Supplementary Information

Optical and Magneto-Optical Properties of Gold Core – Cobalt Shell Magnetoplasmonic Nanowire Arrays.

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Fabrication

Sputtered thin films are deposited on a glass substrate; 10nm of tantalum pentoxide (Ta_2O_5) as an adhesion layer, 8nm gold (Au) to form electrical contact at the bottom of the pores for electrodeposition, and approximately 300nm of aluminium (Al). The Al is anodised in a cold bath of 0.3M sulphuric acid at 35 Volts to create a quasi-hexagonal nanoporous array with centre-to-centre spacing of ~80nm. A chemical etch, using 30mM NaOH, is required to remove the oxide layer at the bottom of the pores and can be used to tailor the pore size to the required dimensions.

The fabrication of the Au-core / Co-shell structures uses a second chemical etch to re-widen pores before the deposition of the Co layer. Due to the high conductivity and large surface area of the Au wires, the result is a rapid Co deposition including the formation of a Co cap on top of the Au wires. The dimensions of the Au-core / Co-shell sample used in the study are shown in Fig. 1.

For the Co tubes however, a sacrificial polymer is first deposited in the pores after the same initial steps. The polymer used is Polypyrrol (Ppy), the chemistry of which is very different to metal solutions with regard to electrodeposition. A thicker anodised aluminium oxide (AAO) template is required (400nm) as the polymer tends to rapidly exceed the height of the pores at the edges of the sample. Also, ten seconds additional chemical etch is required as the low PH of the cobalt sulphate solution reduces the lateral dimensions of the Ppy, leaving room for electrodeposition of the Co shell. This growth is nucleated from the Au layer at the bottom of the pore, as Ppy is no longer conductive in its solid form, meaning there is no Co cap; provided the deposition is terminated before the tubes reach the critical length and completely surround the Ppy. This is another reason why the initial template and the length of the Ppy wires are larger than usual. The growth conditions are carefully monitored with *in-situ* optics and voltage/current settings. Figure S1 displays a high angle annular dark field (HAADF) image by scanning transmission electron microscopy (STEM). STEM imaging allows a fine electron probe, ~1nm, which can be freely positioned, to be focused onto the sample. The interaction of this electron probe with the sample produces characteristic x-rays making energy dispersive X-ray (EDX) line scans possible. Furthermore, HAADF-STEM acquisition mode provides a Z-contrast image, meaning that elements with higher atomic number give a higher contrast, providing simultaneous chemical and structural information.
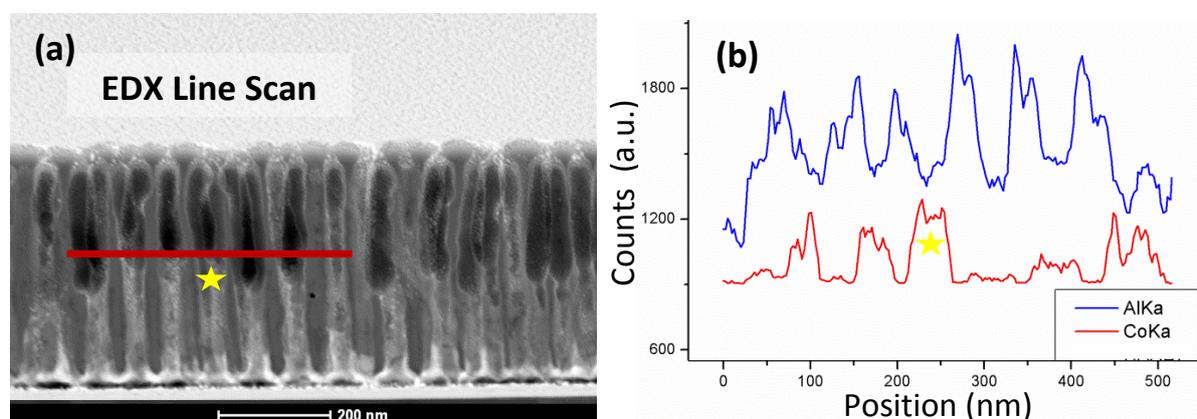
Figure S1 a) HAADF-STEM cross section image of hollow Co tubes b) EDX line scan (red line in (a)) showing the distribution of Co in the AAO template. The Al kα and Co kα signal – blue and red lines respectively.

From the STEM-HAADF image it can be seen that the height of the Co tubes varies around 200nm. However, from the EDX line scan we can see that in some pores there are still small traces of Co present at that height, despite appearing absent in the image. A closer look at the third tube from the left, marked with the yellow asterisk, reveals a spike in the counts of cobalt at each side of the tube. From both HAADF-STEM image and EDX, it can be concluded that the thickness of the Co shell is approximately 5nm. This is similar to the dimensions observed in Fig. 1 of the main manuscript. The controlled nature and *in-situ* monitoring of the growth procedure would indicate that the Co tubes are uniform in height, in the central section of the sample, and are indeed approximately the same length as the core/shell sample, 260nm. However, this is not seen in Fig. S1 due to the lamella preparation by focused ion beam (FIB) milling, which, has indiscriminately removed the cobalt shell in places despite the use of very low beam currents for the final series of cuts, thinning more the top of the sample. However, it is worth mentioning that it is at the bottom of the sample that the thickness of the Co tubes can be better appreciated. The volume of Co in the hollow tubes is very similar to that of the core – shell sample, the differences in geometry being a slightly larger wall thickness in the tubes but without the 15nm cap on top.

The Co wires made for comparison with Fig. 2b & 2c, were deposited from the same cobalt sulphate solution. No STEM analysis was conducted on this sample however it is known from the fabrication of many such samples that these dimensions also match those of the core/shell array; 40nm diameter, 260nm length.

Au and Co Nanowires
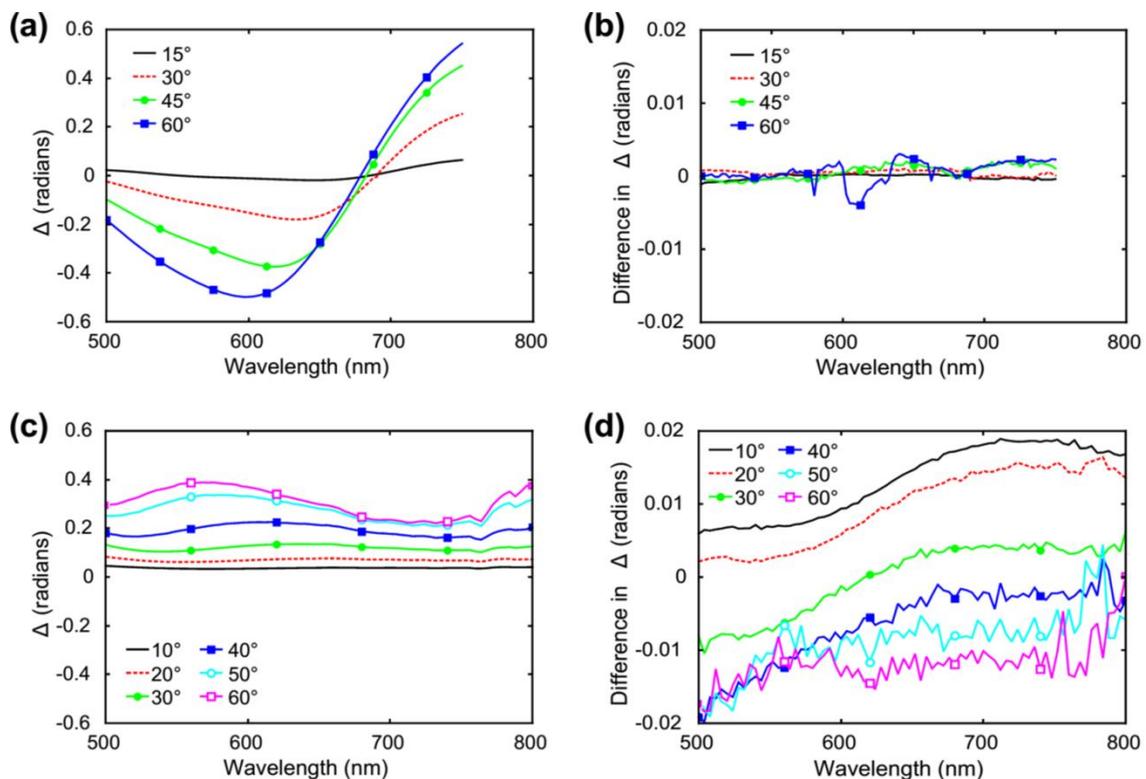
Figure S2 a) Ellipsometric measurements of Del at various AOI's for a Au nanowire array, b) changes in Del in a Au nanowire array with switching the direction of magnetic field, c) Del at varying AOI for a Co nanowire array, d) changes in Del for Co wires by reversing the direction of magnetic field.

Figure S2 demonstrates the difference between Co and Au nanowire arrays. The Au array has a phase shift consonant with plasmonic resonance but cannot be affected by externally applied magnetic fields (the difference in Del at 60 degrees is attributed to noise). The Co array has a low valued, featureless value of Del for all AOIs and across the wavelength range, similar to the Co tubes, but we can induce a change in signal be applying an external magnetic field in opposite directions. Figure 4, however, shows the advantage of the Au-core / Co-shell nanowires. The phase shift displays characteristics consistent with plasmonic resonance, and we can also induce a change in signal with externally applied magnetic fields.