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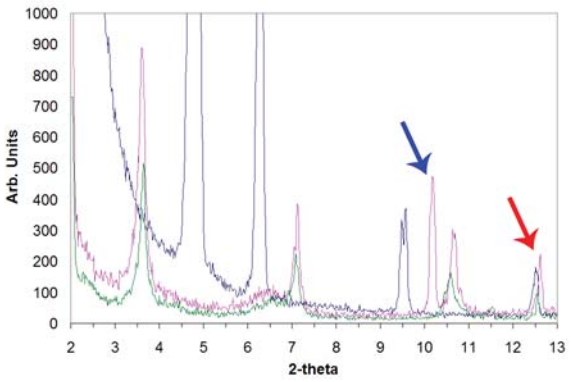
FIGURE S1



XRD plot of homo- and mixed-ligand nanoparticles.
The pink and green curves were obtained from diffraction of **OT:MPA** 2:1 silver nanoparticles (3.8 nm in diameter). The particles were first analyzed at room temperature (green curve) and subsequently heated at 100 °C for 1 hour and allowed to cool down at room temperature slowly. This type of heating cycle is known to help the formation of interdigitated super-lattices of nanoparticles, and consequently to generate new peaks in the XRD spectrum or to enhance the intensity of existing ones (see reference 27). As evident from the comparison of the pink (spectrum taken on the same sample after thermal treatment) and the green curves, new peaks appear (one of them is indicated by the blue arrow) but a few peaks stay unmodified. These latter peaks were assigned to the ordered domains formed on the nanoparticles' ligand shell. In particular, the peak indicated by the red arrow ($2\theta = \sim 12.5°$) indicated a domain spacing of ~0.7 nm, similar to the STM measured spacing (0.92 nm see table). The blue curve is an XRD plot of **OT:MPA** 2:1 gold nanoparticles (5.1 nm in diameter). In this case the ripple spacing measured with XRD ($2\theta = \sim 12.5°$, equivalent to 0.7 nm) agrees even better with the one measured in STM (0.72 nm, see table). These curves are representative of all of the curves that we obtained. The homo-ligand nanoparticles showed no temperature independent peak at small angles, while the mixed ligands showed one or more temperature independent peak at 2θ ranging from 2.5° to 13°

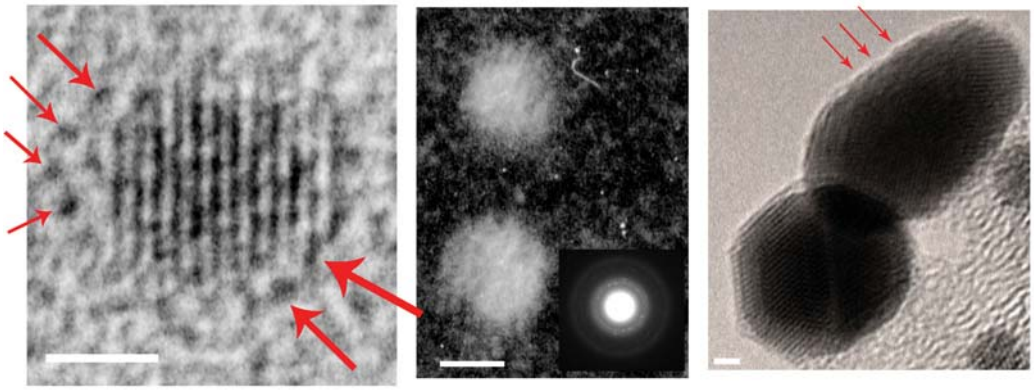


FIGURE S2

Transmission electron microscopy of OT: MPA (2:1) gold nanoparticles.
a, Bright field image of a nanoparticle showing quasi-ordered dots spaced 0.5 nm around the particle. These dots are attributed to ions captured but the MPA head groups. **b,** Dark field image of the same sample acquired by positioning the detector at the first diffraction angle (see inset) corresponding to a 0.55 nm spacing. It should be noted that in dark field the particle diameter appears smaller. We believe that this is due to the fact that the image is created by the ripples diffracting. The alignment of the ripples' planes and their number are lower at the edges of the nanoparticles, thus the diffracting area of a particle is smaller than that of the whole particle. This same effect seems to rule out the fact that the images are produced by high diffraction order scattering of gold planes. **c,** Bright field image on a holey carbon grid, The imaged particles are shown extending over the hole. The red arrows show the sinusoidally shaped contour of the ligands that coat the particles. Scale bars 1 nm.

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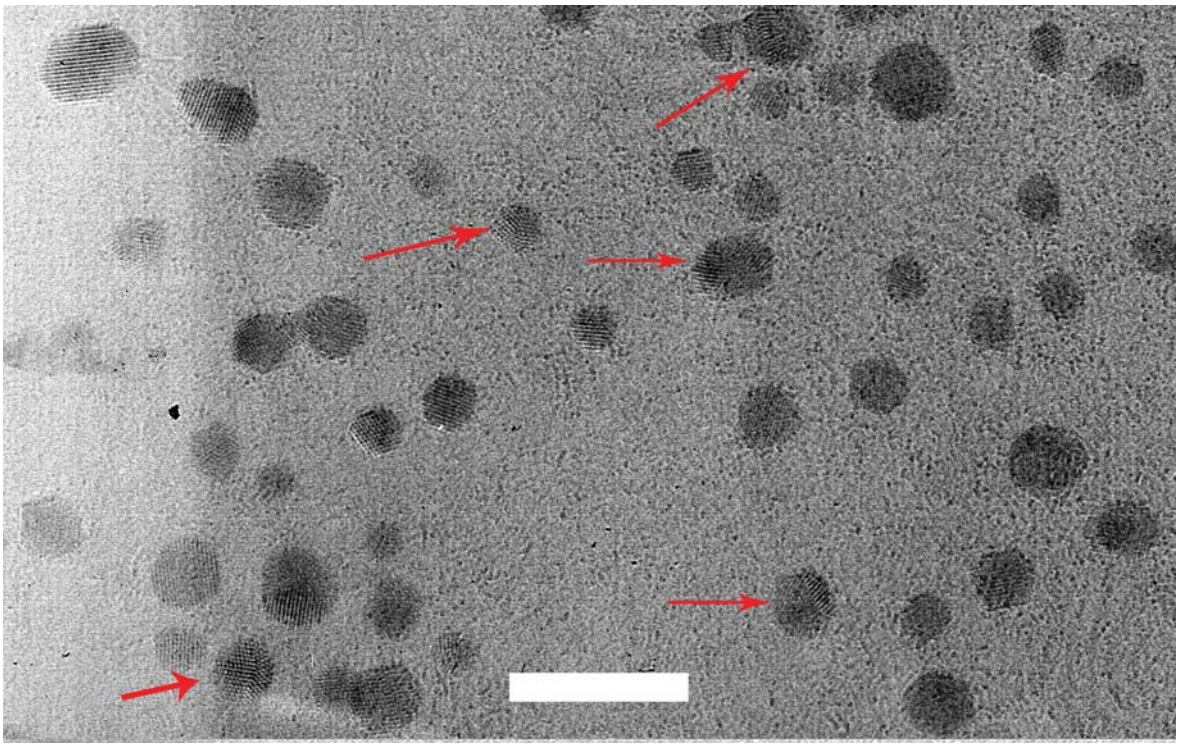

FIGURE S3

TEM image of MPMN coated with OT:MPA 2:1 mixture.
The arrows point to twinned nanoparticles. In the corresponding STM images (**Fig. 1**) there is no sign of such a phenomenon occurring on the ligand shells. Scale bar 10 nm.

Table 1

Metal Nanoparticles' Parameters

Metal	Ligand[a] 1	Ligand[a] 2	Au:Total Ligand (molar ratio)	Ligand 1: Ligand 2 (molar ratio)	Morphology [b]	Periodicity[c] (nm)	Solubility[d] Toluene	Ethanol	Average Diameter[e] (nm)
Au	HT	MPA	1:1	2:1	R	0.95	4	3	3.5
Au	OT	MPA	1:1	2:1	R	1	4	1	3.8
Au	DT	MPA	1:1	2:1	R	1	4	2	3.5
Au	DDT	MPA	1:1	2:1	R	.55-.75[f]	4	1	3.5
Au	OT	MUA	1:1	2:1	R	1	1	4	3.7

Au	DDT	MUA	1:1	2:1	R	0.62	1	3	3.7
Au	OT	MPA	1:1	1:2	R	0.66	4	2	3.6
Au	OT	MPA	1:1	2:1	R	1	4	2	3.8
Au	OT	MPA	1:1	10:1	D	--	4	2	3.5
Au	OT	MUA	1:1	2:1	R	1	1	4	3.7
Au	OT	MUA	1:1	5:1	D	--	2	4	3.6
Au	OT	MUA	1:1	20:1	D	--	4	1	3.6
Au	OT	MPA	1:1	2:1	R	1	4	1	3.8
Au	OT	MPA	2:1	2:1	R	0.82	4	1	4.3
Au	OT	MPA	5:1	2:1	R	0.73	4	2	5.1

Au	OT	MUA	1:1	2:1	R	1	1	4	3.7
Au	OT	MUA	3:1	2:1	R	0.8	1	4	4.9
Au	HT	APT	1:1	1:2	R	0.6	4	1	3.5
Au	OT	APT/MUA	1:1	1:1:1	D	--	1	3	3.6
Au	OT	MPA	3:1	30:1	--	--	4	1	
Au	OT	MPA	1:1	substituted	partial ripples	0.75	1	1	3.6
Ag	OT	MPA	1:1	2:1	R	0.92	2	2	3.8

a) $HOOC\text{-}(CH_2)_2\text{-}SH$, **MPA**; $HOOC\text{-}(CH_2)_{11}\text{-}SH$, **MUA**; $H_2N\text{-}C_6H_4\text{-}SH$, **APT**; $CH_3\text{-}(CH_2)_n\text{-}SH$ n=5, **HT**, n=7, **OT**, n=9, **DT**, n=11, **DDT**; 1b) R= Ripples; D= Domains; c) Peak to peak spacing as determined by STM images; d) all sample were prepared as described in the experimental section, 4= highly soluble, i.e. no precipitation visually observed, 3= soluble, i.e. little precipitation observed over time with consequent slight decoloration of the solution; 2= slightly soluble, i.e. most of sample precipitated but a small coloration of the solution remains, 1= totally insoluble. All observations performed 1 month after stirring of the solution was stopped. e) metallic core size as determined by TEM. f) The spacing is probably highly affected by the chain mobility of the **DDT**.