Figure Captions:

Figure. 2. Scanning electron microscope (SEM) images of biotemplated CoPt nano- and micro‑patterned line surface. (a) Zoomed out and (b) zoomed in SEM image of biotemplated nano-lines. (c) Annotated version of image (b) to show areas functionalised with biotemplating peptide (cys\_CoPt, average width (AW) is 226 ± 20 nm), which are mineralised with a closely packed layer of MNPs, and areas functionalised to resist mineralisation (PEG, AW is 109 ± 13 nm). (d) Micro-patterned lines of dark contrast are the gold surface that was protected against biomineralisation by the μCP PEG thiol (AW is 6.1 ± 0.4 μm). Lines of light contrast were backfilled with the cys\_CoPt peptide before metallisation with CoPt, and are covered in a biotemplated layer of MNPs (AW is 8.1 ± 0.3 μm). (e) A closer view of an edge of a patterned line showing the clean, unbiomineralised gold (bottom of image) and the surface biotemplated MNPs (top of image). (f) A closer view of the biotemplated MNPs, showing a consistent layer of smaller MNPs. Scale bars: (d) 20 μm, (a & e) 2 μm, (b, c & f) 200 nm.

Figure 3. Electron microscopy and grainsize analysis of surface biotemplated CoPt and appropriate controls. Transmission electron microscopy (TEM) images of (a) particles precipitated from a bulk solution in the absence of a biotemplating peptide (**blue**), (b) in the presence of the biotemplating peptide (**green**) and (d) the cys\_CoPt peptide (**orange**). (e) Scanning electron microscope (SEM) image of the patterned biotemplated surface showing a lines pattern (black). (c) Grainsize distribution graph fitted with Gaussian distributions and (f) aspect ratio fitted with Extreme fit in Origin. Scale bars (a, b & d) 20 nm and (e) 200 nm. Text files of grainsize (c) and shape (f) data displayed in figure in data files.

Figure. 4. Electron microscopy, energy dispersive X-ray (EDX) maps and spectra, and X-ray Diffraction (XRD) spectra from the biotemplated patterned surface. (a) SEM image of a biotemplated patterned surface (scale bar 20 μm). Maps to show relative abundances of (b) gold, (d) cobalt and (e) platinum of the area shown in (a) (recorded at 15 keV). (c) EDX spectra and (f) XRD spectra from the biomineralised part of the pattern (**black**) and the non-biomineralized part of the surface (**grey**). The peaks indicated with asterisks (\*) are a good match for some carbon peaks (e.g. see Wang *et al.*)1. Spectra are offset vertically to show the reflections more clearly, and peak positions and assignment is summarised in Table S1. EDX and XRD spectra displayed in the figure are available in data files.

Figure. 5. MFM plots of biotemplated CoPt patterned surface (separate plots show in **Fig.** **S12** & **S13**). A negative phase shift indicates attraction (**red**) between the tip and the surface, and a positive phase shift indicates repulsion (**blue**). The topography is recorded in tapping mode and the phase shift recorded at a lift height of 50 nm (a) 2.5 μm2 scan area surface and (b) 1 μm2 of IL nano‑patterned biotemplated CoPt. (c) 25 μm2 and (d) 5 μm2 scan area of μCP micro-patterned biotemplated CoPt surface. Green area in (c) highlights the location scanned for plot (d). 5 μm2 scan area recorded at a 90° angle to image (c). These IL and μCP surfaces show zones of attraction and repulsion that extend across multiple MNPs on the surface. Separate topography and phase information plots available as text files.

Fig. S2. Scanning electron microscope (SEM) images of biotemplated patterned lines, biomineralised at 18°C at (a) low, (b) medium and (c) high magnifications. Lines of dark contrast are the gold surface that was protected against biomineralisation by the μCP PEG thiol. Lines of light contrast were backfilled with the biotemplating cys\_CoPt peptide before metallisation with CoPt, and are covered in a biotemplated layer of MNPs. Scale bars (a) 100 μm, (b) 20 μm, and (c) 2 μm.

Fig. S3. Scanning electron microscope (SEM) images of biotemplated patterned squares, biomineralised at 18°C at (a) low, and (b) high magnifications. Light contrast squares are peptide biotemplated CoPt MNPs, dark contrast background is protected against metallisation by μCP PEG thiol. Scale bars (a) 10 μm and (b) 2 μm.

Fig. S4. Scanning electron microscope images (SEM) of biotemplated patterned lines metallised at a higher temperature of ≈35°C. (a) The areas functionalised with the cys\_CoPt biotemplating peptide (dark contrast) are unable to template MNPs. At higher magnification (b) the biotemplating areas can be seen to be coated in a thin discontinuous layer of black and white speckling. It is likely that the higher temperatures in the lab in summer significantly increased the rate of the metallisation reaction. The higher temperatures may also have reduced the solubility of CoPt MNP precursors and/or inhibited the ability of the CoPt biotemplating peptide immobilised on the surface to bind to the forming particles. For any or all of these reasons, the biotemplating peptide is not able to control the mineralisation of MNPs onto surfaces at 35˚C, but instead forms this thin, discontinuous film. Scale bars (a) 100 μm and (b) 1 μm.

Fig. S5. Energy dispersive X-ray (EDX) spectra from the powder control samples recorded in the TEM. All samples show peaks that pertain to the formvar carbon coated copper grids (Cu, C, O, Si), and chlorine is probably from the buffer the peptide was stored in (PBS). The non-peptide bulk templated particles (**blue**) also show peaks for Co and Pt, with quantification showing atomic percentage ratio of 25:75. Quantification of the non-cysteine tagged peptide bulk templated particles (**green**) Co:Pt is 88:12, and the cys\_CoPt bulk templated particles (**orange**) is 59:41. The stoichiometry of Co:Pt in the metallisation solution was 75:25, and the ideal ratio for L10 CoPt is 50:50. Despite the large excess of Co in the mineralisation solution, the bulk precipitated particles are dominated by Pt, so much of the Co must remain unreacted in the solution. It is likely that much of the Co detected in the peptide templated samples may be bound by the organic peptide matrix. As there was no detection of Co reflections in the SAED (see Fig. S7 below), this indicates that the peptide is able to bind cobalt well, and may form an amorphous or poorly crystalline cobalt phase that can be seen to surround the MNPs.

Fig. S6. X-ray diffraction (XRD) data for the powder controls. Bulk precipitated (**blue**), bulk peptide biotemplated (**green**) and bulk cysteine tagged peptide biotemplated (**orange**) powders show strong peaks for CoPt3 rather than the CoPt L10 structures. There are also two peaks, labelled with asterisks (**\***) that are likely to be due to carbon in this biotemplated sample. Scans are vertically offset for clarity, and details of peak assignments are shown in Table S1.

Fig. S7. Transmission electron microscope (TEM) and selected area electron diffraction (SAED) of the controls for the biotemplated surface MNPs. The TEM images (a, c & e) show clusters of multiple particles, which are likely self-assembled due to magnetic interactions between the particles. This made imaging and diffraction on these samples difficult. (a) TEM image of bulk precipitated particles in the absence of any biotemplating peptide (bulk) and (b) SAED pattern showing reflections for A1 CoPt. (c) TEM image of MNPs templated with a non-cysteine tagged version of the CoPt peptide in the bulk solution (peptide), with the particles imbedded in a matrix of material that looks like it is organic. (d) SAED pattern from this sample, showing reflections for CoPt3 (111) and A1 CoPt (112) and (212). (e) TEM image of MNPs templated by the cysteine tagged CoPt templating peptide in the bulk solution (cys\_CoPt), again the particles seem to be embedded in a matrix that looks like organic material. (f) SAED diffraction pattern from the cys\_CoPt peptide in the bulk solution templated particles, showing L10 CoPt reflections (003) and A1 reflections (212) and (301). Details of indexing is shown in Table S2. Scale bars 50 nm.

Fig. S8. Vibrating sample magnetometry (VSM) measurements of a biotemplated CoPt surface. Loops were recorded with the field applied perpendicular (**red**) or parallel (**black**) to the sample surface. The lower gradient (loop shear) seen in the perpendicular loop (**red**) may be due to shape anisotropy of the film contributing to demagnetising effects.

Fig. S9. Magneto-optical Kerr effect (MOKE) measurements of a biotemplated CoPt surface. There is little difference between the width of the hysteresis loops (the coercivity) measured perpendicular (**red**) or parallel (**black**) to the sample surface. The loop shear seen in the perpendicular VSM measurements (Fig. S8) is also observed in these MOKE measurements.

Fig. S10. Hysteresis loops recorded using MOKE on biotemplated surfaces formed (a) perpendicular and (b) parallel to a 0.2 T DC field. The loops show that the samples are ferromagnetic, but have low coercivity. Samples biomineralised with the field applied out-of-plane (oop, (a)) and in-plane (ip, (b)). The hysteresis loops were recorded at 90˚ to the surface (polar) and then parallel to the surface at two different angles (longitudinal 0˚ and 90˚) to measure the magnetic response of the surfaces in the *x*, *y*, and *z* planes. Again, the MNPs have a more rapid switching when the field is applied in-plane for the measurements when compared to it being applied out-of-plane.

Fig. S11. Scanning electron microscopy (SEM) of unpatterned biotemplated surfaces metallised in the presence of an applied field of 0.2 T. (a & b) Biotemplated CoPt MNPs formed when the field is applied parallel to the surface during mineralisation. (c & d) Biotemplated CoPt formed when the field is applied perpendicular to the surface during mineralisation. Scale bars (a & c) 2 μm and (b & d) 200 nm.

Fig. S12. Separate magnetic force microscopy (MFM) of nano-patterned surfaces. (a & b) Tapping mode height, and (c & d) the respective phase contrast due to magnetic interactions between the magnetised tip and the IL nano-patterned MNPs biotemplated onto the surface. The magnetic nanoparticles show mainly repulsion (**light**) that extend across multiple MNPs on the surface. These same plots are shown combined in Fig. 5a & b.

Fig. S13. Separate MFM of micro-patterned surfaces. Separated scans to show the height contrast (a & b) and corresponding phase contrast in non-contact mode (c & d) of the μCP MFM plots also shown in Fig. 5c & d. 25 μm2 scan area of biotemplated μCP CoPt line patterned surface, a topography recorded in tapping mode and c phase shift recorded at a lift height of 50 nm. There is significant attraction of a few degrees between the magnetised tip and the patterned biotemplated CoPt MNPs. 5 μm2 scan area (b) topography and (d) phase contrast recorded at a lift height of 50 nm and a 90° angle to image (a). Here, the gold substrate appears as close to zero (i.e. non-magnetic) in the phase shift when compared to the MNPs. The magnetic nanoparticles clearly show zones of attraction (**dark**) and repulsion (**light**) that extend across multiple MNPs on the surface.

Fig. S14. Combined MFM plots of same area of line micro-patterned biotemplated CoPt MNPs, scanned at a 45° angle to the line pattern. (a & b) colour keys to show phase shift in plots (c & d) respectively A negative phase shift indicates attraction (**red**) between the tip and the surface, and a positive phase shift indicates repulsion (**blue**). (c) 2 μm2 scan area of biotemplated CoPt surface, topography recorded in tapping mode and phase shift recorded at a lift height of 30 nm. (d) 2 μm2 scan of the same area recorded at a lift height of 30 nm with the scan direction rotated by 90°. Both plots show similar zones of attraction and repulsion that extend across multiple MNPs on the surface, parallel to the long axis of the line pattern. These are shown as separated height and magnetic plots in Fig. S15.

Fig. S15. Separate MFM plots of the same area of line micro-patterned biotemplated CoPt MNPs, scanned at + and - 45° angle to the line pattern, also shown in Fig. S14 as combined plots. (a & b) Height plots recorded in tapping mode at a 90˚C angle to each other, and (c & d) the respective phase contrast recorded at a lift height of 30 nm. Arrows indicate the direction of the long axis of the micro-patterned lines on the surface. The magnetic nanoparticles show zones of attraction (**dark**) and repulsion (**light**) that extend across multiple MNPs on the surface. Magnetic phase contrast appears to align with the long axis of the pattern, indicating that shape anisotropy of the assembly influences the magnetic alignment of the biotemplate MNPs.

Fig. S16. Combined MFM plots of biotemplated CoPt micro-patterned surface. (a & b) colour keys to show phase shift in plots (c & d) respectively A negative phase shift indicates attraction (**red**) between the tip and the surface, and a positive phase shift indicates repulsion (**blue**). (c) 5 μm2 scan area of biotemplated CoPt line patterned surface, topography recorded in tapping mode and phase shift recorded at a lift height of 50 nm, green area highlights area scanned for image (d). d, 2 μm2 scan area recorded at a lift height of 30 nm and a 90° angle to image (c). Again, both plots clearly show zones of attraction and repulsion that extend across multiple MNPs on the surface. Again, these zones appear to run roughly parallel to the long axis of the patterned biotemplated lines, even when the scale of the image and the scan direction is changed. These are shown as separate height and magnetic contrast plots in Fig. S17**.**

Fig. S17. Separate MFM plots of micro-patterned surfaces. (a & b) Tapping mode height, and (c & d) the respective phase contrast due to magnetic interactions between the magnetised tip and the MNPs biotemplated onto the surface. The magnetic nanoparticles show significant zones of attraction (**dark**) and repulsion (**light**) that extend across multiple MNPs on the surface. These same plots are shown combined in Fig. S16.

Fig. S18. Combined MFM plots of biotemplated CoPt unpatterned surface. (a & b) colour keys to show phase shift in plots (c & d) respectively A negative phase shift indicates attraction (**red**) between the tip and the surface, and a positive phase shift indicates repulsion (**blue**). (c) 10 μm2 scan area of biotemplated CoPt surface, topography recorded in tapping mode and phase shift recorded at a lift height of 50 nm. (d) 2 μm2 scan area recorded at a lift height of 30 nm. Both plots clearly show zones of attraction and repulsion that extend across multiple MNPs on the surface. These appear to wider than those observed on the biotemplated lines pattern. They also do not appear to have any preferred alignment or orientation. These are shown as separate height and magnetic plots in Fig. S19.

Fig. S19. Separate MFM plots of unpatterned biotemplated CoPt surfaces. (a & b) Tapping mode height, and (c & d) the respective phase contrast due to magnetic interactions between the tip and the unpatterned MNPs biotemplated onto the surface. The magnetic nanoparticles show significant zones of attraction (**dark**) and repulsion (**light**) that extend across multiple MNPs on the surface. These same plots are shown combined in Fig. S18.

Bibliography

1 Y. Wang, J. E. Panzik, B. Kiefer and K. K. M. Lee, *Sci. Rep.*, 2012, **2**, Article No. 520.