Scattering Spectroscopy of Plasmonic Janus Particles

Supplementary Material

Felix H. Patzschke, A. Markus Anton and *Frank Cichos

Orientation Definition

The orientation of the system is characterised by the angles between three unit vectors;

$\hat{k}$, the propagation direction of the incident light, with respect to which scattering angles are defined,

$\hat{o}$, the "forward" direction along the optical axis, equivalent to the central axis of the objective's collection cone and

$\hat{z}$, the symmetry axis of the particle, oriented such that the Au cap lies in the positive and the PS side in the negative z-direction.

In terms of these, the out-of-plane orientation of the JP is defined as $\alpha := \measuredangle(\hat{o}, \hat{z})$. Similarly, we define the illumination angle $\zeta := \measuredangle(\hat{k}, \hat{z})$, which, in this model, is the only orientation parameter needed to define the light-matter interaction of the JP.

Scattering angles are defined for each plane-wave contribution to the scattered field: Let that contribution have a propagation direction $\hat{k}'$. Then, $\theta := \measuredangle(\hat{k}', \hat{k})$ is the polar component of the scattering angle. Additionally, there is an azimuthal component ϕ, the choice of reference point for which being somewhat arbitrary. In the following, it will be chosen such that if $\hat{k}'$ lies in the $(\hat{k}, \hat{z})$ plane, then $\phi = 0$.

These definitions are illustrated in [Figure] .

Derivation of the probability density for the out-of-plane angle

The model JP is cylindrically symmetric. Therefore its orientation in space is completely described by the direction of its axis of symmetry, $\hat{z}$. For the random orientation, let $\hat{z}$ be evenly distributed over $\mathcal{S}^2$. For a PDF p_Ω w.r.t. the solid angle Ω, this means

$$\iint_{\mathcal{T}} p_\Omega \, \mathrm{d}\Omega = \frac{\iint_{\mathcal{T}} \mathrm{d}\Omega}{\oiint_{\mathcal{S}^2} \mathrm{d}\Omega} \quad \forall\, \mathcal{T} \subseteq \mathcal{S}^2 \, .$$

$\oiint_{\mathcal{S}^2} \mathrm{d}\Omega$ is nothing else than the surface area of the unit sphere, 4π. Therefore,

$$\iint_{\mathcal{T}} p_\Omega \, \mathrm{d}\Omega = \frac{1}{4\pi} \iint_{\mathcal{T}} \mathrm{d}\Omega \quad \forall\, \mathcal{T} \subseteq \mathcal{S}^2 \, ,$$

which implies that

$$p_\Omega = \frac{1}{4\pi} \, .$$

Now, we parametrise $\mathcal{S}^2$ in spherical coordinates (α, β), where α is the polar coordinate and β is the azimuthal coordinate. The differential solid angle is

$$\mathrm{d}\Omega = \sin\alpha \, \mathrm{d}\alpha \, \mathrm{d}\beta \, .$$

The PDFs w.r.t. these coordinates, p_α and p_β, must satisfy

$$\begin{aligned}
\text{a)} \qquad & \int_0^\pi p_\alpha \,\mathrm{d}\alpha = 1 \\
\text{b)} \qquad & \int_0^{2\pi} p_\beta \,\mathrm{d}\beta = 1 \\
\text{and c)} \quad & \iint_\mathcal{T} p_\alpha \cdot p_\beta \,\mathrm{d}\alpha\,\mathrm{d}\beta = \iint_\mathcal{T} p_\Omega \,\mathrm{d}\Omega \quad \forall\, \mathcal{T} \subseteq \mathcal{S}^2 \,.
\end{aligned}$$

From c), it follows that

$$\begin{aligned}
p_\alpha \cdot p_\beta \,\mathrm{d}\alpha\,\mathrm{d}\beta &= p_\Omega \,\mathrm{d}\Omega \\
&= \frac{1}{4\pi} \sin\alpha \,\mathrm{d}\alpha\,\mathrm{d}\beta \,.
\end{aligned}$$

Cancelling $\mathrm{d}\alpha\,\mathrm{d}\beta$ yields

$$p_\alpha \cdot p_\beta = \frac{1}{4\pi} \sin\alpha \,.$$

To separate p_α and p_β, let's assume that p_α and p_β are invariant w.r.t. β and α, respectively. Then, substitution in a) yields

$$\begin{aligned}
\frac{1}{p_\beta} \int_0^\pi p_\alpha \cdot p_\beta \,\mathrm{d}\alpha &= 1 \\
\frac{1}{4\pi\, p_\beta} \underbrace{\int_0^\pi \sin\alpha \,\mathrm{d}\alpha}_{=2} &= 1 \quad \therefore \quad p_\beta = \frac{1}{2\pi} \,,
\end{aligned}$$

which also satisfies b). It follows that

$$p_\alpha = \frac{\sin\alpha}{2} \,.$$

Integration of the Far-Field Intensities

For a given incident wavelength and illumination angle, let the scattering intensity in the far-field be

$$I_\text{far}(\theta, \phi) \,.$$

The data retrieved from the simulation are I_n, samples of I_far in points (θ_n, ϕ_n) on the unit sphere $\mathcal{S}^2$. To compute

$$\iint_\mathcal{T} I_\text{far} \,\mathrm{d}\Omega \approx \sum_{n | (\theta_n, \phi_n) \in \mathcal{T}} I_n \,\Delta\Omega_n \,,$$

we must determine the values of $\Delta\Omega_n$. [TODO: Notation]

We project the spherical coordinates (θ_n, ϕ_n) into the Euclidean plane, obtaining the cartesian coordinates (x_n, y_n).[1] We then compute the Voronoi tesselation which maps each projected sample point (x_n, y_n) to an area element[2]

$$P_n = \left\{ (x, y) \in \mathbb{R}^2 \middle| \arg\min_{n'} \Big(m_\text{E}\big((x, y) - (x_{n'}, y_{n'})\big) \Big) = n \right\}$$

with the Euclidean metric m_E. The area elements $\Delta\Omega_n$ are then given as

$$\Delta\Omega_n = \mu\left(P_n \cap \mathcal{T}\right) \cdot \tau\left(\theta_n, \phi_n\right)$$

where $\mu(\cdot)$ denotes the measure of a subset of $\mathbb{R}^2$ and τ is the area element transfer function associated with the chosen projection.

More Angular Distributions

[1] The choice of projection is somewhat arbitrary, we chose the stereographic projection.

[2] the fact of which being a convex polygon being computationally convenient, yet analytically irrelevant

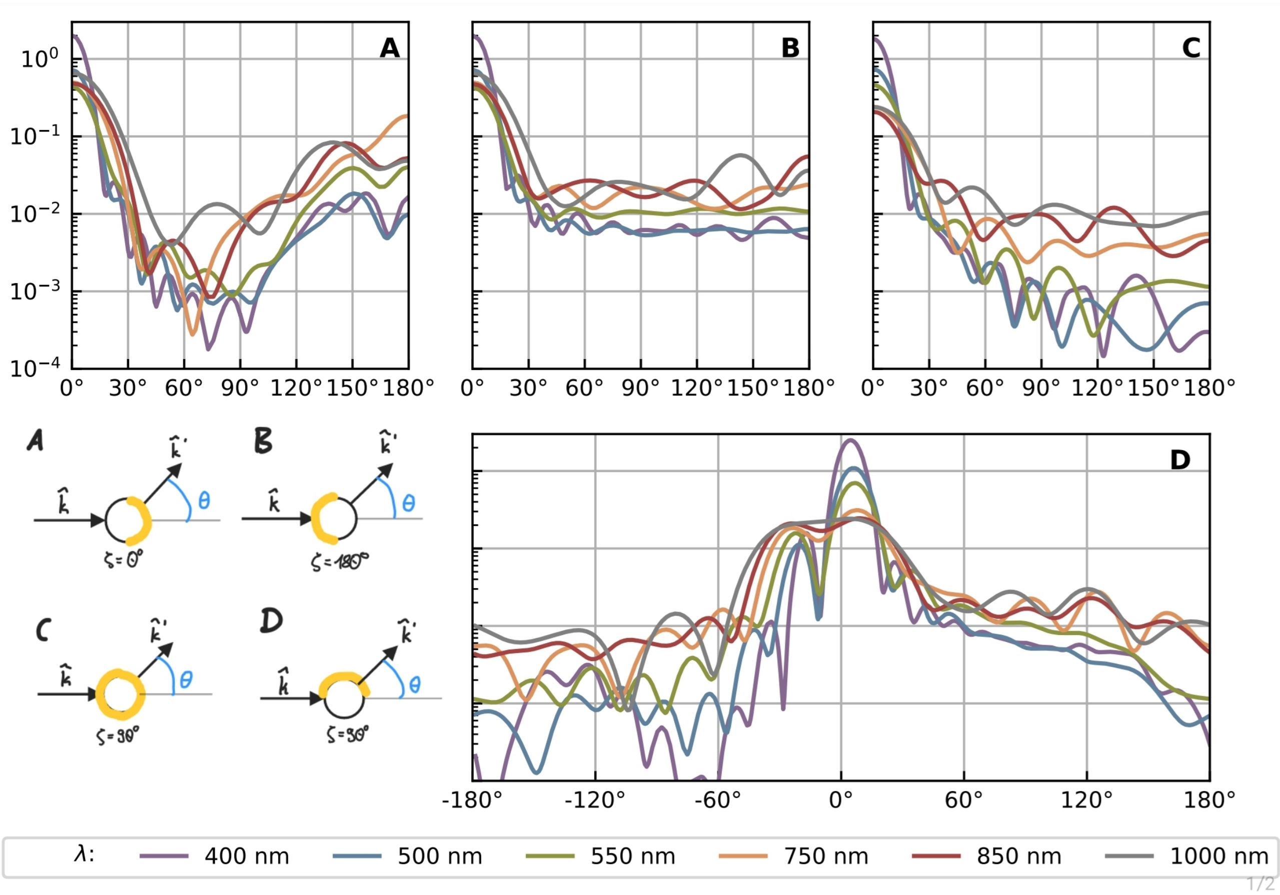

Figure 1: Scattering intensity of the JP versus scattering angle. **A** and **B** show the cylindrically symmetric cases of illumination from the PS side and from the Au side, respectively, i.e. where $\hat{k} \parallel \hat{z}$. In **C** and **D**, the light is incident side-on ($\hat{k} \perp \hat{z}$, $\zeta = \pi/2$). The scattering intensities are taken from the $(\hat{k}, \hat{y})$ plane in **C** (note, that the system is still symmetric under inversion of y) and from the $(\hat{k}, \hat{z})$ plane, where spatial symmetry is entirely broken, in **D**. Here, the Au side lies in the positive θ direction.