

## Supplementary information

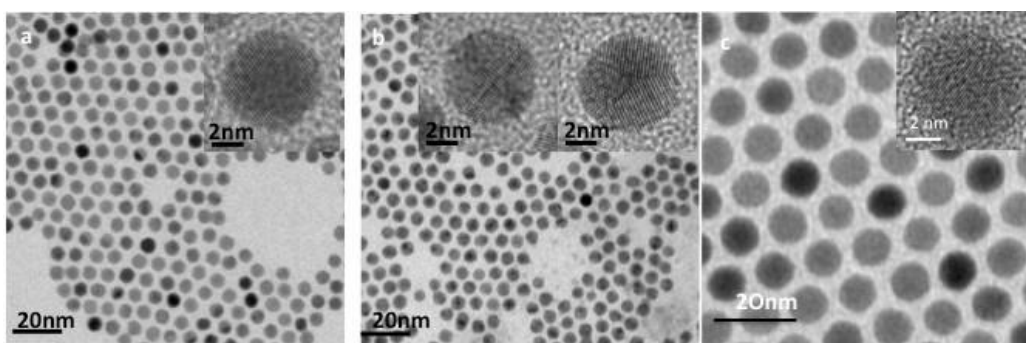
# Au nanocrystal superlattices: nanocrystallinity, vicinal surfaces, and growth processes

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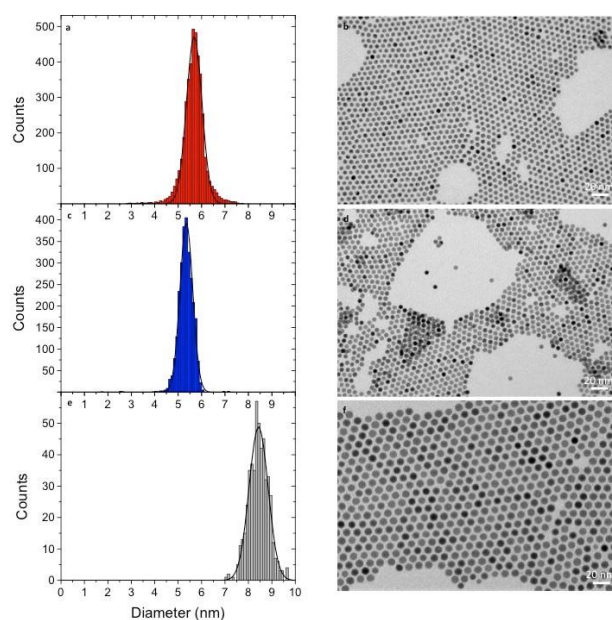
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b) CEA/IRAMIS, CEA Saclay, 91191, Gif-sur-Yvette, France

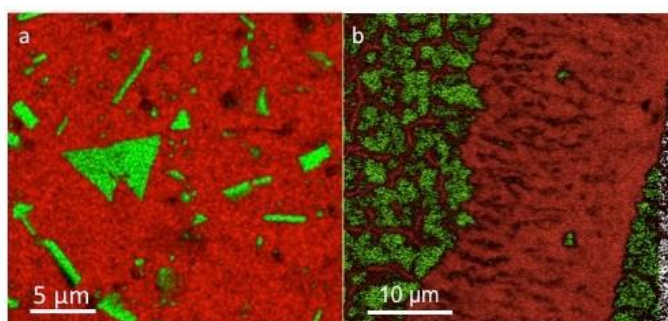
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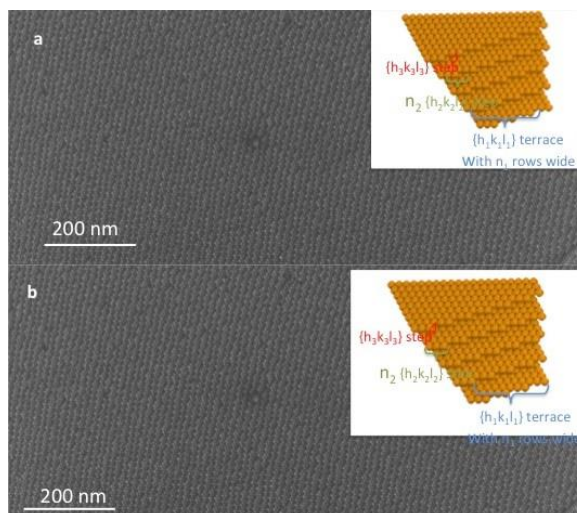
**Figure S1:** TEM images of Au<sub>single</sub> (a) Au<sub>poly</sub> (b) and Co(ε) (c) NCS. Insets corresponding HRTEM images



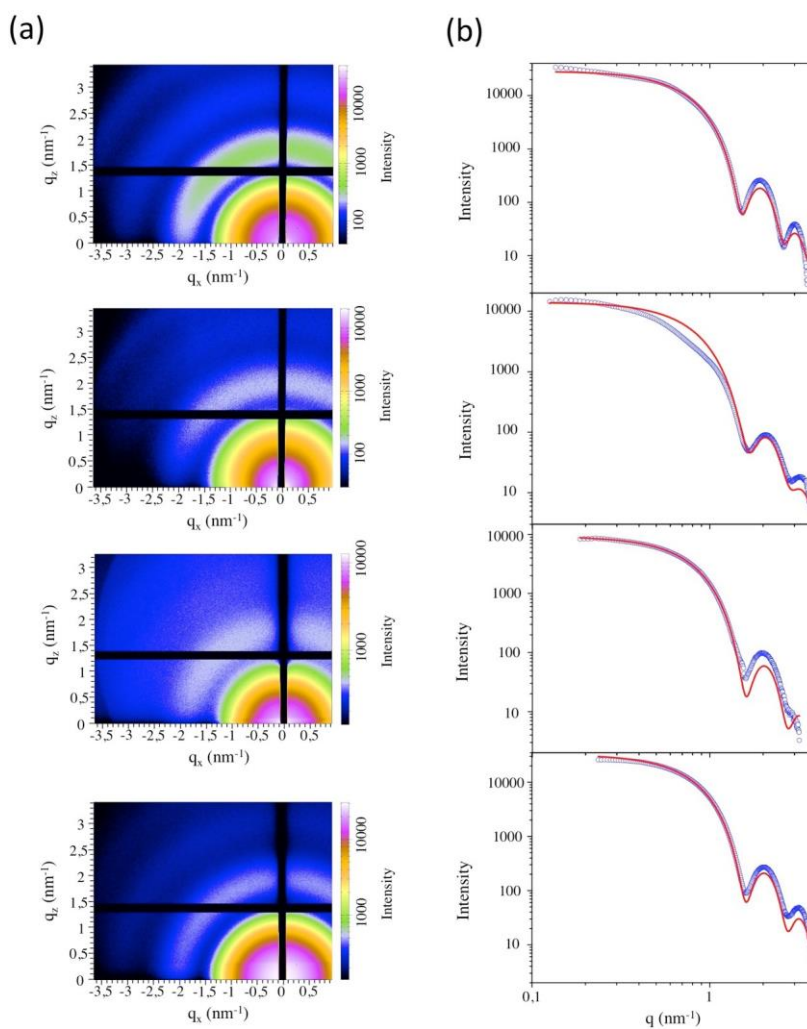
**Figure S2.** Au<sub>poly</sub> (red), Au<sub>single</sub> (blue) and Co(ε) (grey) nanoparticle size distributions (a) and their corresponding TEM pictures (b).



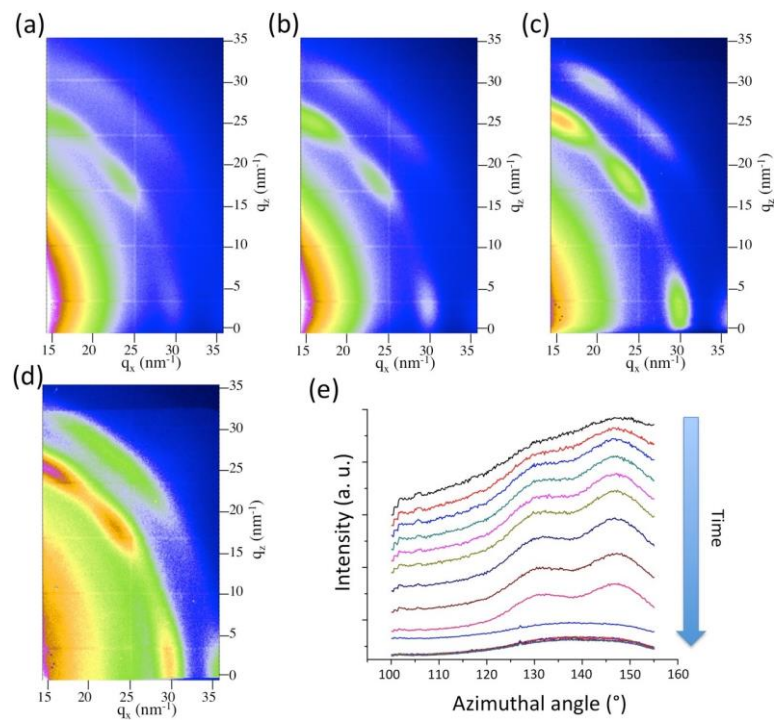
**Figure S3.** Elemental maps corresponding to L<sub>α1</sub> of Au (green) and K<sub>α1</sub> of Co (ε) NCS (red). (a) Self-assembly of Au<sub>single</sub> and Co(ε) NCS. Note the phase separation of Au<sub>single</sub> and Co supracrystals with well defined Au shape supracrystals. (b) Self-assembly of Au<sub>poly</sub> and Co(ε) NCS. Note that Au<sub>poly</sub> and Co(ε) NCS form smaller grains superlattices.



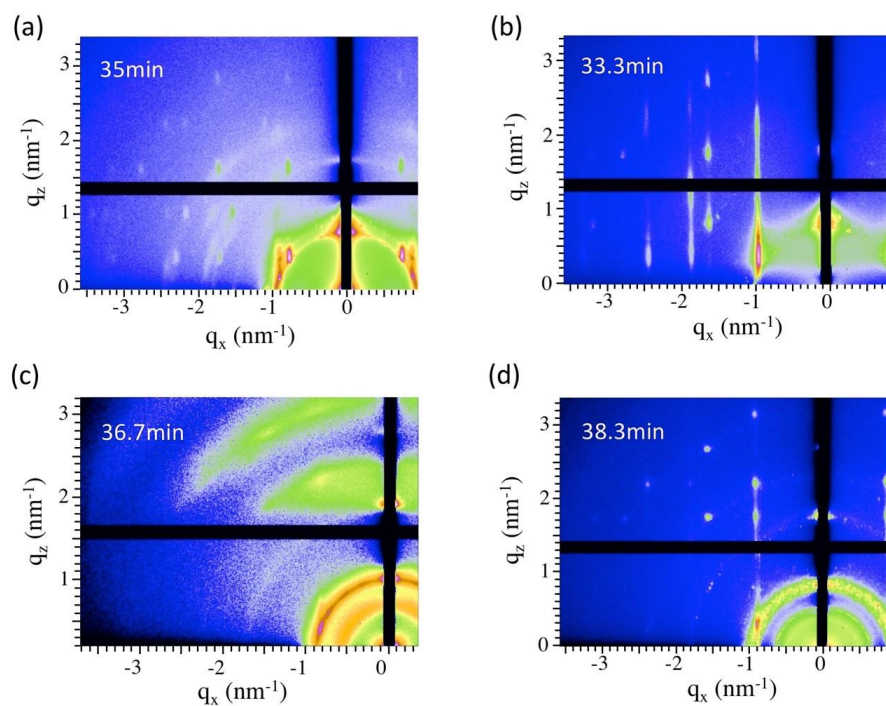
**Figure S4.** Various vicinal surfaces of Au<sub>single</sub> supracrystals in presence of Co( $\epsilon$ ).



**Figure S6.** GISAXS patterns (a) and their corresponding radially integrated intensity profiles (b) of the initial colloidal solution drop for Au<sub>single</sub>/Co( $\epsilon$ ), Au<sub>poly</sub>/Co( $\epsilon$ ), Au<sub>single</sub> and Au<sub>poly</sub>, from the top to bottom.



**Figure S7.** Evolution of GIWAXS patterns of  $\text{Au}_{\text{single}}$  NCs at 20s (a), 50s (b), 100s (c) and 140s (d). Evolution of corresponding WAXS profiles (e).



**Figure S8** GISAXS patterns after final evaporation of  $\text{Au}_{\text{single}}$  (a),  $\text{Au}_{\text{poly}}$  (b),  $\text{Au}_{\text{single}}/\text{Co}(\epsilon)$  (c) and  $\text{Au}_{\text{poly}}/\text{Co}(\epsilon)$  (d).

		Toluene atmosphere	Hexane atmosphere
		Without Co	With Co
$Au_{\text{single}}$	$a_{\text{compressed\_fcc}} = 12.3\text{nm}$ $c_{\text{compressed\_fcc}} = 11.2\text{nm}$ $d_{\text{pp}} = 3.3\text{nm}$	$a_{\text{fcc}} = 12.3\text{nm}$	$a_{\text{fcc}} = 12.4\text{nm}$
		$d_{\text{pp}} = 3.3\text{nm}$	$d_{\text{pp}} = 3.4\text{nm}$
$Au_{\text{poly}}$	$a_{\text{hcp}} = 8.2\text{nm}$ $c_{\text{hcp}} = 13.5\text{nm}$ $d_{\text{pp}} = 2.6\text{nm}$	$a_{\text{hcp}} = 8.2\text{nm}$ $c_{\text{hcp}} = 13.4\text{nm}$	$a_{\text{hcp}} = 8.2\text{nm}$ $c_{\text{hcp}} = 13.2\text{nm}$
		$d_{\text{pp}} = 2.6\text{nm}$	$d_{\text{pp}} = 2.5\text{nm}$

**Table S1.** Au SC parameters differing by their nanocrystallinity, single domain ( $Au_{\text{single}}$ ) and polycrystals ( $Au_{\text{poly}}$ ), deduced from GISAXS patterns in the various experimental conditions. Let admit an error of 0.1nm no influence of Co( $\epsilon$ ) NCs.

Sample	$D_{\text{SAXS}}$	$\sigma_{\text{SAXS}}$	$D_{\text{TEM}}$	$\sigma_{\text{TEM}}$
$Au_{\text{single}}$	5.54	0.40	5.4	0.4
$Au_{\text{poly}}$	5.56	0.40	5.6	0.4
$Au_{\text{single}} + \text{Co}$	5.84	0.44		
$Au_{\text{poly}} + \text{Co}$	5.30	0.54		

**Table S2.** NC diameter determined by SAXS ( $D_{\text{SAXS}}$ ) and TEM ( $D_{\text{TEM}}$ ) with their corresponding standard deviation ( $\sigma$ ).