QUANTITATIVE PHOTOTHERMAL MICROSCOPY

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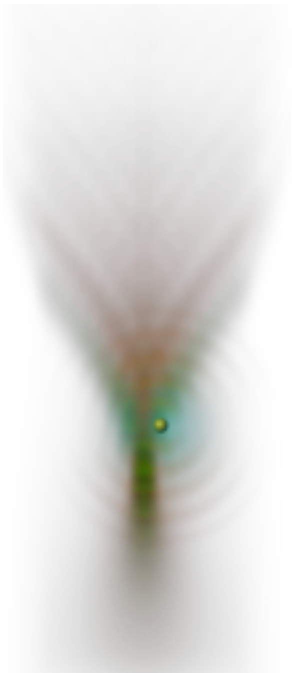
Fig.1: Detection and heating foci

Within the growing field of nano-sciences a fruitful new domain of metal nano-particles (NPs) such as AuNPs has emerged: NPs of different shapes and composition have been used as markers, field-/temperature-sources and sensors in many studies. Single particle photothermal (PT) microscopy was developed as an imaging technique to overcome the scattering cross-section limitation of regular dark/wide-field and confocal microscopes. It is based on the (Lock-In-)detected refractive index gradient (by a detection laser) generated by an absorption-induced point-source temperature field created by a second modulated heating laser (which is possibly offset along the optical axis). Therefore, even small particles down to 1nm may be detected easily due to the weaker scaling of the absorption cross-section with size as compared to the scattering cross-section.
Nonetheless, neither a rigorous nor a simple model exists which is able to describe for example the axial profiles of the detected signal, nor does a quantitative theory for the signal strength at all exist. This gap, which so far separates the ultra-sensitive imaging technique (i.e. PT) from a quantitative tool, was bridged by the development of quantitative theories which are able to reproduce the entirety of observed phenomena so far.

Here, we present ab-initio calculations of the full physical situation within a modified vector-field electromagnetic framework (GLMT [1]) that includes highly focussed beams, aberration, the refractive index gradient (as a multilayered sphere) and interfaces as well as the finite collection of the fields' energy fluxes in forward (detection) direction under the microscope. The obtained results match the experimental findings in absolute value and their parameter-dependencies (such as detection and heating focus displacements, radius, power). More importantly, they give support to 2 much simpler intuitive models which elucidate the exact physical origin of the PT signal. Such a simple model is Fresnel-diffraction with a collected phase-advance, while a further revealing model was obtained from a ray-optics treatment in combination with ABCD Gaussian beam propagation. The lens-like character of the PT signal is represented here in form of a lens-matrix with a focal length obtained through the exact solution of Fermat's least optical path differential equation.

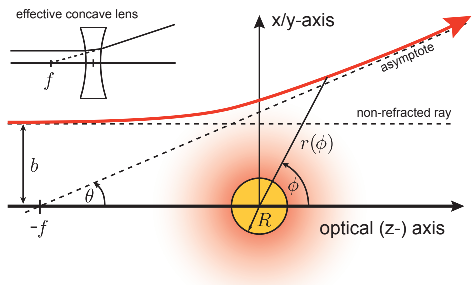

Fig.2: Solution to Fermat's differential equation

Thus, for the first time absorption coefficients may be obtained through the PT microscopy technique and it is thereby possible to accurately determine the induced temperatures of (layered) metal NPs.

[1] G. Gouesbet, J.A. Lock, G. Gréhan, "Generalized Lorenz–Mie theories and description of electromagnetic arbitrary shaped beams: Localized approximations and localized beam models, a review", *Journal of Quantitative Spectroscopy & Radiative Transfer*, **112,** (2011) 1-27