Nanointerferometric amplitude and phase reconstruction of tightly focused vector beams

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Highly confined vectorial electromagnetic field distributions are an excellent tool for detailed studies in nano-optics, such as nonlinear microscopy1, advanced fluorescence imaging2,3 or nanoplasmonics4,5. Such field distributions can be generated, for instance, by tight focusing of polarized light beams6–9. To guarantee high resolution in the investigation of objects with subwavelength dimensions, precise knowledge of the spatial distribution of the exciting vectorial field is of utmost importance. The full-field reconstruction methods presented to date involve, for example, complex near-field techniques10–13. Here, we demonstrate a simple and straightforward-to-implement measurement scheme and reconstruction algorithm based on the scattering signal of a single spherical nanoparticle as a field probe. We are able to reconstruct the amplitudes and relative phases of the individual focal field components with subwavelength resolution from a single scan measurement without the need for polarization analysis of the scattered light. This scheme has the potential to improve microscopy and nanoscopy techniques.

In the optical analysis of subwavelength objects such as cellular structures14 or plasmonic particles15,16,17, nano-optical tools including highly resolving microscopy techniques are used. Because such methods utilize complex and highly confined vector fields, exact knowledge of the corresponding spatial field distributions is crucial. In recent decades, several techniques have been proposed to map these focal fields, for example using metal knife edges16,17 to probe the total electric energy density distribution, fluorescence molecules18, tapered fibres19,20 or tip-based methods10 to image specific field orientations, or near-field scanning optical microscope (NSOM) techniques to extract amplitude and even phase information11–13. The NSOM-based methods require complex measurement and detection schemes and calibration procedures to allow for amplitude and phase mapping of individual field components. As an alternative approach to measure phase information, a single-particle scattering scheme has also been proposed21, where the authors show that Mie scattering can distinguish the topological charge of vortex beams.

We now demonstrate a precise and easily implementable reconstruction technique for highly confined field distributions created by arbitrary focusing systems, based on what we call ‘Mie scattering nanointerferometry’. The basic concept of this reconstruction method can be understood as follows. We use a metallic nanoparticle on a glass substrate as a local field sensor and scan it stepwise through the field distribution under investigation. By collecting the transmitted light in an angularly resolved form22 and exploiting the interference signal between incoming field and the field scattered off the nanoparticle at each point in the angular spectrum, the initial three-dimensional focal field distribution can then be determined accurately in amplitudes and relative phases of the individual field components. The interference, and hence the phase information, is preserved herein by effectively changing the observation direction.

To analytically describe the full scattering process and thus incorporate the disturbance of the initial field distribution by the probe, the unknown focal field is first expanded into electromagnetic multipoles. With this choice of a basis system, only the lower expansion orders have to be taken into account for highly confined fields (Supplementary Section 7)13,24. The focal electric field distribution can thus be expressed as

$$E_{in}(r) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} A_{mn} N_{mn}(r) + B_{mn} M_{mn}(r)$$

where $N_{mn}$ and $M_{mn}$ are regular vector spherical harmonics representing differently oriented electric and magnetic multipoles25,26 expanded around the geometrical focus. This transfers the full information of the unknown focal field from the complex-valued electric field components $E_{in}(r)$ at each point in the focal plane to the complex-valued multipole expansion coefficients $A_{mn}$ and $B_{mn}$. This representation allows the relation of distinct far-field patterns with the focal field distribution under investigation, where electric dipoles can be associated with the local electric field $E_{in}(s)$ at the expansion point $r_s$ and higher-order multipoles with higher-order moments of the field. In addition, it permits a simple treatment of scattering problems via the T-matrix approach22.

The focal electric field distribution can thus either be reconstructed by determining all multipole coefficients of the focal field at one single point or by scanning the dipole contributions in the focal plane and relating them to one common expansion point via the translation theorem for vector spherical harmonics27. In both cases, to achieve the necessary high position accuracy and repeatability, the probe is immobilized on a substrate in our experiments, which introduces an interface in the scattering problem (Fig. 1). The resulting transmitted field $E_i$ can then be expressed as the incoming field $E_{in}$ and a scattering field term $E_s$, taking the interaction of the focal field with the nanoprobe and interface fully into account:

$$E_i(r) = E_{in}(r) + E_s(r)$$

For a simplification of the description of the interface, we transform the basis functions in the forward hemisphere to $N_{mn}$ and $M_{mn}$, leaving the expansion coefficients $A_{mn}$ and $B_{mn}$ unaltered and thus keeping the low-order multipole expansion (Supplementary Section 1). The scattered electric field $E_s$ is then related to the incoming field $E_{in}$ by describing the
interaction of the nanoprobe and the substrate with the incoming light field by an effective scattering matrix $T_{\text{eff}}$:

$$E_{\text{in},\text{t}} = (1 - T_{L_{\text{R}}}^{(1)})^{-1} T_{L_{\text{R}}}^{(1)} E_{\text{in},\text{s}} = T_{\text{eff}} E_{\text{in},\text{s}} \quad (3)$$

where $T$ is the full scattering matrix of the probe particle determined by Mie theory (including information about the particle shape, size and its optical properties) and $L_{\text{R}}^{(1)}$ represents the reflection operators of the substrate (Fig. 1, Supplementary Section 1). Both the presence of the substrate and the nanoparticle change the field distribution in the plane of observation (the focal plane of the focusing lens). The $T$-matrix in equation (3) relates these changes to the input field and allows for its reconstruction. Hence, this method describes the scattering process rigorously, relying only on the precise knowledge of the parameters of the nanoprobe. These can be determined experimentally with sufficient precision by means of scanning electron microscopy and spectral measurements.

To relate the unknown input field to a directly measurable quantity, the resulting power transmitted at an angle $(\theta, \varphi)$ (Fig. 2a) can then be expressed by

$$P_{\text{in}}(\theta, \varphi) = P_{\text{in}}(\theta, \varphi) + P_{\text{t}}(\theta, \varphi) + P_{\text{ext}}(\theta, \varphi) \quad (4)$$

following the classical Mie scattering problem\(^{29}\), with

$$P_{\text{in}} = \frac{1}{2} \text{Re}[E_{\text{in},\text{t}} \times H_{\text{in},\text{t}}]$$
$$P_{\text{t}} = \frac{1}{2} \text{Re}[E_{\text{t},\text{t}} \times H_{\text{t},\text{t}}]$$
$$P_{\text{ext}} = \frac{1}{2} \text{Re}[E_{\text{ext},\text{t}} \times H_{\text{ext},\text{t}} + E_{\text{ext},\text{s}}^* \times H_{\text{ext},\text{s}}] \quad (5)$$

as the incoming, scattered and extinct power measured in transmission. Here, the magnetic field components $H_{\text{t}}$ are determined from the electric field components $E_{\text{t}}$ in the far-field using the plane wave spectrum of the transmitted multipoles (Supplementary Section 1). The far-field interference term $P_{\text{ext}}$, which depends on both $\theta$ and $\varphi$, not only allows for the extraction of amplitude information about the multipole expansion coefficients, but also the phase relation between them. A reference field that can interfere with the light field scattered off the nanoprobe in the far-field (here, the transmitted input field) is therefore essential for the proposed technique (see equation (5)). Hence, it is applicable to all systems and scenarios that exhibit such a far-field reference term.

It can be shown that, for an unambiguous reconstruction of both the amplitudes and relative phases of the individual electric field components, the combination of the following collection schemes is sufficient. First, the transmitted power $P_{\text{t}}$ has to be recorded and integrated in a given solid angle $\theta_{\text{a}}$ around the optical axis for each position of the particle in the focal plane (Fig. 3a, top and centre). With this integration over the whole range in $\varphi$, some phase information is lost. Thus, to regain the full information about the relative phases, an effective break of the cylindrical symmetry of the collection system (objective) is introduced. This can be realized by additionally choosing a collection sector and thus performing a $\varphi$-integration from $\varphi_1$ to $\varphi_2$ (with $|\varphi_1 - \varphi_2| < 2\pi$; Fig. 3a, bottom). With this choice, light emitted in a certain direction relative to the optical axis is analysed. Both schemes can be realized easily in the experiment, as discussed below. With the precisely known scattering matrix $T_{\text{eff}}$, the powers transmitted into different angular ranges for each position of the nanoprobe relative to the input beam thus establish a system of positive quadratic forms of the unknown multipole expansion coefficients. The inversion of this equation system then leads to the vectorial focal electric field distribution. Further collected angular ranges, as well as an adapted choice of step size, can additionally increase the accuracy of the reconstructed focal field distribution (Supplementary Sections 2 and 5 to 7).

For an experimental demonstration of the introduced nanointerferometric vectorial field reconstruction scheme, a custom-made

**Figure 1** Schematic of the scattering process. The interface leads to a coupling of the reflected field from the substrate $L_{\text{R}}^{(1)} \cdot E_{\text{in}}$ and $L_{\text{R}}^{(1)} \cdot E_{\text{in}}$ and the scattered field off the spherical scatterer $E_{\text{s}}$.

**Figure 2** Sketch of the experimental implementation of the reconstruction scheme. a. A spherical metallic nanoparticle adhered to a glass substrate is scanned through the focal field distribution under investigation. The resulting scattered and transmitted intensity is collected with a variable solid angle $(\theta_{\text{a}}, \varphi)$, b. Experimental set-up. An arbitrarily structured input beam is focused by high-NA microscope objectives (NA = 0.9). The nanoprobe (spherical gold nanoparticle; diameter 82 nm) can be precisely scanned through the focal plane using a three-dimensional piezo stage. The transmitted light is then collected by an immersion-type microscope objective (NA = 1.3) and its back focal plane is imaged onto a CCD camera. The reflected light can also be measured to ensure the exact focus position.
Figure 3 | Experimental results and theoretical comparison for a radially polarized vector beam. 

(a) Image of the back focal plane of the collection objective with NA = 1.3 for one position of the nanoprobe relative to the focal field. The measured intensity scan images in transmission for a wavelength of 530 nm correspond to three different collecting solid angles—within full aperture of NA = 0.9 and 0.4, and with an azimuth angle of φ = π/2 rad for an NA = 0.9—all derived from the same measured back focal plane images for different probe positions. The intensity is normalized to the total intensity of the input beam. 

(b) Squared electric field components |E|^2 and relative phases Φ, in the focal plane reconstructed from the measured intensity distributions up to a multipole order of n = 8. 

(c) Energy density distribution for the three field components in the focal plane of the same beam calculated via vectorial diffraction theory. 

Insets: calculated phase distribution, with the same scale and colourmap as in (b).
By choosing a radially polarized doughnut beam as an exemplary input field, a three-dimensional vectorial field distribution was generated under tight focusing conditions, exhibiting an on-axis longitudinal field component26,28 and off-axis transverse field components (Fig. 3c, calculated via vectorial diffraction theory29). Such a tightly focused cylindrical vector beam finds applications in several fields of nano-optics and imaging29,30. Figure 3a presents the experimental results (Supplementary Section 8 also shows a different input field configuration). For every position of the nanoparticle (scan step size: 25 nm; Supplementary Section 6) a single image of the back focal plane was recorded (Fig. 3a, left column for one position of the particle). From this camera data, two-dimensional scan images were derived by plotting the transmitted intensity integrated over the corresponding angular range in the back focal plane for each probe position (Fig. 3a, right column; Supplementary Section 3). We chose three different angular ranges for integration in the measured back focal plane images to reduce the influence of experimental noise (Supplementary Section 2). Solid angles were chosen, corresponding to two full apertures with NA = 0.9 and 0.4 and a sector of $\varphi_s = 1\, \text{rad}$ at NA = 0.9. The non-rotational symmetric collection angle preserves the interference information (Fig. 3a, bottom). This signal is highly sensitive to the actual relative permittivity of the nanoprobe, which was determined experimentally for the selected nanoprobe and wavelength to be $\varepsilon_{\text{NANP}} = -3.0 + 2.1\, \text{i}$ (Supplementary Section 4).

The electric energy densities and phases of the focal field components reconstructed from these intensity distributions (Fig. 3b) show a very good overlap with the calculated field amplitudes shown in Fig. 3c. When compared with these theoretical data, slight deviations are visible in both reconstructed amplitude and phase distributions. These deviations are predominantly caused by the experimental imperfection of the focal field distribution itself and are not introduced by the reconstruction algorithm (Supplementary Sections 3 to 7). Furthermore, the influence of experimental noise in the scan images on the final reconstruction is also shown to be significantly smaller than the observed deviations. Hence, these deep subwavelength differences can be attributed to imperfections of the field distribution under study, introduced, for instance, by aberrations of the focusing system and resolved by our reconstruction technique (Supplementary Fig. 3 and Section 6).

In summary, we have shown an easily applicable reconstruction scheme to determine the full vectorial amplitude and relative phase distributions of highly confined electromagnetic fields. The technique relies on nanointerferometry, that is, the interference between the input field and the field scattered off a nanoprobe, as well as an angularly resolved measurement of the resulting far-field intensity. By adapting the developed scheme, a reconstruction of the field distributions for other focusing systems or confined fields with a propagating far-field reference (equation (5)) can also be realized, including, for example, the near-field distribution of NSOM tips. Furthermore, we believe that the proposed scheme can also help to significantly improve imaging techniques such as laser scanning microscopy.

Methods

Experimental set-up. A tunable light source (NKT Photonics SuperK PowerPlus with an AA-Opto-Electronic MDScnT-NC acousto-optical tunable filter) provided a linearly polarized Gaussian beam at a wavelength of 530 nm, which was optionally converted into a radially or azimuthally polarized doughnut beam by a liquid-crystal radial polarization converter (ARCoptix). The beam was then guided via four mirrors top-down onto a microscope objective with NA = 0.9 (Leica HCX PL FLUOTAR x100/1.3 OIL). In addition, the reflected light was collected by the focusing objective. The angular distributions of the transmitted and optionally the reflected light were detected via imaging the back focal plane of the corresponding objectives on CCD cameras (The Imaging Source DMK 23G618).

Sample preparation. The spherical gold nanoparticle was fabricated at the Max Planck Institute for Polymer Research by laser-induced melting of a commercial colloidal gold solution. The resulting solution of spherical nanoparticles was drop-coated on a microscope coverslip with a thickness of 170 µm and prestructured gold markers in order to reproducibly measure one single nanoparticle.

Received 8 May 2013; accepted 26 September 2013; published online 10 November 2013

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Acknowledgements

The authors thank M. Neugebauer and S. Batz for discussions and M. Schmelzeisen from the Max Planck Institute for Polymer Research in Mainz for the fabrication of the scattering particle.

Author contributions

G.L., P.B., S.O. and U.P. conceived the idea. P.B. and T.B. designed the experiment. S.O. and P.B. developed the theoretical algorithm and procedure. T.B. performed the experiment. S.O. and T.B. analysed the data. G.L. and P.B. supervised all aspects of the project. All authors contributed to the text of the manuscript.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to P.B.

Competing financial interests

The authors declare no competing financial interests.