SUPPORTING INFORMATION

# Influence of Anisotropy on Heterogeneous Nucleation with Gold Nanorods

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### 1 Controlled Nucleation in Solution



**Figure S 1. Controlled nucleation of gold nanorods in dispersion.** UV-vis-NIR spectra of the destabilisation process of gold nanorods with different aspect ratios of A) 3.00, B) 2.25, and C) 1.75. The CTAB stabilised gold nanorods can be destabilised in a controlled manner by the addition of ethanol. During the destabilization process, the longitudinal plasmonic bands centered at A) 830 nm, B) 730 nm, and C) 630 nm of single gold nanorods decrease while new bands at longer wavelengths emerge.

#### 2 Reaction Order

**Table S 1. Determination of the reaction order.** Determination of the reaction order for the nucleation of gold nanorods in solution with aspect ratios of 3.00, 2.25, and 1.75 (Figure S 1). The optical density at the respective absorption maxima is plotted linearly against the experiment time dependent on the assumed reaction order. The R<sup>2</sup> values, which describe the quality of the linear fit, are given.

assumed reaction order	plot	3.00	2.25	1.75
0. order	Abs → t	0.58	0.81	0.73
1. order	ln(Abs) → t	0.69	0.89	0.89
2. order	1/Abs → t	0.79	0.93	0.93

#### 3 Interaction with Substrates



**Figure S 2. Effect of sulfonate-functionalised surfaces on the nucleation process.** Gold nanorod dispersions with an aspect ratio of 3.00 were destabilised by adding ethanol in the presence of mica substrates functionalised with sulfonate. Two hours after adding ethanol, the UV-vis-NIR spectra of the investigated gold nanorod dispersions show that sulfonate-functionalised mica can trigger the nucleation of gold nanorod-based superstructures (red), while in their absence such processes do not occur (green). The experiment was repeated four times.

**Table S 2. Counted gold nanorod superstructures.** The superstructures formed by gold nanorods with different aspect ratios were analyzed with SEM and the percentage of the analyzed substrate surface covered with superstructures, the average size of superstructures from sizes of 1  $\mu$ m<sup>2</sup> and bigger, and the size of the biggest superstructure are given.

aspect ratio	% area	average size [µm²]	biggest superstructure [µm²]
3.00	1.85	2.18	4
2.25	6.76	2.21	15
1.75	11.62	3.62	23



**Figure S 3. Structure area dependent on the nanoparticle anisotropy.** Superstructures of gold nanorods formed on sulfonate-functionalized substrates with different aspect ratios of A) 3.00, B) 2.25, C) 1.75 were analyzed with SEM. The structure area of the counted superstructures are plotted in histograms.

#### 4 Simultaneous UV-vis-NIR Measurement



**Figure S 4. Simultaneous UV-vis-NIR measurement.** UV-vis-NIR spectra of gold nanorod dispersions with aspect ratios of A) 3.00, B) 2.25, and C) 1.75. The respective absorption maxima remain constant throughout the experiment time, indicating that the gold nanorod dispersions are stable, and the particles are not aggregating in dispersion.

# 5 Interfacial Energies

**Table S 3. Interfacial Energies.** Interfacial energies were determined for the heterogeneous nucleation of gold nanorods on sulfonate-functionalised mica substrates. A contact angle of 60 to 120° is assumed.

	3.00	2.25	1.75
interfacial energy [J/m²]	3.83 – 6.72 10 <sup>-07</sup>	4.27 – 7.48 10 <sup>-07</sup>	4.98 - 8.74 10-07