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Supplementary Information for: Orientation dependence of optical activity in light scattering by nanoparticle clusters

Atefeh Fazel-Najafabadi, *a,b* and Baptiste Auguié**a,b*

 ^a School of Chemical and Physical Sciences, Victoria University of Wellington, PO Box 600, Wellington 6140, New Zealand
 ^b The MacDiarmid Institute for Advanced Materials and Nanotechnology *E-mail: baptiste.auguie@vuw.ac.nz

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1 Dimer of Au nanorods

Figure S1 presents the analytical orientation averaged results (solid lines) and the numerical averaging over x, y, z incident directions (dashed lines) for absorption, scattering, and extinction circular dichroism spectra in the "fingers crossed" dimer configuration, with different scaling factor (f). The structure comprises two identical Au spheroids with semi-axes f.a, f.b, with nominal dimensions a = 15 nm, c = 40 nm, and centre-to-centre separation f.d, with d = 100 nm. For small scaling factors ($f \ll 1$), the dimer is much smaller than the wavelength and averaging over just the 3 orthogonal x, y, z directions provides an accurate orientation averaging. However, the accuracy suffers as the scaling factor increases beyond $f \sim 0.8$. We also note that for small dimers, extinction is largely dominated by absorption and the circular dichroism spectrum has an antisymmetrical line shape, very reminiscent of exciton coupling in molecular CD. For larger scaling factors, scattering contributes more to the overall extinction CD with an increase in the intensity of the CD spectrum and a deviation from the antisymmetric curve predicted by exciton coupling theory. The intermediate regime where absorption is on par with scattering leads to interesting spectral profiles, such as a transition in the scattering lineshape from a monosignate band below f = 1.2, evolving to a bisignate band resembling more that of absorption at larger sizes¹.



Fig. S1: Absorption (red), scattering (green), and extinction (blue) circular dichroism spectra for the "fingers crossed" dimer of Fig. 2, with different scaling factor (f). The solid lines show the analytical orientation averaged results and the dashed lines are for the average over x, y, z. Each particle in the dimer is represented as a prolate ellipsoid with semi-axes f.a and f.c (a = 15 nm and c = 40 nm) and center-to-center separation f.d (d = 100 nm).



Fig. S2: Angular maps of the extinction CD at different wavelengths for the "fingers crossed" dimer consisting of two Au prolate spheroids with semi-axes a = 30 nm and b = 80 nm, interparticle separation d = 200 nm (scaling factor f = 2 in Fig. S1, corresponding to Fig. 2 of the main text), and relative orientation $\theta = \pi/4$.

Figure S2 presents multiple maps of the angular pattern of extinction circular dichroism for the dimer (f = 2) presented in the main text, Fig. 2. The different panels are for wavelengths spanning the spectral range 520–800 nm, showing the evolution between the two extremes shown in the main text. Note that the colour scale is normalised to unity in each panel, to facilite comparisons of the patterns, as the absolute intensities vary strongly across the spectrum.

2 GoldHelix



Fig. S3: Angular response of a GoldHelix cluster² with N = 40 particles (4 × 10 ribbon). Upper panels: Extinction and scattering spectra for three perpendicular incident directions, *x*-axis (solid red line), *y*-axis (solid blue line), *z*-axis (solid green line), the numerical average over the three (N.A., dashed black line), and the analytical orientation-averaged result (A.A., solid black line). Lower panels: the corresponding results for circular dichroism. Note that the numerical and analytical orientation averages are mostly indistinguishable, except for panel (d).

Figure S3 complements Fig. 4 (a–d) of the main text, with far-field spectra for the truncated GoldHelix instead of the full structure. The 3D view on the right illustrates the geometry in more detail: a 4×10 ribbon is formed by a square array of Au nanospheres, wrapped in a helix around a central template². The template is not part of the electromagnetic simulations, however its effect on optical properties is expected to be relatively weak, as it is made of silica.



Fig. S4: Spherical harmonic analysis of the circular dichroism angular patterns shown in Fig. 4 (e,f). We use the function sphharm from the Matlab library chebfun^{3,4} to compute the decomposition of the angular map into spherical harmonics. For each harmonic degree *l* we collapse the corresponding 2l + 1 orders as $|a_l| = \sum_{m=-l}^{l} |a_{lm}|^2$; the result summarises the total weight of degree *l* in the pattern (higher degrees correspond to higher angular frequencies). For ease of comparison the scale is logarithmic, and we normalise the results such as $\max a_l = 1$. The two cases considered correspond to the full GoldHelix cluster (blue triangles, 320 particles), and the truncated structure (red circles, 80 particles).

3 Si chain

Figure S5 shows the extinction cross section of a single Si nanosphere with radius r = 150 nm (black solid line). The spectra ED (red dashed line) and MD (blue dashed line) refer to electric dipole and magnetic dipole contributions to the extinction cross section, separately. It can be seen that spectra of ED and MD intersect at $\lambda = 1208$ nm corresponding to the so called first Kerker condition.

Fig. S6 shows the normalised extinction efficiency $Q_{\text{ext}} = C_{\text{ext}}/(N.C_{\text{ext},1})$ for a chain of N = 50 Si nanospheres, where $C_{\text{ext},1}$ is the extinction cross section of a single silicon particle at λ_{Kerker} . The structure is illuminated normal to the chain axis with a circularly polarised plane wave at $\lambda = 1208$ nm, and we vary the interparticle distance (pitch).



Fig. S5: Extinction spectrum of a single Si nanosphere with radius r = 150 nm in air. The total spectrum (solid black line) is decomposed into electric dipole (ED, dashed red line) and magnetic dipole (MD, dashed blue line). A vertical dotted line at $\lambda = 1208$ nm indicates the Kerker condition where electric dipole and magnetic dipole terms are equal.



Fig. S6: Extinction efficiency at $\lambda = \lambda_{\text{Kerker}} = 1208 \text{ nm}$ for a chain of N = 50 silicon nanospheres as a function of pitch/ λ_{Kerker} . The chain is illuminated with circular polarisation at normal incidence.

4 GoldHelix cluster description

The following listing contains the cartesian positions of the 320 Au nanospheres used in the GoldHelix model of Fig. 4 of the main text.

material x y z radius		
Au 1.7320508080e+01 1.000000000e+01 -2.139583333 Au 1.000000000e+01 1.732050808e+01 -2.085416667	e+025 e+025	
Au 1.224646799e-15 2.00000000e+01 -2.031250000	e+02 5	
Au -1.00000000e+01 1.732050808e+01 -1.977083333	e+02 5	
Au -1.732050808e+01 1.000000000e+01 -1.922916667	e+025	
Au -1.732050808e+01 -1.00000000e+01 -1.814583333	e+02 5 e+02 5	
Au -1.000000000e+01 -1.732050808e+01 -1.760416667	e+02 5	
Au -3.673940397e-15 -2.000000000e+01 -1.706250000	e+02 5	
Au 1.000000000e+01 -1.732050808e+01 -1.652083333	e+025	
Au 2.000000000e+01 -1.0000000000e+01 -1.597910007	e+02 5 e+02 5	
Au 1.732050808e+01 1.00000000e+01 -1.489583333	e+02 5	
Au 1.000000000e+01 1.732050808e+01 -1.435416667	e+02 5	
Au 2.388680239e-14 2.000000000e+01 -1.381250000	e+02 5	
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Au	1.109006785e+01 -1.664362927e+01 -7.274550949e+00	5
Au Au	1./920095120+01 -8.868//18330+00 -1.8578842830+00 1.995883968e+01 1.282/65851e+00 - 5.5070320/a-00 5	ъ
Au	1.664362927e+01 1.109006785e+01 8.975449051e+00 5	
Au	8.868771833e+00 1.792609512e+01 1.439211572e+01 5	
Au	-1.282465851e+00 1.995883968e+01 1.980878238e+01	5
Au 4	-1.109006/85e+01 1.664362927e+01 2.522544905e+01	5
Au	-1.995883968e+01 -1.282465851e+00 3.605878238e+01	5
Au	-1.664362927e+01 -1.109006785e+01 4.147544905e+01	5
Au	-8.868771833e+00 -1.792609512e+01 4.689211572e+01	5
Au	1.282465851e+00 -1.995883968e+01 5.230878238e+01	5
AU A11	1.105000512e+01 -1.664362927e+01 5.772544905e+01 1.792609512e+01 -8 868771833e+00 6 3172115720+01	5 5
Au	1.995883968e+01 1.282465851e+00 6.855878238e+01 5	2
Au	1.664362927e+01 1.109006785e+01 7.397544905e+01 5	
Au	8.868771833e+00 1.792609512e+01 7.939211572e+01 5	
Au A	-1.282465851e+00 1.995883968e+01 8.480878238e+01	5
ли Au	-1.792609512e+01 8.868771833e+00 9.564211572e+01	5
Au	-1.995883968e+01 -1.282465851e+00 1.010587824e+02	5
Au	-1.664362927e+01 -1.109006785e+01 1.064754491e+02	5
Au	-8.868771833e+00 -1.792609512e+01 1.118921157e+02	5
Au Au	1.202405851e+00 -1.995883968e+01 1.173087824e+02 1.109006785e+01 -1.664362927e+01 1.2272544010+02	5 5
Au	1.792609512e+01 -8.868771833e+00 1.281421157e+02	5
Au	1.995883968e+01 1.282465851e+00 1.335587824e+02 5	
Au	1.664362927e+01 1.109006785e+01 1.389754491e+02 5	
AU Au	-1.282465851e+00 1 995883968e+01 1 4080878240.00	5
Au	-1.109006785e+01 1.664362927e+01 1.552254491e+02	5
Au	-1.792609512e+01 8.868771833e+00 1.606421157e+02	5
Au	-1.995883968e+01 -1.282465851e+00 1.660587824e+02	5
Au	-1.664362927e+01 -1.109006785e+01 1.714754491e+02	5
AU A11	-0.000//18330+00 -1./926095120+01 1.7689211570+02 1.2824658510+00 -1.9958839680+01 1.8230878240+02	5 5
Au	1.109006785e+01 -1.664362927e+01 1.877254491e+02	5
A	1 7026005120+01 -8 8687718330+00 1 0314211570+02	5

Au	1.995883968e+01 1.282465851e+00 1.985587824e+02 5	
Au	1.664362927e+01 1.109006785e+01 2.039754491e+02 5	
Au	8.868771833e+00 1.792609512e+01 2.093921157e+02 5	_
Au	-1.282465851e+00 1.995883968e+01 2.148087824e+02	5
Au	-1.109006785e+01 1.664362927e+01 2.202254491e+02	5
Au	-1./92609512e+01 8.868//1833e+00 2.25642115/e+02	5
Au	-1.995883968e+01 -1.282465851e+00 2.310587824e+02 1.6642620270+01 1.1000067850+01 2.2647544010+02	5
Au	1 072656258e+01 -3 205865383e+00 -1 801826508e+02	5
Au	1.873163702e+01 7.008078143e+00 -1.747650931e+02	5
Au	1 271758444e+01 1 543577164e+01 -1 693493264e+02	5
Au	3 295865383e+00 1 972656258e+01 -1 639326598e+02	5
Au	-7.008978143e+00 1.873163702e+01 -1.585159931e+02	5
Au	-1.543577164e+01 1.271758444e+01 -1.530993264e+02	5
Au	-1.972656258e+01 3.295865383e+00 -1.476826598e+02	5
Au	-1.873163702e+01 -7.008978143e+00 -1.422659931e+02	5
Au	-1.271758444e+01 -1.543577164e+01 -1.368493264e+02	5
Au	-3.295865383e+00 -1.972656258e+01 -1.314326598e+02	5
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Au	1.873163702e+01 7.008978143e+00 -1.097659931e+02	5
Au	1.271758444e+01 1.543577164e+01 -1.043493264e+02	5
Au	3.295865383e+00 1.972656258e+01 -9.893265976e+01	5
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Au	-1.873163702e+01 -7 008078143e+00 -7 72650930e+01	5
Au	-1 271758444e+01 -1 543577164e+01 -7 184932642e+01	5
Au	-3.295865383e+00 -1.972656258e+01 -6.643265976e+01	5
Au	7.008978143e+00 -1.873163702e+01 -6.101599309e+01	5
Au	1.543577164e+01 -1.271758444e+01 -5.559932642e+01	5
Au	1.972656258e+01 -3.295865383e+00 -5.018265976e+01	5
Au	1.873163702e+01 7.008978143e+00 -4.476599309e+01	5
Au	1.271758444e+01 1.543577164e+01 -3.934932642e+01	5
Au	3.295865383e+00 1.972656258e+01 -3.393265976e+01	5
Au	-7.008978143e+00 1.873163702e+01 -2.851599309e+01	5
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Au	-1.972656258e+01 3.295865383e+00 4.731734024e+01	5
Au	-1.873163702e+01 -7.008978143e+00 5.273400691e+01	5
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Au	7.008978143e+00 -1.873163702e+01 6.898400691e+01	5
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Au	1.9/20502580+01 -3.2958055830+00 /.981/340240+01	5
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Au	-7.008978143e+00 1.873163702e+01 1.014840069e+02	5
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Au	-1.972656258e+01 3.295865383e+00 1.123173402e+02	5
Au	-1.873163702e+01 -7.008978143e+00 1.177340069e+02	5
Au	-1.271758444e+01 -1.543577164e+01 1.231506736e+02	5
Au	-3.295865383e+00 -1.972656258e+01 1.285673402e+02	5
Au	7.008978143e+00 -1.873163702e+01 1.339840069e+02	5
Au	1.543577164e+01 -1.271758444e+01 1.394006736e+02	5
Au	1.972656258e+01 -3.295865383e+00 1.448173402e+02	5
Au	1.8/3163/02e+01 /.0089/8143e+00 1.502340069e+02 5	
Au	1.2/1/58444e+01 1.5435//164e+01 1.556506/36e+02 5	
AU	-7 0080781/3010 1 8731627020101 1 664040000000	5
ли А.,	-1 543577164e+01 1 271758444e+01 1 7100067760+02	5
An	-1.972656258e+01 3.295865383e+00 1 773173402e±02	5
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Au	1.543577164e+01 -1.271758444e+01 2.044006736e+02	5
Au	1.972656258e+01 -3.295865383e+00 2.098173402e+02	5
Au	1.873163702e+01 7.008978143e+00 2.152340069e+02 5	
Au	1.271758444e+01 1.543577164e+01 2.206506736e+02 5	
Au	3.295865383e+00 1.972656258e+01 2.260673402e+02 5	_
Au	B 0000B0110 00 1 0B0107555 55 55 55 55 55	-
	-7.008978143e+00 1.873163702e+01 2.314840069e+02	5
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Au Au	-7.008978143e+00 1.873163702e+01 2.314840069e+02 -1.543577164e+01 1.271758444e+01 2.369006736e+02 -1.972656258e+01 3.295865383e+00 2.423173402e+02 1.872162702.01 2.0802781402+02 2.423173402e+02	5 5 5

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