Temporal evolution of the internal structure of an evaporating droplet of suspension reflected in the distribution of the intensity of scattered light

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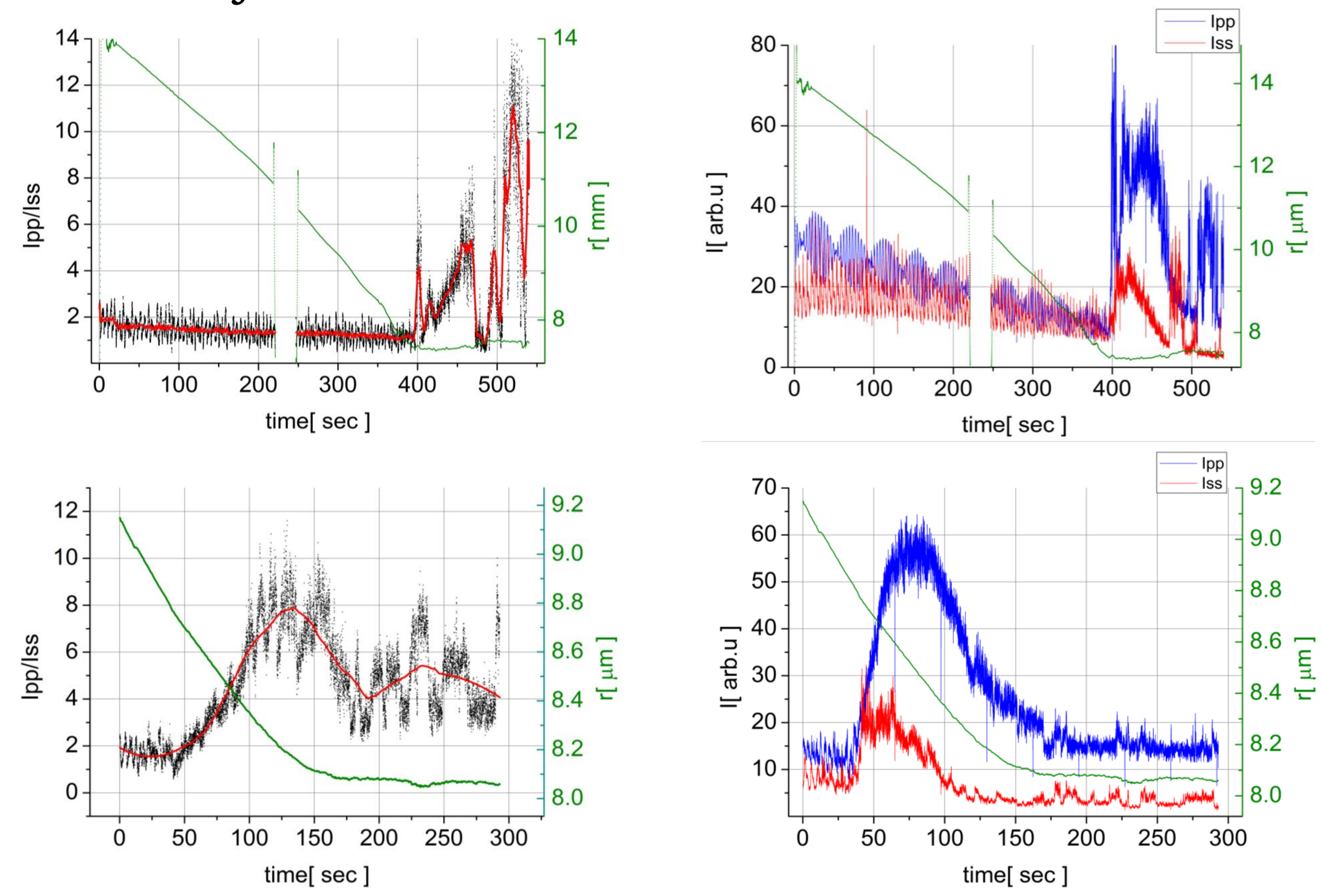
Abstract:

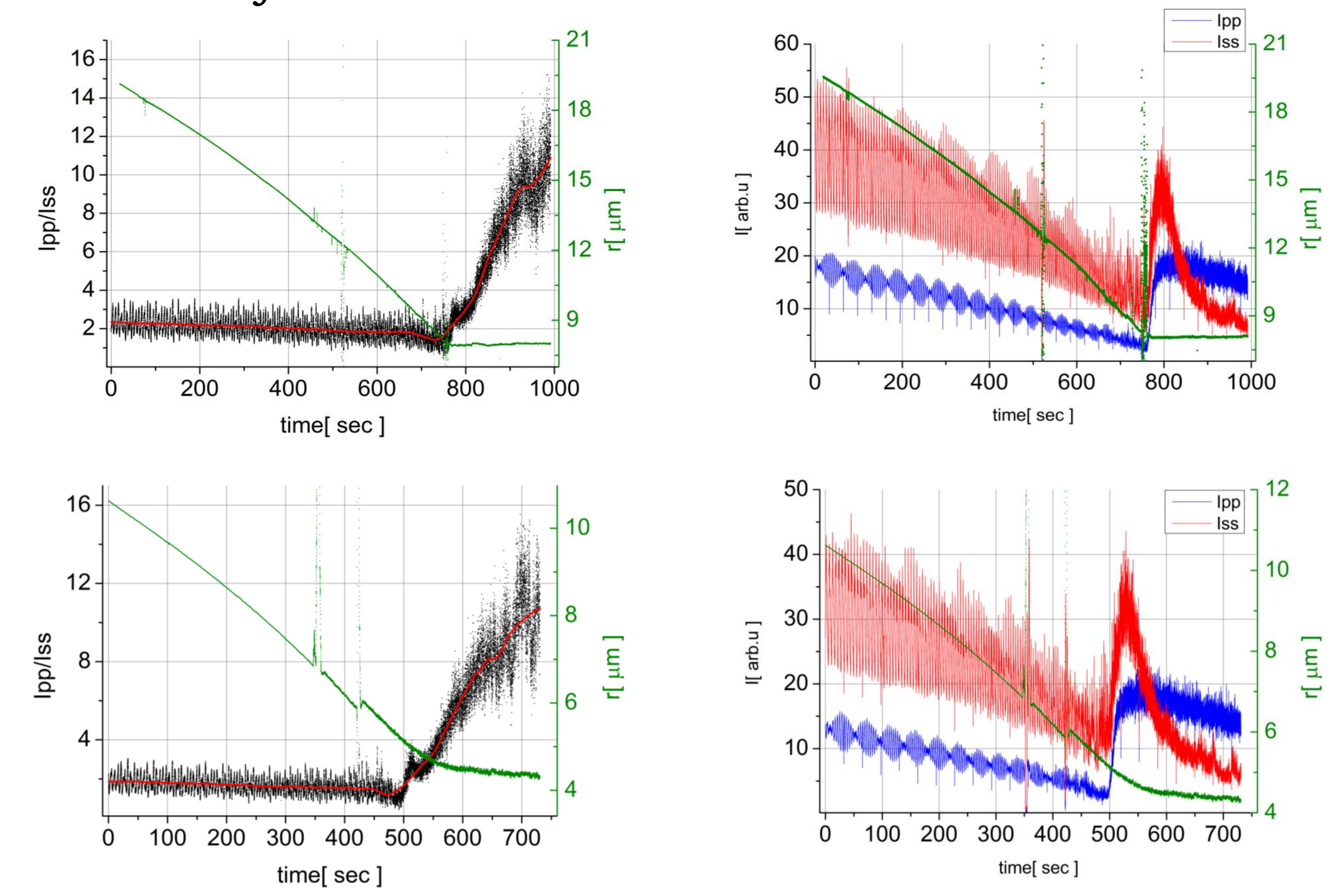
Despite extensive research on light scattering by droplets of suspension, many key aspects remain poorly understood—particularly the correlation between a droplet's internal structure and its light scattering properties. Traditional models, such as Mie theory, approximate droplets as homogeneous spheres with an effective refractive index. While this simplification is adequate in many cases, it fails when the inherent optical inhomogeneities—namely, the discrete nanoparticles—approach the size of the incident wavelength. Under these conditions, especially when inter-particle distances become comparable to the wavelength, Mie theory may underpredict scattered intensity by up to a factor of six. In our study, we investigate the relationship between the internal arrangement of nanoparticles in a droplet and its light scattering properties. Levitated, evaporating droplets confined in an electrodynamic trap allow us to precisely control the system while continuously tracking the evolution of the droplet's radius during evaporation. Because the evaporation rate depends on both the internal nanoparticle configuration and the state of the droplet's surface—which may be partially covered by suspended particles—the radius evolution serves as an indirect probe of structural changes and surface conditions. To model optical inhomogeneities, we employ SiO_2 particles of various sizes (50 nm, 100 nm, 250 nm, and 450 nm), enabling us to investigate how nanoparticle size influences the evolving scattering intensity. Additionally, two laser beams with wavelengths of 447 nm and 568 nm are used. Monitoring the dynamic evolution of scattered light intensity at these wavelengths provides estimates of the predominant structural scales within the evaporating droplet. For droplets containing small nanoparticles (<200nm), as evaporation proceeds, the particles within the droplet converge, reducing the inter-particle distance until it reaches the order of the incident wavelength. At this critical spacing, we observe a pronounced maximum in scattered light intensity, attributed to constructive interference resulting from enhanced spatial correlations among the particles. With further evaporation, as particles are forced into even closer proximity such that the spacing falls below the wavelength, the scattering intensity diminishes markedly. In contrast, droplets containing larger nanoparticles display a markedly different behavior. Due to their increased size, these particles cannot approach one another as closely, preventing sub-wavelength separations. Consequently, the droplet maintains an effective optical inhomogeneity, and the scattered light intensity remains high throughout the evaporation process.

Droplet of SiO2 nanoparticle suspension (NPs).

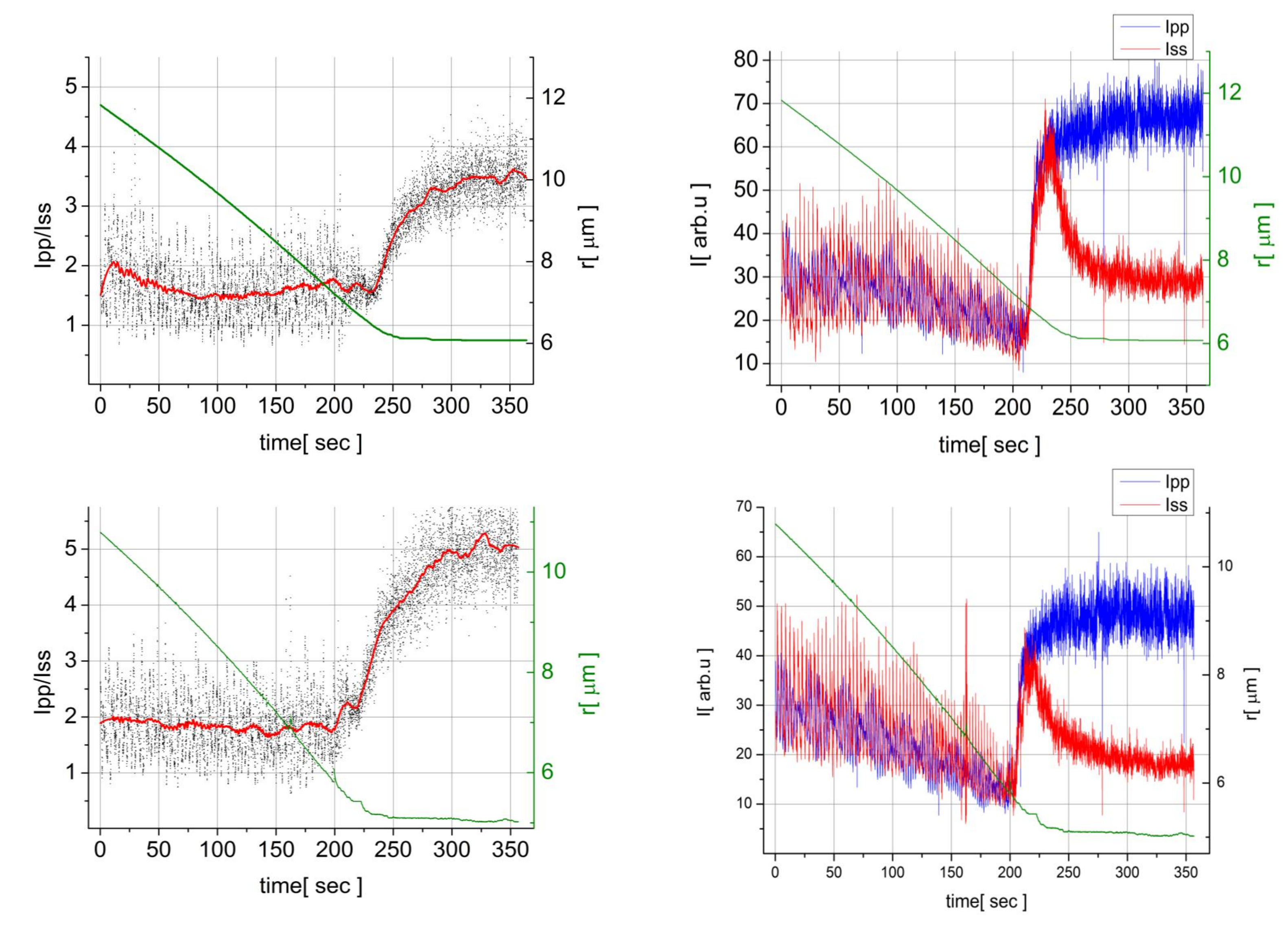
Diameter of NPs = 50nm



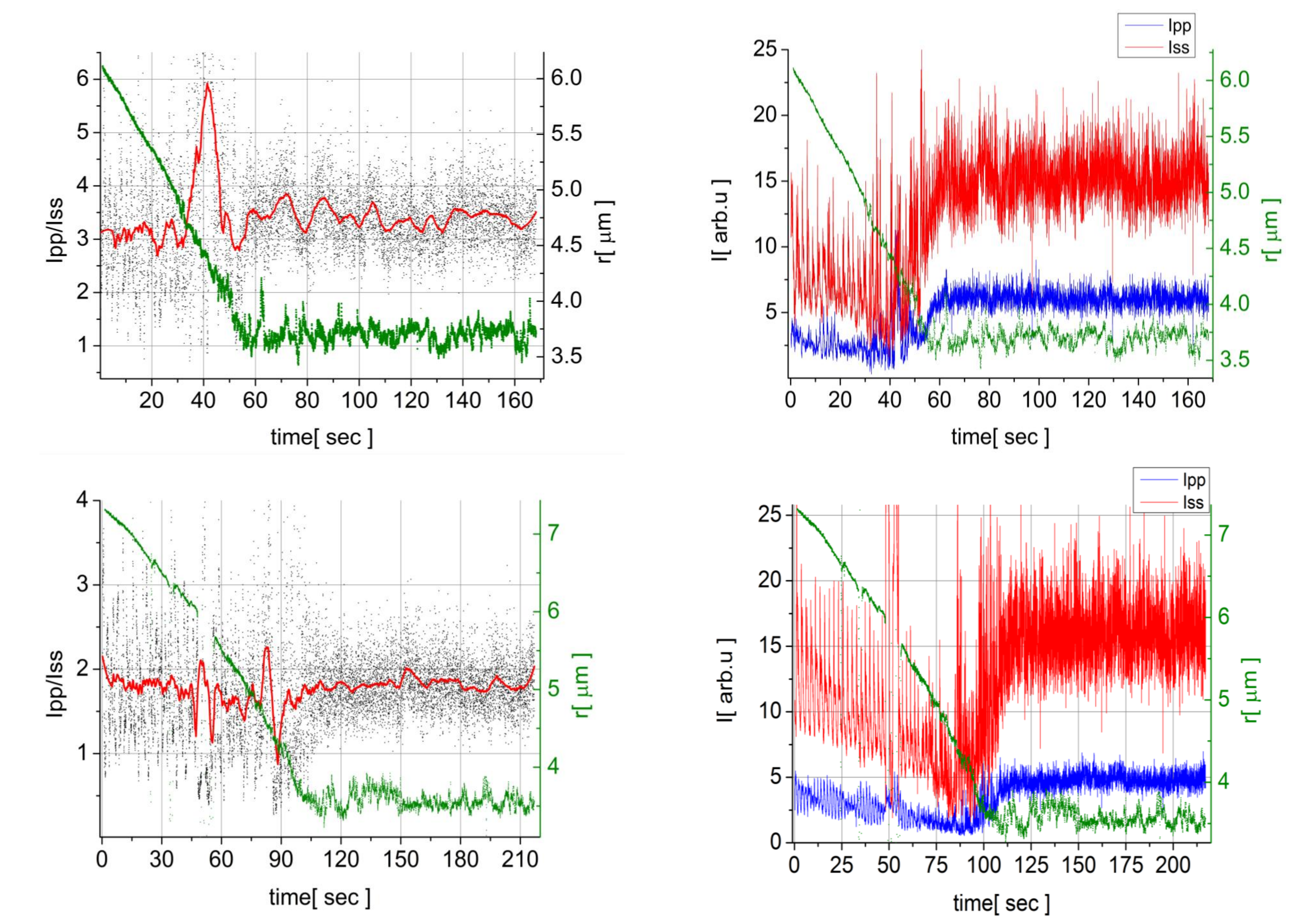
Diameter of NPs = 100nm



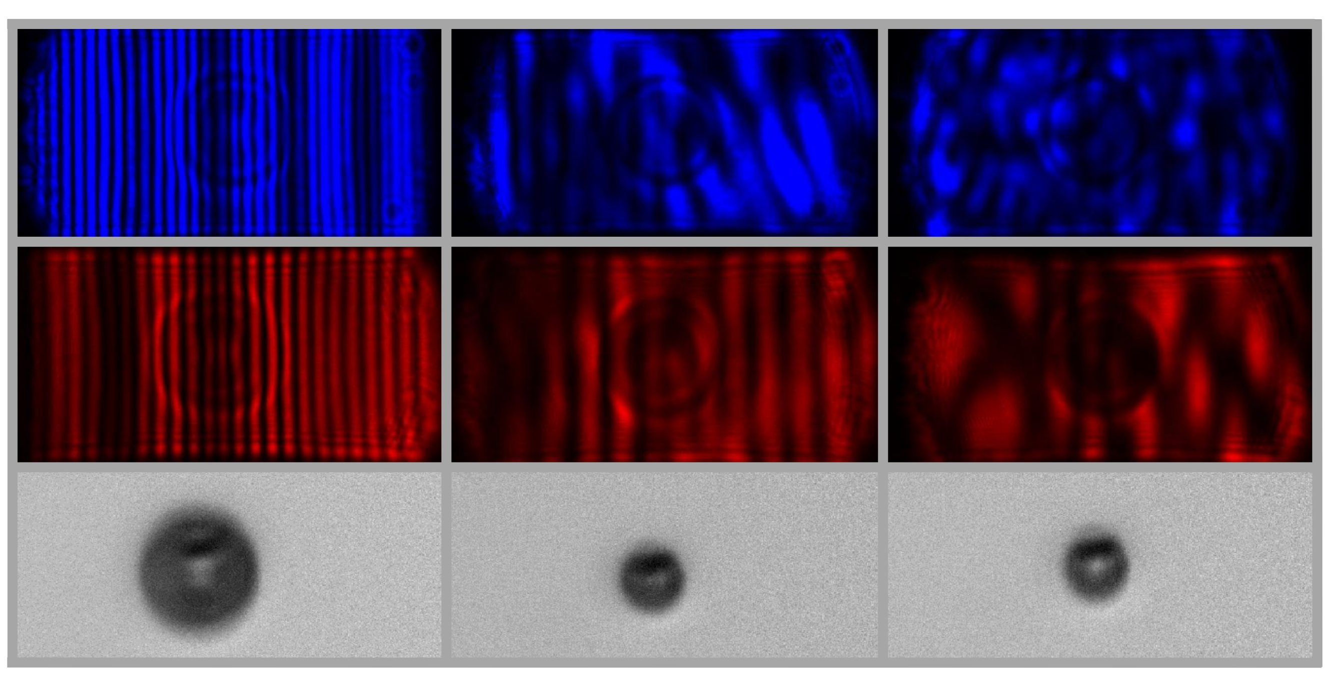
Diameter of NPs = 250nm



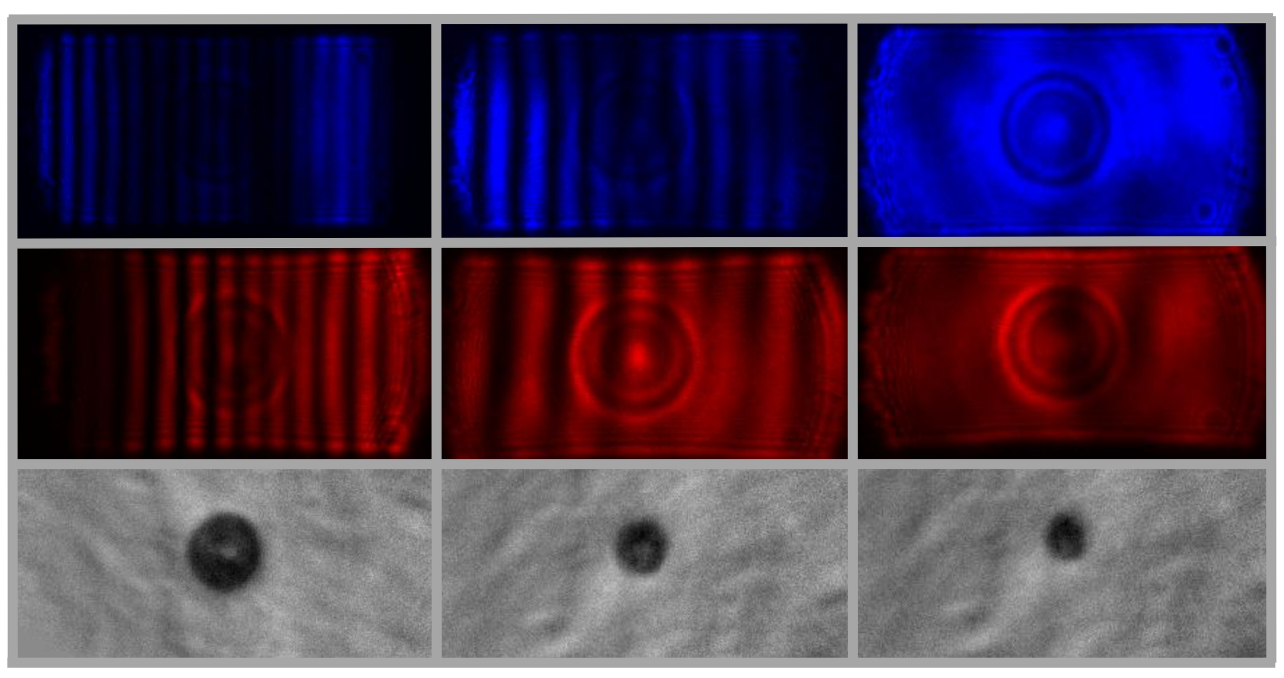
Diameter of NPs = 450nm



Exaple of light scattering on droplet of suspension (50 nm SiO_2 NPs)



Exaple of light scattering on droplet of suspension (450 nm SiO_2 NPs)



The graphs represent temporal evolution of averaged intensity of light scattered on a droplet of SiO2 NPs suspension in dipropylene glycol: Ipp(t) (blue line) p-polarized light λ = 447[nm] and Iss(t) (red line) s-polarized light λ = 658 [nm] intensities of scattered light. The green solid line represents the time evolution of the radius of the evaporating droplet. The pictures on the left represent light interference patterns (out-of-focus; red and blue) and sharp image of shadow (gray)