Light absorption in two-dimensional crystals covered by randomly distributed plasmonic nanoparticles

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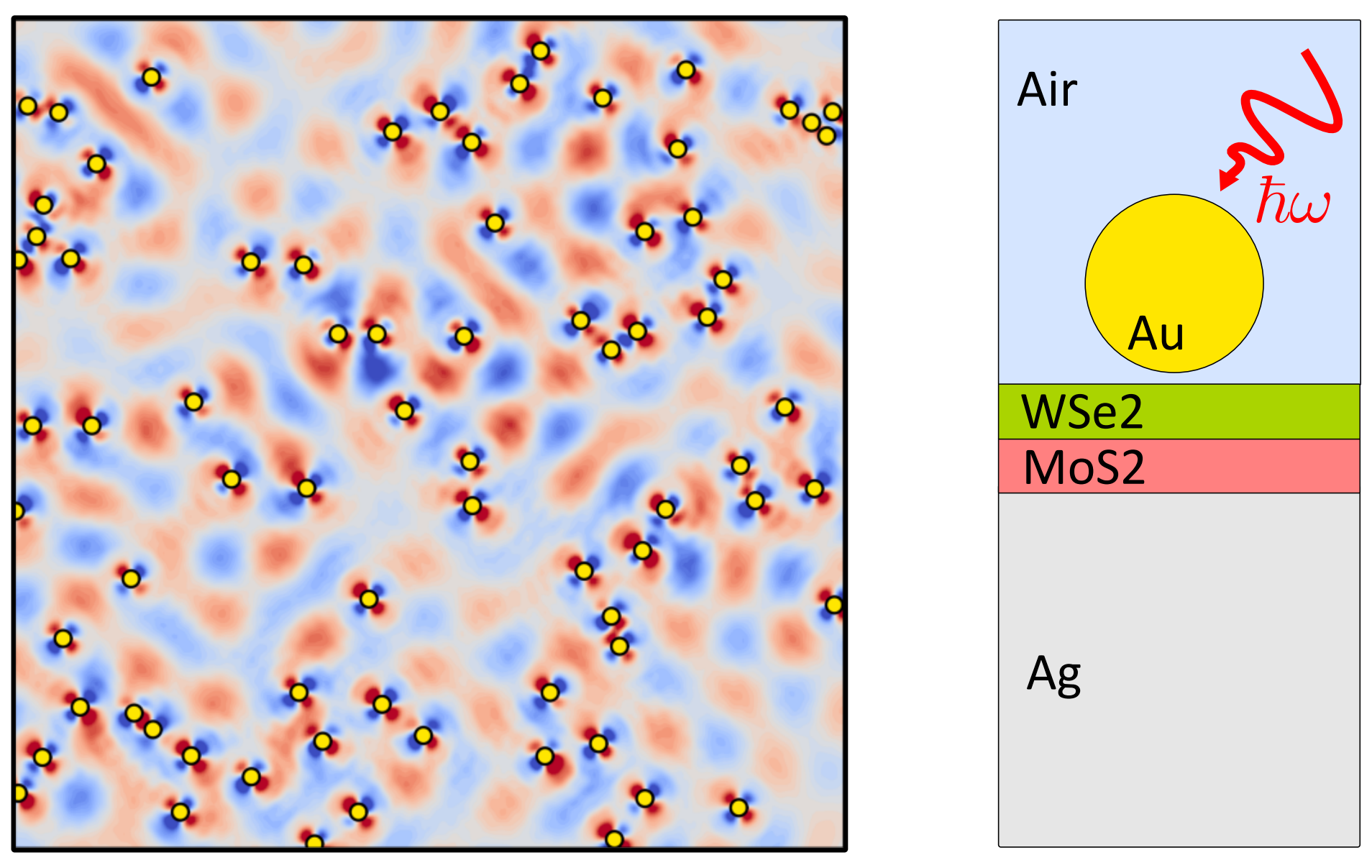
Abstract

The discovery of many thermodynamically stable two-dimensional crystals attainable, amonst others, by the mechanical exfoliation method [1] has, since the 2000s, been inspiring the investigation of a range of electronic and, more recently, optoelectronic systems featuring such atomically thin layers either individually or stacked into so-called van der Waals heterostructures. In case of van der Waals heterostructure photovoltaics [2], light trapping strategies gain in significance as the active layer thickness is typically two orders of magnitude smaller than the wavelength.
We pursue ways to increase the light absorption efficiency using plasmonic nanoparticles randomly distributed on top of two-dimensional crystals [3]. To study the light absorption in disordered systems, we employ SMUTHI [4,5], an open-souce T-matrix based Python package for simulating light scattering on particles embedded in arbitrary layered systems. In numerical simulations, we assume that a number N_p of nanoparticles is arranged over a square area A with a uniform distribution. As macroscopic clusters (such that $A \sim 1\mathrm{mm}^2$ or more) are way too large to simulate numerically, our method relies on simulating sufficiently large (typically $N \sim 100$) ensembles of smaller randomly generated clusters and estimating the absorption enhancement of a macroscopic system from the ensemble average.

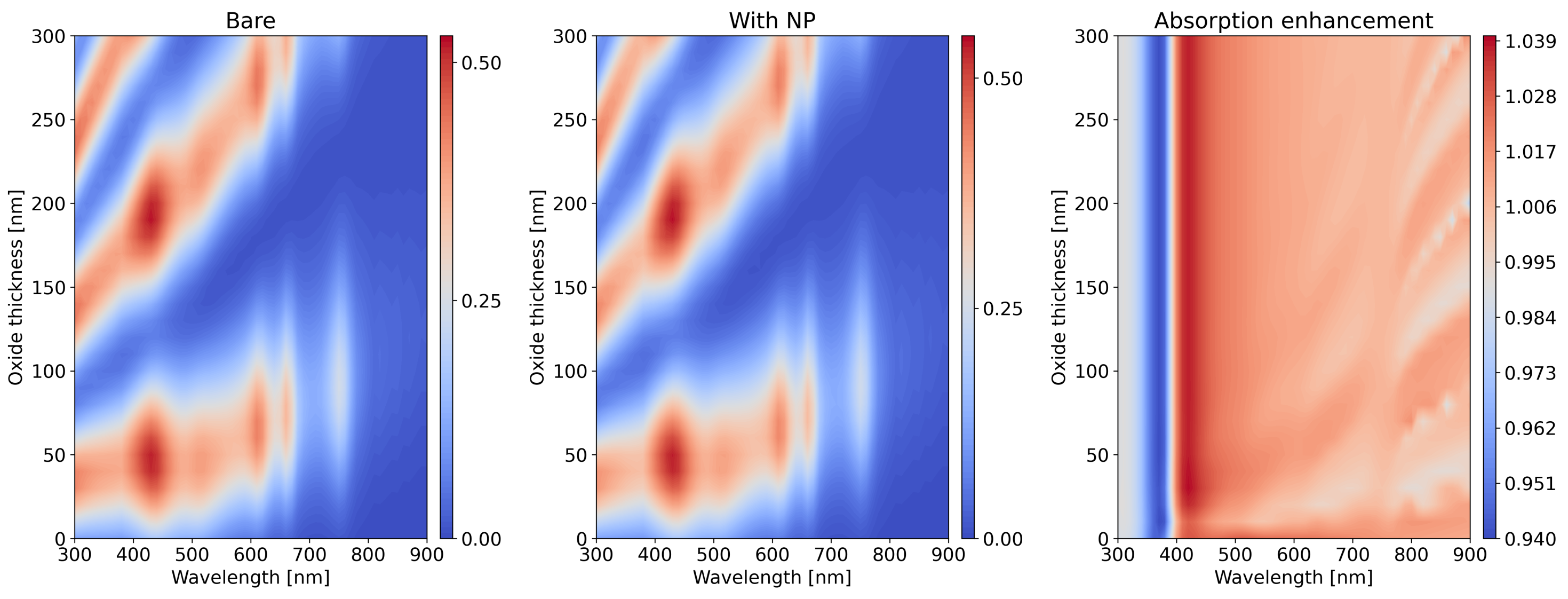
Modeling light scattering with SMUTHI



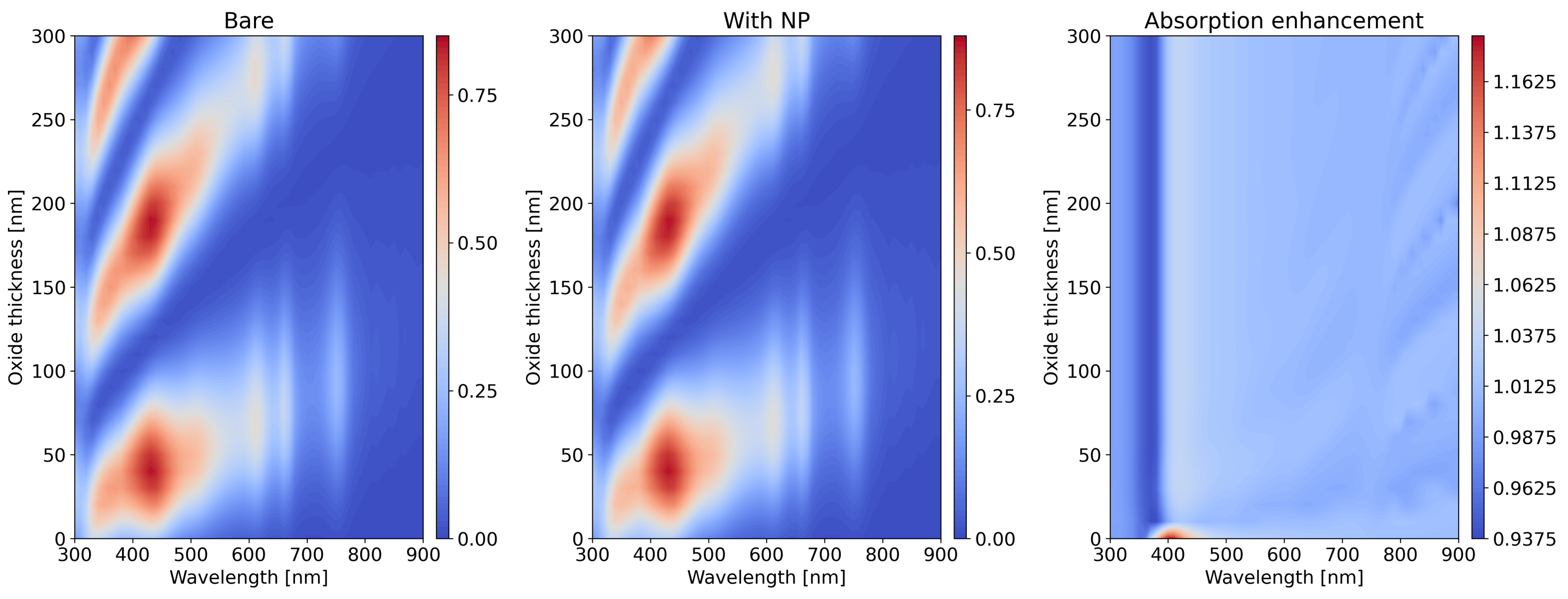
Left panel: a typical electric field distribution evaluated using SMUTHI. Here 75 gold nanoparticles with a 100 nm diameter are randomly distributed over a $5 \times 5 \mu m^2$ area on top of a $\mathrm{WSe}_2/\mathrm{MoS}_2$ heterostructure placed on a silver film, as indicated in the schematic (right panel). The incoming beam has a wavelength of 550 nm, a perpendicular incidence and is y-polarized, while the map shows the real part of the electric field phasor x-component, normalized to the incoming field magnitude.

Layer configuration

The effect plasmonic nanoparticles have on the absorption in the nanometer thin active PV layer is strongly dependent on the layer configuration underneath. To illustrate this, here we consider the case where the substrate is made of gold or silver. The active layer, here assumed as a $\mathrm{WSe}_2/\mathrm{MoS}_2$ van der Waals heterostructure comprising a monolayer of WSe_2 (thickness of 0.65 nm) on top of a MoS_2 monolayer (thickness of 0.62 nm), is assumed to be separated by a SiO_2 layer whose thickness is allowed to vary from 0 nm to 300 nm, while the area considered is $1 \times 1 \mu m^2$ around a single nanoparticle.



Gold substrate. Absorption without any particles ('Bare'), with one 60 nm silver nanoparticle ('With NP') and the ratio of the two ('Absorption enhancement').



Silver substrate. Absorption without any particles ('Bare'), with one 60 nm silver nanoparticle ('With NP') and the ratio of the two ('Absorption enhancement').

Absorption enhancement by random nanoparticle clusters

The effect of the nanoparticles is quantified by the local absorption enhancement factor η, defined as the ratio of the local light absorption rate density with (w_{np}) and without (w_0) the plasmonic nanoparticles $\eta = \frac{w_{np}}{w_0}$, whereby the light absorption rate density is evaluated by invoking the Poynting's theorem in linear dispersive media with losses

$$w = \omega \mathrm{Im}\{\varepsilon(\omega)\} \left(|E_x|^2 + |E_y|^2\right)$$

The absorption enhancement rate per area A is calculated for the entire cluster as the integral of the local absorption enhncement rate normalized to A

$$\eta_A = \frac{1}{A}\int_A \eta(x,y)dxdy.$$

To assess η_A^{macro}, the absorption enhancement observed in a macroscopic sample, one would, in principle, need to solve the field scattering problem involving millions of nanoparticles distributed over a very large area

$$\eta_A^{\mathrm{macro}} = \eta_{A\to\infty},$$

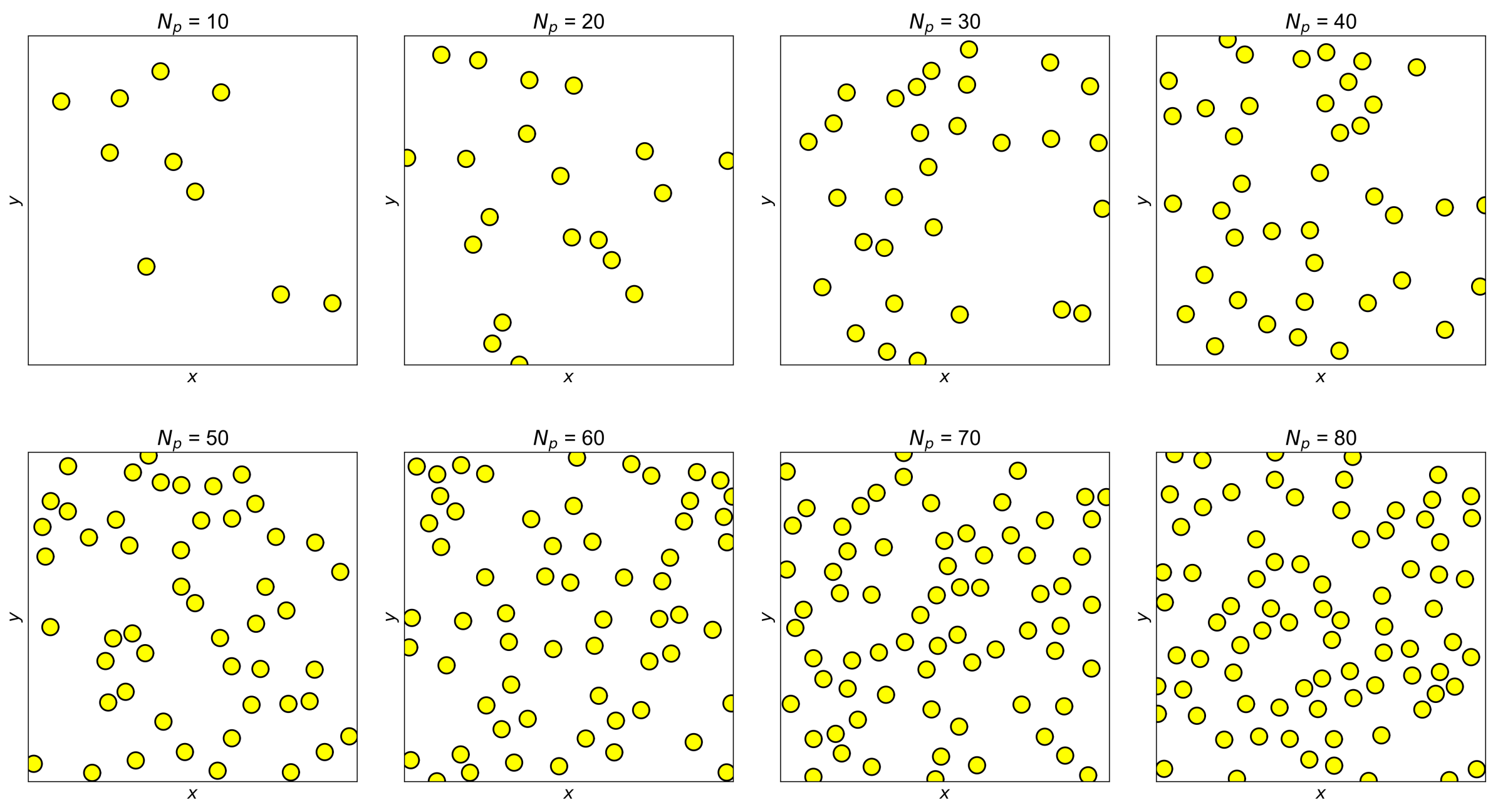
which is not feasible. Instead, we assess η_A^{macro} by analyzing the values of η_A calculated over an ensemble of clusters with smaller number of particles. The ensemble average is calculated as

$$\overline{\eta}_A = \frac{1}{N}\sum_{i=1}^{N} \eta_A^i,$$

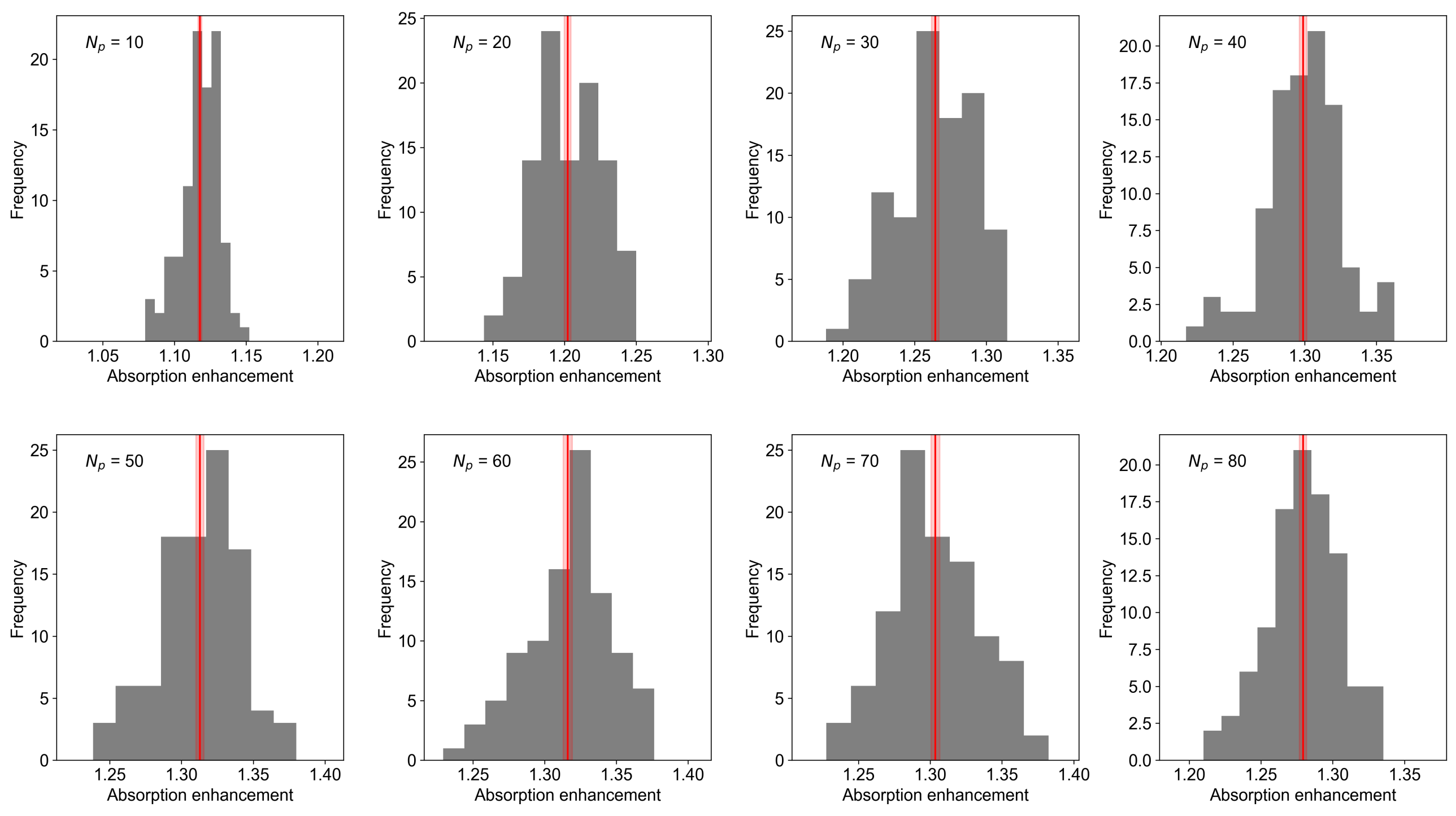
where η_A^i denotes the enhancement observed in the i-th cluster of the ensemble. If evaluated properly, $\overline{\eta}_A$ should represent an accurate estimate for η_A^{macro}, where the error bound is given by the standard error of the mean

$$\Delta\overline{\eta}_A = \frac{1}{\sqrt{N}}\sigma_A, \quad \sigma_A = \frac{1}{\sqrt{N-1}}\sqrt{\sum_{i=1}^{N}(\eta_A^i - \overline{\eta}_a)^2}.$$

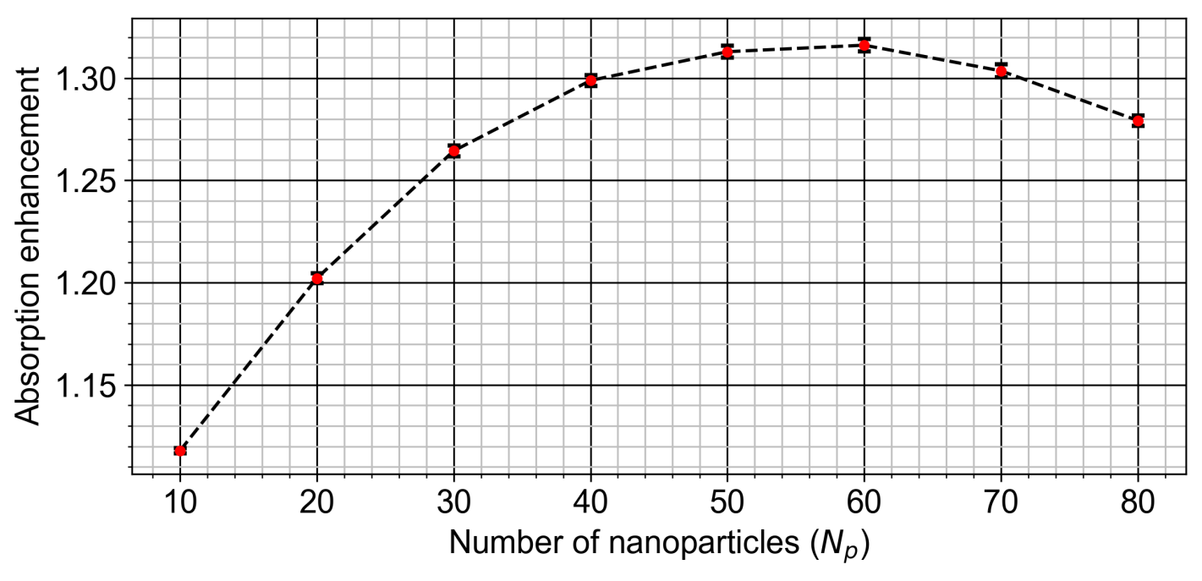
Optimal particle density



Here we demonstrate the use of numerical simulations for finding the optimal particle density. Monodisperse gold nanoparticles of 100 nm diameter are randomly distributed over a 1 nm thick layer of MoS_2 on top of an opaque gold film. The figure on left shows the representative nanoparticle clusters with varying N_p distributed over $A = 4\mu\mathrm{m}^2$.



Absorption enhancement histograms calculated for ensembles containing $N = 100$ clusters with $A = 4\mu\mathrm{m}^2$ with particle number increasing from $N_p = 10$ (top left) up to $N_p = 80$ (bottom right).



Sample averages of the apsorption enhancement drawn as a function of N_p while keeping the cluster area fixed at $A = 4\mu\mathrm{m}^2$, showing that a maximum is reached around $N_p = 60$.

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