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Direct Observation of Plasmonic Modes in Au Nanowires Using High-Resolution Cathodoluminescence Spectroscopy

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ABSTRACT

We use cathodoluminescence imaging spectroscopy to excite and investigate plasmonic eigenmodes of Au nanowires with lengths of 500–1200 nm and ∼100 nm width. We observe emission patterns along the Au nanowire axis that are symmetric and strongly wavelength dependent. Different patterns correspond to different resonant modes of the nanowire. From the observed patterns, we derive the spatial and spectral properties of the wire eigenmodes and determine the dispersion relation for plasmonic Au nanowire modes.

The ever-increasing speed and decreasing size of electronic circuits will meet fundamental limits in the near future. Integration of electronics with optics is promising because optical components can provide a very high bandwidth. However, a simple downscaling of conventional optics to nanometer scale is not possible due to the diffraction limit, hindering true nanoscale applications. Surface plasmon polaritons (SPPs) are electromagnetic waves that are strongly coupled to electron plasma oscillations at a metal−dielectric interface. They are characterized by a dispersion below the light line, enabling reduced wavelengths at optical frequencies.[1,2] Moreover, they are strongly bound to the metal surface, leading to high lateral confinement. SPPs propagating along metal nanowires therefore allow photonic manipulation below the diffraction limit.

At small length scales, metal nanowires with finite length behave as plasmonic cavities with resonant eigenmodes.[3] Properties of metal nanowire plasmon resonances have been investigated by extinction measurements on large ensembles.[3,4] To study the nature of the resonances in greater detail, individual wires have been investigated using scanning near-field optical microscopy (SNOM).[5,6] With SNOM, however, one is still limited in resolution by the size of the tip (>50 nm). Furthermore, the tip proximity can have a very strong influence on the optical behavior of the sample.[7,8]

Here we study photonic properties of metal nanowires at high resolution using electron radiation. The electron beam generates a broad spectrum of SPPs, which can subsequently generate radiation that can be detected.[9−11] In this Letter, we use CL imaging spectroscopy for direct observation of plasmonic nanowire modes and determine both spectral and spatial properties of plasmonic eigenmodes. On the basis of the data, we determine the dispersion relation of SPPs on the Au nanowires.

Au nanowires were fabricated on a doped silicon substrate using electron beam lithography and lift-off. Figure 1a shows a scanning electron micrograph of a nanowire specimen. The nanowire width is ∼100 nm and the length is 725 nm. Additional nanowires were fabricated with lengths in the 500−1200 nm range. The nanowires were separated by 5 μm so that no interaction between wires is expected. The silicon provides a highly conductive substrate avoiding charging during electron beam irradiation, but does give a weak CL background signal that is subtracted from the data.

CL spectroscopy was performed in a scanning electron microscope (SEM) extended with a Gatan ParaCL cathodoluminescence system. The electron beam (30 kV, waist <5 nm) is sent through a hole in a parabolic mirror that is mounted above the substrate. The mirror collects light that is emitted from the sample within a large opening angle (0.5π sr) and collimates it into a spectrometer that is equipped with a CCD detector with a 1340 × 100 pixel array, recording spectra in the 390−950 nm wavelength range.

Figure 1b shows the result of spectral collection during an electron beam scan of a 725 nm long Au nanowire. The electron beam was scanned in seven rows and 30 columns, and for every pixel, a spectrum was collected. The figure shows emission in three wavelength windows from this scan, around 592, 640, and 730 nm. For 592 nm, we observe a pattern of four maxima, while for 640 and 730 nm, three maxima are observed. These images demonstrate that the electron beam can excite multiple resonances on the wire that each have a specific spatial profile.

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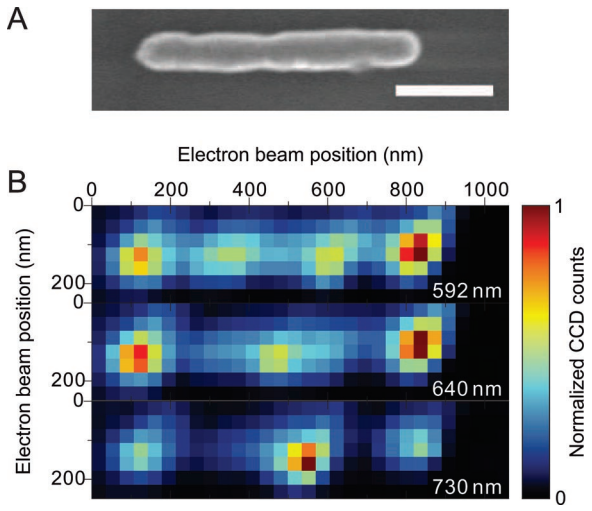

Figure 1. (a) Scanning electron micrograph of a 725 nm long Au nanowire on a Si substrate, fabricated using e-beam lithography. Scalebar is 250 nm. (b) Cathodoluminescence images of the Au nanowire, at wavelengths of 592, 640, and 730 nm. The light emission pattern is strongly wavelength dependent, indicating excitation of different plasmonic modes.

For a more detailed investigation of the data in Figure 1, the electron beam was scanned along the wire axis, and for every beam position spaced by 14 nm, a spectrum was collected (see Figure 1a for a SEM image). The result of this scan is shown in Figure 2b. The spectral axis is plotted horizontally and beam position is plotted vertically. The color scale indicates the recorded intensity. Before further analysis, we removed from the dataset the peak around wavelength 730 nm and position 0.53 μm using a two-dimensional Gaussian fit.[12]

Figure 3a shows the CL spectra for two positions on the wire, which are indicated by X and Y in Figure 2b. Gaussian fits are performed on these data and show for spectrum X predominant contributions of the resonances peaking at 540 and 592 nm, while spectrum Y is made up mostly by the 540 and 640 nm resonance. The fitted spectral widths are in the range 40−70 nm, corresponding to a characteristic cavity Q of ∼10.

Figure 3b plots relative contributions of each resonance at each position on the nanowire. Three clear maxima are observed along the wire for the 640 and 690 nm resonances; four maxima are observed for the 592 nm resonance and, less pronounced, five resonances are observed for the shortest-wavelength resonance at 540 nm. To verify the quality of the analysis, the line scan data in Figure 2b were reconstructed by convoluting the four resonance spectra with the spatial profiles. This reconstruction is drawn in Figure 2c and shows that the four resonances provide a good representation of the data.

For a further analysis, we treated the nanowire resonances as organ-pipe-like modes, i.e., interfering SPPs travelling in opposite directions that reflect at the wire ends. Furthermore, we assume that the CL emission spectrum from the sample is directly related to the intensity of different eigenmodes at the spot of the electron beam.[9,13,14]

In this picture, the distance between two intensity maxima in the profiles of Figure 3b corresponds to $\lambda_p/2$, where λ_p is the SPP wavelength. For a wave in a dispersive medium, with complex index of refraction, there is a nontrivial phase shift upon reflection from the wire end that can be wavelength-dependent. For this reason, we only determine λ_p from peaks within the wire to avoid the effect of this phase shift.

From the data in Figure 3b, and additional measurements on Au nanowires with different lengths in the 700−1200 nm range, several values for λ_p were determined for the corresponding resonance wavelength.

Combining the resonance energy with the corresponding SPP wavelength (λ_p) yields a data point of the dispersion relation of SPPs on a Au nanowire. Dispersion data for all

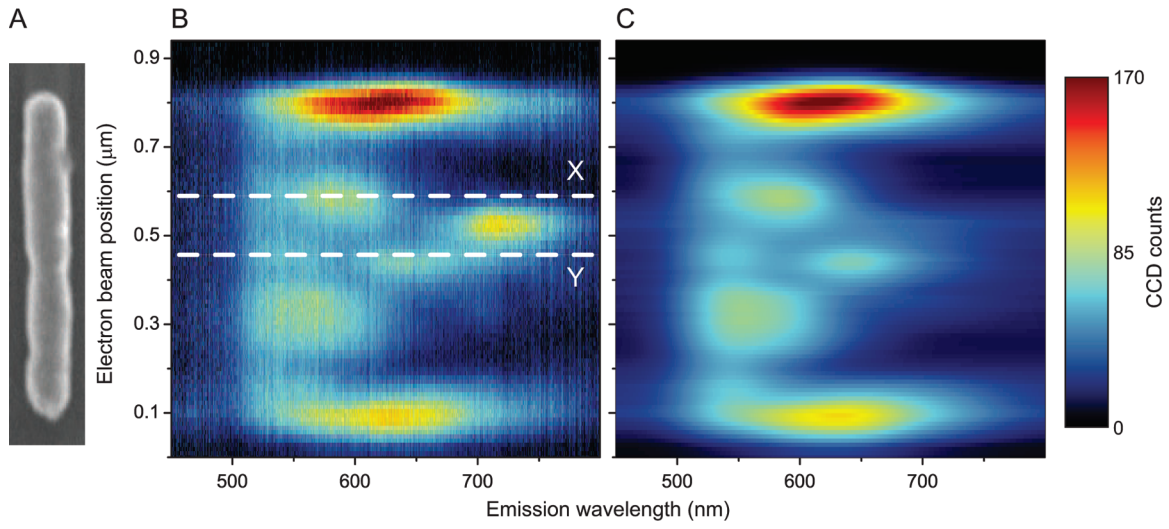

Figure 2. (a) SEM image of the Au nanowire. (b) Cathodoluminescence spectral line scan along the axis of a 725 nm long and ∼100 nm wide Au nanowire on Si. For every beam position (vertical axis), a spectrum is plotted (horizontal axis). The lines X and Y indicate the position at which the spectra that are plotted in Figure 3 are taken. (c) Reconstruction of the line scan image from the fit results, showing that the four resonant mode spectra that have been used represent all resonances of the wire. The artifact in (b) around 730 nm was removed before fitting because it does not correspond to a geometric resonance.

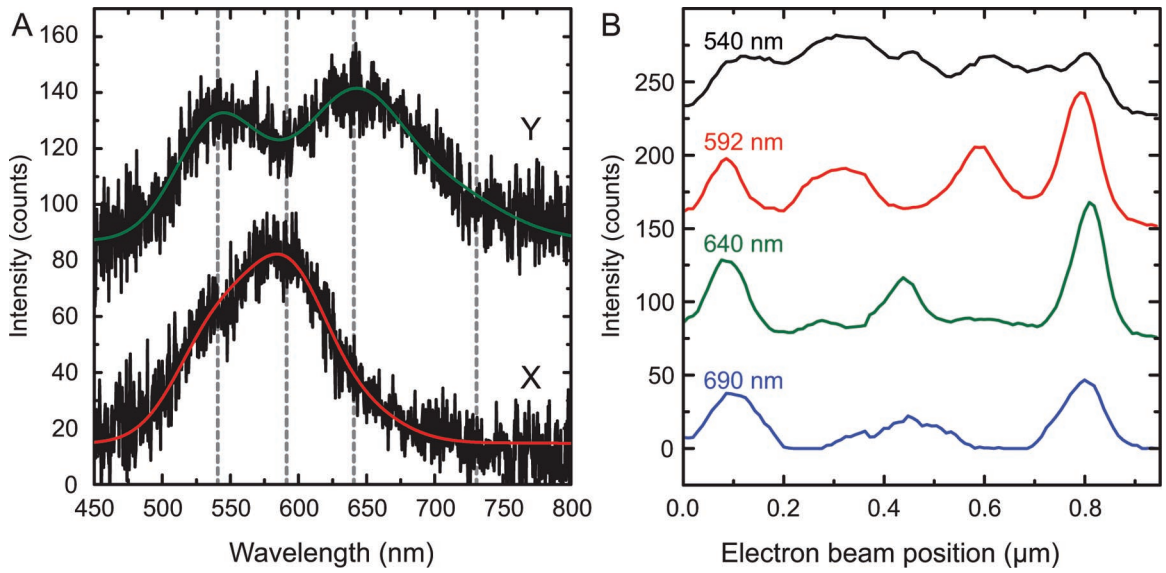

Figure 3. (a) CL spectra taken at two different positions along a 725 nm long Au nanowire (black curves). X and Y correspond to positions as indicated in Figure 2b. Also plotted in the figure are fits (green and red curves) to these data by a set of four Gaussian resonances at center wavelengths of 540, 592, 640, and 690 nm, respectively, indicated in the figure by vertical dotted lines. (b) Fitted intensity of each resonance plotted as a function of electron beam position. The 640 and 690 nm modes show three maxima, the 592 nm mode shows four maxima, and five maxima are vaguely observed for the 540 nm mode.

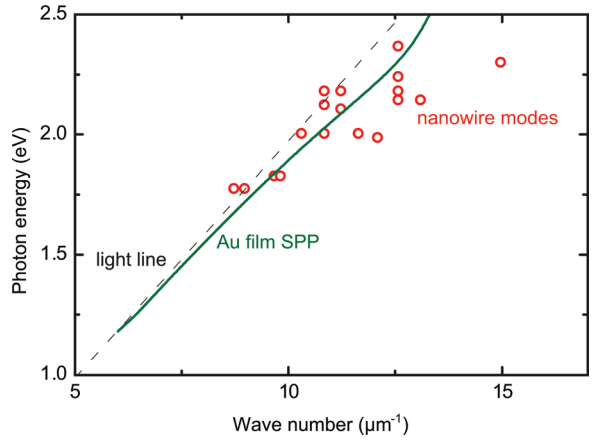

Figure 4. Experimentally determined dispersion of surface plasmon modes on Au nanowires with different lengths (circles). Dotted line: light line in vacuum. Solid line: dispersion relation of a SPP on an infinite Au film.

measurements are plotted in Figure 4. The SPP dispersion for a planar Au film as well as the vacuum light line have also been plotted. As can be seen in Figure 4, the wire modes are close to the dispersion relation for SPPs on a Au−vacuum interface.

Finally, we note that the spatial profiles in Figure 3b for 640 and 690 nm are quite similar. This is possibly due to the fact that the wires were not embedded in a homogeneous medium but were fabricated on top of a silicon substrate, which may cause splitting of wire resonances. More work is required to investigate this further.

In conclusion, we have used cathodoluminescence imaging spectroscopy to observe the plasmonic behavior of gold nanowires. We show that Au nanowires behave as plasmon resonators with eigenmodes with distinct spatial profiles. The simultaneous measurement of spectral and spatial characteristics of the modes allowed us to determine the dispersion of SPPs on Au nanowires.

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