefore, in our investigated regime, the radiation in the $xy$ plane is dominant. Nevertheless, we notice that the radiation along the $+z$ direction can be enhanced when launching helical mode along the taper via a novel excitation scheme.\textsuperscript{5,6}

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Figure S4. Calculated radiation across planes normal to the (a) +z, (b) +x, (c) -x, and (d) +y directions when the electron scans along the shaft of a 13° taper. The coordinate with the respect to the taper is depicted in the inset above.