

Farfield method for detection of geometrical patterns of self-assembled gold nanospheres

Sandra Krämer*, Kathrin Kroth**, Thomas Sure*, Peter J. Klar**

*Institut für Optik und Mikrosysteme, Technische Hochschule Mittelhessen

**1. Physikalisches Institut, Justus-Liebig-Universität Gießen

mailto:sandra.kraemer-2@me.thm.de

Gold nanospheres are measured with the method of Coherent Fourier Scatterometry. The observed scattering behavior of a single particle matches our calculations. Furthermore, different geometrical patterns of gold nanospheres are distinguished by the intensity distribution in the Fourier plane.

1 Introduction

We present the simulation and detection of single spherical nanoparticles with the method of Coherent Fourier Scatterometry (CFS), an easy-to-implement far-field optical method. We directly image the Fourier plane of a high numerical aperture microscope objective onto a CCD to evaluate disturbances of the wavefront that are caused by particles smaller than the diffraction limit. For the simulation of this scattering problem, a modification of Sommerfeld's half-space problem is used. In addition, we distinguish different geometric assemblies of nanospheres based on the intensity distribution of the exit pupil and present a method to determine the angle of the link axis of two adjacent particles.

2 Methods

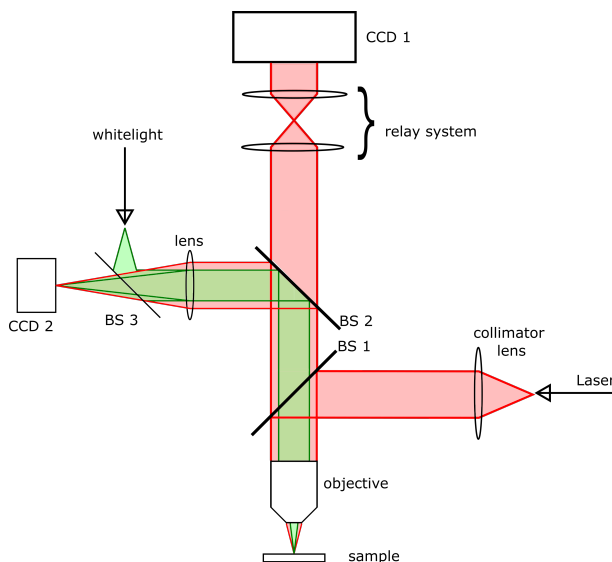


Fig. 1 Sketch of the experimental setup used for CFS.

Figure 1 shows a sketch of the experimental setup. The CFS setup (red beam path) is extended with another optical path that operates as a white light microscope. For CFS measurements, the same objective that focuses the incoming light also collects

the scattered light. The numerical aperture is $NA = 0.9$ and we use a wavelength of 633 nm . The exit pupil is imaged onto a CCD using a relay system.

The examined samples are spherical gold nanoshells showing a diameter of 300 nm . Besides single particles, we produced geometric assemblies in the form of L's and lines.

For the simulation of the scattering signal of a single particle, a modification of SOMMERFELD'S half-space-problem is used. This problem describes an oscillating dipole with defined orientation positioned at a certain height above a dielectric surface [1]. We assume that the incoming light induces a dipole moment in the nanoparticle. The calculations relate to [2] and use certain simplifications for the far-field. The solution we use is also known as the SOMMERFELD-RAYLEIGH-System, presented in [3]. According to [4] the SOMMERFELD far-field vector potential $\vec{\Pi}$ can be written as

$$\vec{\Pi} = \Pi_x \vec{e}_x + \Pi_z \vec{e}_z \quad (1)$$

with

$$\vec{\Pi} = \nabla \times (c\Psi) \quad (2)$$

where Ψ is a generating function that is determined by the symmetry of the problem. To obtain the electric far-field components we solve the wave equation

$$\vec{E} = k^2 \cdot \vec{\Pi} + \nabla(\nabla \cdot \vec{\Pi}) \quad (3)$$

With this solution we can predict the resulting intensity distribution of the Fourier plane.

3 Results

Figure 2 shows the intensity distribution of the exit pupil which was calculated according to Equation 3 and the measured, background corrected intensity distribution for a single particle using linear polarized light. As can be seen in 2 the qualitative behavior of the intensity distribution is well described by our theoretical approach.

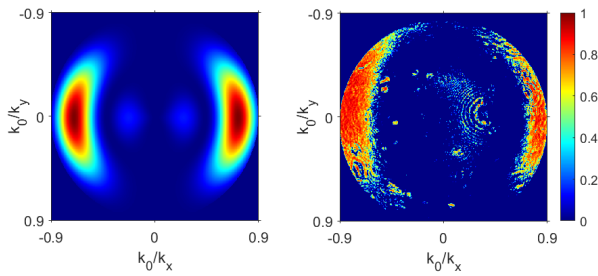


Fig. 2 Left: Simulation, Right: measured intensity distribution of the exit pupil for single particle using linear polarized light.

For the measurement of the geometric assemblies, circular polarized light was used to avoid a stimulation of the structures in a preferential direction.

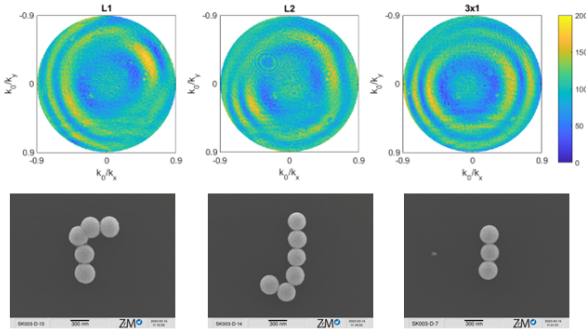


Fig. 3 SEM images of the structures measured and their corresponding CFS signals.

Figure 3 shows SEM images of three geometrical structures and their corresponding CFS signals. The symmetry of the assemblies determines the intensity distribution. The L1-structure shows a ring shaped intensity distribution in the entire left half of the pupil. In the right half, the upper area in particular stands out with a region of high contrast. This intensity distribution can be traced back to the orientation of L1. The pupil image for L2 is to be interpreted analogously. The pupil images of the three neighboring particles, 3x1, show intensity accumulations and a higher modulation perpendicular to the vertical line direction. The intensity distribution in the upper and lower area of the pupil, i.e. in the direction of the line, is of lower contrast.

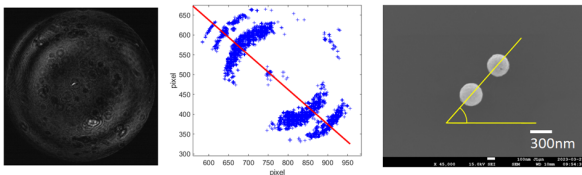


Fig. 4 Illustration of finding the angle of link axis of neighboring particles.

A qualitatively equivalent behavior is observed for two neighboring particles, which are separated by a distance of at least 170 nm. We exploit this observation to develop a method for finding the link axis of neighboring particles. For this purpose, a thresh-

old is applied to the pupil image. A linear regression with the remaining data points is done. We obtain the angle of the connecting axis by rotating the slope of the regression by 90° . We have applied this method to more than ten different structures. The determined link axes show a deviation of only $\pm 4^\circ$ compared with the angles determined from SEM images. The procedure is illustrated in Figure 4.

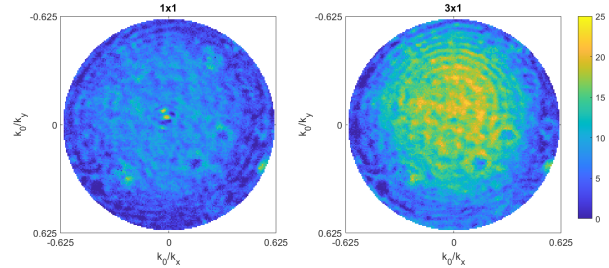


Fig. 5 Left: single particle, Right: three neighboring particles. CFS measurement with smaller aperture of 0.625.

The used samples show a particle size tight below the resolution limit. We reduce the NA from 0.9 to 0.625 to demonstrate that it is possible to detect particles and also distinguish them, if the particle size is *certainly* below the resolution limit. Figure 5 shows that with this smaller aperture it is still possible to clearly distinguish single particles from three neighboring particles. The single particles show a rotationally symmetric slight decrease in intensity towards the edges of the pupil, while in the 3x1 structures a stronger intensity modulation perpendicular to the link axis appears.

4 Conclusion

We demonstrated the detection of nanosphere assemblies of different geometries by an CFS approach. For the simulation of the scattering behavior of single particles a modification of SOMMERFELD'S half-space problem was used. The measured and calculated intensity distribution match well. Shapes and geometries of the nanoparticle assemblies are clearly distinguishable despite the small sizes of the nanostructures, i.e. at least one lateral dimension below the resolution limit.

References

- [1] A. Sommerfeld, "Über die Ausbreitung der Wellen in der drahtlosen Telegraphie," *Annalen der Physik* **333**(4), 665–736 (1909).
- [2] G. Videen, "Light scattering from a sphere on or near a surface," *J. Opt. Soc. Am. A* **9**(5), 844–845 (1992).
- [3] G. Videen, M. G. Turner, V. J. Iafelice, W. S. Bickel, and W. L. Wolfe, "Scattering from a small sphere near a surface," *J. Opt. Soc. Am. A* **10**(1), 118–126 (1993).
- [4] C. Bohren and D. Huffman, *Absorption and Scattering of Light by Small Particles*, chap. 1, pp. 7–8 (John Wiley and Sons, Ltd, 1998).