Transverse Kerker Scattering for Ångström Localization of Nanoparticles

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(Dated: April 27, 2018)

Ångström precision localization of a single nanoantenna is a crucial step towards advanced nanometrology, medicine and biophysics. Here, we show that single nanoantenna displacements down to few Ångströms can be resolved with sub-Ångström precision using an all-optical method. We utilize the transverse Kerker scattering scheme where a carefully structured light beam excites a combination of multipolar modes inside a dielectric nanoantenna, which then upon interference, scatters directionally into the far-field. We spectrally tune our scheme such that it is most sensitive to the change in directional scattering per nanoantenna displacement. Finally, we experimentally show that antenna displacement down to 3 Å is resolvable with a localization precision of 0.6 Å.

Nanoparticles are fundamental building blocks in modern nanophotonic devices and experimental schemes. Accordingly, depending on the actual application, various antenna designs have been proposed and investigated in recent years [1–4]. In bio-sensing applications, bowtie metal antennas can be used to substantially enhance the field locally, thus enabling single molecule detection and surface enhanced Raman spectroscopy [5, 6]. On the other hand, Yagi-Uda type antennas are important components in optical circuitry to realize far-field to near-field coupling and vice-versa [7, 8]. Furthermore, the spectral composition of an excitation field can be deduced from the pattern of the light scattered off a bi-metallic nanoantenna [9].

Besides the aforementioned complex antenna designs, even single-element antennas such as cylinders and spheres can exhibit interesting scattering properties, such as, directional emission and coupling via polarization dependent spin-momentum locking [10–12]. Another example are scatterers, which fulfill Kerker’s condition [13–15]. This effect is typically associated with simultaneous excitation of electric and magnetic dipoles (Huygens’ dipole), resulting in enhanced or suppressed forward/backward scattering [16–19]. In Refs. [11, 20–22], a localization scheme was proposed and discussed based on enhanced or suppressed scattering in the direction transverse to propagation. Here, we present a detailed spectral analysis of transverse Kerker scattering off a single silicon particle. First, we elaborate on the general concept for a particle in free-space. Then, we present an actual experimental implementation and tune the wavelength of the excitation field to optimize the position-dependent directional scattering. By choosing optimal parameters, nanoantenna displacements down to few Ångströms can be resolved with sub-Ångström precision and accuracy.

As proposed by Kerker et al. [14], a plane-wave-like excitation of a small spherical particle (radius ≪ λ) can result in asymmetric forward/backward scattering depending on the amplitudes and phases of the induced electric and magnetic dipole moments. A Huygens’ dipole with zero back-scattering can be achieved when the induced electric and magnetic dipolar scattering coefficients are equal in amplitude and phase [23, 24], as can be seen in the sketch in Fig. 1(a). Equivalently, upon interference of a longitudinal and a transverse dipole, it is possible to achieve what we refer to as transverse Kerker scattering, that is, directional scattering perpendicular to the propagation direction of the beam (transverse Kerker scattering). A tightly focussed azimuthally polarized beam propagating along the z-axis can induce longitudinal magnetic $m_z$ and transverse electric $p_y$ dipoles, resulting in transverse Kerker scattering [inset similar to (a)].

![FIG. 1. Kerker scattering and Huygens’ dipole. (a) Zero backscattering. A y-polarized plane wave with $\mathbf{k} = k_y \mathbf{\hat{e}}_y$ excites transverse electric and magnetic dipoles $p_y$ and $m_z$ of equal amplitudes and phases, resulting in a Huygens’ dipole and zero backscattering. (Inset) Polar plot of the resulting total far-field intensity of the scattered light (black) and the individual emission patterns of $p_y$ (red) and $m_z$ (blue) in the meridional $xz$-plane. (b) Directional scattering perpendicular to the propagation direction of the beam (transverse Kerker scattering). A tightly focussed azimuthally polarized beam propagating along the z-axis can induce longitudinal magnetic $m_z$ and transverse electric $p_y$ dipoles, resulting in transverse Kerker scattering [inset similar to (a)].]
by a tightly focused beam can be treated by generalized Mie theory [28]. In Fig. 2(a), we plot the scattering cross-sections of the electric dipole (ED), magnetic dipole (MD), and magnetic quadrupole (MQ) as red, blue, and purple lines within the visible spectral range for the particle in free-space. In the dominant part of the depicted spectrum, the MQ contribution can be neglected and we can approximate the nanoparticle as a dipole, such that the induced dipole moments are proportional to the local electromagnetic field components, \( \mathbf{p} \propto T_{\text{ED}} \mathbf{E} \) and \( \mathbf{m} \propto T_{\text{MD}} \mathbf{H} \) [20]. The proportionality factors, \( T_{\text{ED}} \) and \( T_{\text{MD}} \) are the electric and magnetic dipole scattering coefficients calculated using Mie theory, which define the strength and phase of the induced dipole moments [29]. They are linked to the scattering cross-sections by \( \sigma_{\text{sca}} \propto |T_{\text{ED}}|^2 \) and \( \sigma_{\text{sca}} \propto |T_{\text{MD}}|^2 \) [29]. Since achieving transverse Kerker scattering depends not only on amplitudes but also on phases, we depict the relative phase between the two dominant ED and MD resonances in the lower graph of Fig. 2(a). The black dotted circles and corresponding gray areas around \( \lambda = 535 \text{ nm} \) and \( 630 \text{ nm} \) denote the wavelength ranges where electric and magnetic dipoles are \( \pi/2 \) out-of-phase. The importance of these wavelengths will become clear, when we discuss the impinging light field in the following.

For excitation, we use tailored inhomogeneous electromagnetic field distributions obtained by tightly focusing azimuthally and radially polarized vector beams [30–32]. Fig. 2(b) shows the focal-plane electric and magnetic intensity and phase distributions of the field components: \( \mathbf{E}_x, \mathbf{E}_y, (E_x \mathbf{e}_x + E_y \mathbf{e}_y), \mathbf{H}_x, \) and \( \mathbf{H}_y, (H_x \mathbf{e}_x + H_y \mathbf{e}_y) \), calculated using vectorial diffraction theory [33, 34]. Both, the intensity and phase distributions of the transverse and longitudinal field components exhibit cylindrical symmetry. The amplitudes of the transverse components \( |\mathbf{E}_y| \) and \( |\mathbf{H}_y| \) are zero on the optical axis for both input beams, and in close proximity to the optical axis (\( r \ll \lambda \)), can be approximated to increase linearly with radial distance [20]. The intensity of the longitudinal field component—only \( \mathbf{E}_x \) (\( \mathbf{H}_z \)) is present for radial (azimuthal) polarization—are maximum on the optical axis and significantly stronger than the transverse components for \( r \ll \lambda \). Another important aspect is the phase retardation of \( \pm \pi/2 \) between the longitudinal and transverse field components [see insets in Fig. 2(b)]. By choosing the excitation wavelengths \( \lambda = 535 \text{ nm} \) and \( 630 \text{ nm} \), the aforementioned phase difference between \( T_{\text{ED}} \) and \( T_{\text{MD}} \) cancels the phase retardation between the longitudinal and transverse components of the excitation fields.

Consequently, when the wavelength is optimized with respect to the resonances of the particle, longitudinal and transverse dipole moments with a relative phase of 0 or \( \pi \) can be induced, resulting in transverse Kerker scattering. Furthermore, the relative amplitudes between longitudinal and transverse dipole moments can be adapted by changing the radial distance between the particle and the center of the beam. Therefore, our system enables tailoring of transverse Kerker scattering, which will be discussed with examples. The nanoantenna positioned in the focal plane at \((x, y) = (50, 0) \text{ nm} \) can be considered as a combination of \( p_z, m_y, \) and \( p_x \) dipoles for the radially polarized beam, and \( m_z, p_y, \) and \( m_x \) dipoles for the azimuthally polarized beam. The relative amplitudes and phases of each dipole moment can be determined from Figs. 2(a) and (b). In Fig. 2(c), we plot the resulting far-field intensity of the scattered light in the meridional \( xx \)-plane for \( \lambda = 535 \text{ nm} \) (green) and \( \lambda = 630 \text{ nm} \) (magenta). Highly directional transverse Kerker scattering...
can be observed for radial polarization at $\lambda_{\text{rad}} = 630$ nm (directionality in negative $x$-direction) and for azimuthal polarization at $\lambda_{\text{azim}} = 535$ nm (directionality in positive $x$-direction). In contrast, the other two corresponding plots indicate a much weaker directionality, highlighting the wavelength dependence of the transverse Kerker scattering, which will be discussed in detail below.

In the actual experimental implementation of transverse Kerker scattering based localization, the nanoantenna is placed on a dielectric interface (air-glass), which substantially modifies the scattering scheme from the aforementioned free-space scenario. To analytically describe the full scattering process, we start with determining the complete scattering matrix $T$ of the nanoantenna sitting on an interface [35], such that the incident field $E^{\text{inc}}$ and the scattered field $E^{\text{sca}}$ are related by $E^{\text{sca}} = TE^{\text{inc}}$. We expand our highly confined focal field into electromagnetic multipoles [36–38] as

$$E^i = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} (a_{mn}^{i} N_{mn}^{i} + b_{mn}^{i} M_{mn}^{i}),$$

where $i$ corresponds to either the incident or the scattered fields. $N_{mn}^{i}$ and $M_{mn}^{i}$ are vector spherical harmonics (regular or irregular type for incident or scattered field respectively) representing the electric and magnetic multipoles expanded around the center of the particle [36].

The complex-valued multipole expansion coefficients $a_{mn}^{\text{inc}}$ and $b_{mn}^{\text{inc}}$ for the incident field contain full information about the electric $E^{\text{inc}}(r)$ and magnetic $H^{\text{inc}}(r)$ field components at each point $r$. Following [29, 39], we model the influence of the interface by considering the effect of incident and scattered light reflected from the interface. Hence, the expansion coefficients $a_{mn}^{\text{sca}}$ and $b_{mn}^{\text{sca}}$ representing the induced multipole moments can be obtained from the auxiliary scattered field above the interface $E^{\text{sca}}(r)$, which is related to $E^{\text{inc}}$ via the effective scattering matrix $T_{\text{eff}}$ as

$$E^{\text{sca}} = T_{\text{eff}} E^{\text{inc}} = \frac{T(1 + L_{R}^{(1)} a_{mn}^{\text{sca}} + b_{mn}^{\text{sca}})}{1 - TL_{R}^{(3)}} E^{\text{inc}},$$

where $L_{R}^{(1,3)}$ are the reflection operators of the interface for the incident and scattered light (see more details in the supplementary of [39]). These expansion coefficients $a_{mn}^{\text{sca}}$ and $b_{mn}^{\text{sca}}$ can then be used to calculate the light emitted into the glass substrate $E^{\text{sca}}$ taking into account the transmission Fresnel coefficients [36, 39]. In particular, we consider the emission at the critical angle, which is equivalent to the transverse plane in free-space. Furthermore, the emission pattern peaks at this angle [34]. The transmitted far-field intensity $I \propto |E_{\text{eff}}^{\text{inc}}|^2$ along the criti-
cal angle is then used to numerically calculate the wavelength and position-dependent transverse Kerker scattering for azimuthally and radially polarized beams.

Regarding the experimental implementation, a sketch of our setup is depicted in Fig. 3(a). We tightly focus an incoming beam with a microscope objective of numerical aperture (NA) of 0.9 onto a silicon nanoantenna (SEM image shown as inset) sitting on a glass substrate, which can be precisely positioned within the focus by a piezo-stage. We collect the transmitted and forward scattered light with a second (oil-immersion type) microscope objective of NA=1.3, and image the back-focal plane (BFP) of said objective onto a CCD camera. Similar to Ref. [20], we only consider the region, NA∈[0.98,1.3], where we can detect the scattered light without the transmitted beam. For each wavelength, we scan our nanoantenna within the focal plane and obtain BFP images for each (x,y) position [examples plotted in Figs. 3(b-c)].

To define the position-dependent strength of the directional scattering, we calculate the difference between the light scattered into opposite directions in k-space (kx,ky): \( D_x = (I_3 - I_1)/I_{tot} \) and \( D_y = (I_2 - I_4)/I_{tot} \), where \( I_{tot} = (I_1 + I_2 + I_3 + I_4)/2 \). Here, \( I_j \) \((j \in [1,4])\) is the average intensity of the \( j^{th} \) region in the BFP, which corresponds to the angular region NA∈[0.98,1.03] around the critical angle as indicated in Figs. 3(b) and (c) for the particle positioned on the optical axis. Figs. 3(b) ii-v and (c) ii-v show exemplary BFP images indicating varying extents of transverse directivity for different wavelengths. For each wavelength, we obtain \( D_x(x) \) and \( D_y(y) \) curves by fitting our experimentally measured directivity for the nanoantenna displacement along the x- and y-axis. In line with the behavior of the transverse electromagnetic fields, the directivity \( D_x(x) \) and \( D_y(y) \) exhibit a linear relationship with displacement within at least 30 nm around the optical axis. In Fig. 3(d), without loss of generality, \( D_x(x) \) is plotted against the x-position of the particle for azimuthally and radially polarized beams of wavelengths \( \lambda_{azim} = 545 \text{ nm} \) and \( \lambda_{rad} = 630 \text{ nm} \). Each data point in these calibration curves in Fig. 3(d) is a statistical representation of more than 50 measurement values. Fitting fluctuations shown as error bars represent the stability of our current experimental setup and do not reflect upon the localization resolution (see more details below). As we can see, a very high directivity of \( D_x \approx 0.35 \) for radial and even higher directivity of \( D_x \approx 0.76 \) for azimuthal polarization can be achieved for a displacement as small as 30 nm. In order to define a parameter describing the position sensitivity of our experimental setup, we use the average change in directivity along the x- and y-axis within the region of linearity, \( \Delta D_x = \partial D_x/\partial x \) and \( \Delta D_y = \partial D_y/\partial y \). With respect to Fig. 3(d), \( \Delta D_x \) represents the slopes of the two linear fits. In Fig. 3(e), we present a spectral analysis of \( \Delta D_x \pm \delta \Delta D_x \) for radial (red) and azimuthally polarized (blue) beams where \( \delta \Delta D_x \) is the fitting error of the slope represented as errorbars. For comparison, we plot the numerically calculated results (bold lines) based on the theoretical model presented earlier. As can be seen, the parameter \( \Delta D_x \) is maximum around \( \lambda_{azim} = 545 \text{ nm} \) and \( \lambda_{rad} = 630 \text{ nm} \), where the electric and magnetic dipoles are induced with a relative phase close to 0 or \( \pi \) [see free-space scenario in Fig. 2(a)]. Also, for azimuthal polarization (\( \lambda_{azim} \approx 545 \text{ nm} \)), the change in directivity \( \Delta D_x(y) \) is more than two times stronger than for radial polarization (\( \lambda_{rad} \approx 630 \text{ nm} \)). The result highlights that, for the utilized silicon particle, the best choice for transverse Kerker scattering based localization is an azimuthally polarized excitation beam with \( \lambda_{azim} = 545 \text{ nm} \), since a stronger directivity leads to enhanced localization accuracy.

In order to demonstrate our best localization accuracy, we consider differential BFP images—differences between two BFP intensity distributions corresponding to two particle positions—for \( \lambda_{azim} = 545 \text{ nm} \), where the relative nanoantenna displacements were less than 1 nm. Such small displacements were not possible to achieve deterministically with our current experimental setup (position uncertainty of ±4 nm), so we utilized the vibrational stochasticity of our system to obtain an ensemble of corresponding nanoantenna positions. Exemplary differential BFP images are shown in Fig. 4. The histogram plots in the center shows the pixel-intensity distribution of \( \Delta I_j \), for the \( j^{th} \) regions marked in the differential BFP images. Region \( j = 1,3 \) (dashed black border) corresponds to a movement \( \Delta x \) and region \( j = 2,4 \) (solid black border) for \( \Delta y \). Gaussian fits to the histograms \( \Delta I_j \) allow us to estimate the relative displacements along x- and y-axis such that

\[
\Delta x = \frac{\langle \Delta I_1 \rangle - \langle \Delta I_3 \rangle}{\Delta D_x}, \quad \Delta y = \frac{\langle \Delta I_2 \rangle - \langle \Delta I_4 \rangle}{\Delta D_y}.
\]

The localization precision is calculated using an error
propagation formula considering both $\partial \Delta D_{xy}$ and $\sigma_M$, where $\sigma_M$ is the standard error of the mean given as $\sigma_M = \sigma / \sqrt{\#}$ of pixels. Fig. 4 shows clearly distinguishable Gaussian peaks for a displacement down to 3 Å with precision of 0.6 Å, whereas, for less than 3 Å displacements, the Gaussian peaks overlap significantly with our current experimental setup.

In conclusion, we demonstrated a simple system based on transverse Kerker scattering, consisting of a tightly focused vector beam and a small spherical dielectric nanoparticle excited in free-space. Furthermore, we discussed the formalism to optimize such scheme for a more sophisticated practical experimental case, which can be applied for arbitrary excitation beams and particle sizes. Our experimental results show that, upon optimization, a displacement of individual nanoparticle down to few Ångström can be resolved with sub-Ångström accuracy. The discussed scheme proves that the localization of nanoparticles can be sensed with ultra-high precision and accuracy, paving the way towards interesting applications, such as, stabilization of positioning systems in microscopy and nanometrology. Moreover, a quadrant-detector based signal detection would allow for an ultra-fast time-resolved tracking of nanoscopic systems.

We thank Thomas Bauer for the fruitful discussions.
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